A Poisson integrator for Gaussian wavepacket dynamics

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Dedicated to Peter Deuflhard on the occasion of his sixtieth birthday.

Abstract

We consider the variational approximation of the time-dependent Schrödinger equation by Gaussian wavepackets. The corresponding finite-dimensional dynamical system inherits a Poisson (or non-canonically symplectic) structure from the Schrödinger equation by its construction via the Dirac-Frenkel-McLachlan variational principle. The variational splitting between kinetic and potential energy turns out to yield an explicit, easily implemented numerical scheme. This method is a time-reversible Poisson integrator, which also preserves the L^2 norm and linear and angular momentum. Using backward error analysis, we show long-time energy conservation for this splitting scheme. In the semi-classical limit the numerical approximations to position and momentum converge to those obtained by applying the Störmer-Verlet method to the classical limit system.

1 Introduction

Gaussian wavepacket dynamics is widely used in quantum molecular dynamics as an approximation to the time-dependent Schrödinger equation, which we write as

$$i\varepsilon \frac{\partial \psi}{\partial t} = H\psi, \tag{1.1}$$

where $\psi = \psi(x,t)$ is the wave function depending on the spatial variables $x = (x_1, \dots, x_N)$ with $x_k \in \mathbb{R}^d$ (e.g., with $d = 1$ or 3 in the partition) and the time $t \in \mathbb{R}$. Here, ε is a (small) positive number representing the scaled Planck constant and i is the complex imaginary unit. The Hamiltonian operator H is written

$$H = T + V$$

with the kinetic and potential energy operators

$$T = -\sum_{k=1}^{N} \frac{\varepsilon^2}{2m_k} \Delta_{x_k} \quad \text{and} \quad V = V(x),$$

where $m_k > 0$ is a particle mass and Δ_{x_k} the Laplacian in the variable $x_k \in \mathbb{R}^d$, and where the real-valued potential V acts as a multiplication operator on ψ.

In Gaussian wavepacket dynamics [10, 11, 12, 13, 19, 3, 2, 20] an approximation to the wave function $\psi(x,t)$ is sought for in the form

$$u(x,t) = e^{i\phi(t)/\varepsilon} \prod_{k=1}^{N} \varphi_k(x_k,t) \tag{1.2}$$

with

$$\varphi_k(x_k,t) = \exp\Big(\frac{i}{\varepsilon}\big(a_k(t)\,|x_k - q_k(t)|^2 + p_k(t)\cdot(x_k - q_k(t)) + c_k(t)\big)\Big), \tag{1.3}$$

where $|\cdot|$ and $\cdot$ denote the Euclidean norm and inner product on $\mathbb{R}^d$, respectively. Here, $a_k = \alpha_k + i\beta_k$ (with $\beta_k > 0$) is a complex width parameter, $c_k = \gamma_k + i\delta_k$ is a complex phase parameter, and ϕ is a real phase. (Only the sum of the phases $\phi + \sum_k c_k$ is determined uniquely. The partition of the phases is made by convenience so that each φ_k is of constant norm.) The parameters $q_k \in \mathbb{R}^d$ and $p_k \in \mathbb{R}^d$ represent the position and momentum average, respectively: $q_k = \langle x_k \rangle$ and $p_k = \langle -i\varepsilon\nabla_{x_k}\rangle$. Here and in the following we denote the average of observables along the approximate wave function u by

$$\langle A \rangle = \langle\, u \,|\, A \,|\, u \,\rangle / \|\, u \,\|^2$$

where $\|\, u\,\|^2 = \langle\, u \,|\, 1 \,|\, u\,\rangle$ is the squared L^2 norm. For real-valued functions A,

$$\langle\, u \,|\, A \,|\, u\,\rangle = \int_{\mathbb{R}^{Nd}} A(x) \prod_{j=1}^{N} \exp\Big(-\frac{2}{\varepsilon}\big(\beta_j|x_j - q_j|^2 + \delta_j\big)\Big)\mathrm{d}x. \tag{1.4}$$

The average $\langle A\rangle$ of a real-valued function A at u thus depends only on the parameters q_j, β_j, and δ_j.

The Dirac-Frenkel-McLachlan variational principle [5, 6, 17] yields equations of motion for these parameters as derived by Heller [11]:

$$\begin{aligned} \dot{q}_k &= \frac{p_k}{m_k} \\ \dot{p}_k &= -\langle \nabla_{x_k} V\rangle \\ \dot{a}_k &= -\frac{2a_k^2}{m_k} - \frac{1}{2d}\langle \Delta_{x_k} V\rangle \\ \dot{c}_k &= \frac{i\varepsilon d a_k}{m_k} + \frac{\varepsilon}{8\beta_k}\langle \Delta_{x_k} V\rangle \end{aligned} \tag{1.5}$$

and

$$\dot{\phi} = \sum_{k=1}^{N} \frac{|p_k|^2}{2m_k} - \langle V\rangle.$$

We show in Section 2 that the system (1.5) has a non-degenerate Poisson structure, or in other terminology, it is a non-canonical Hamiltonian system. In particular, it preserves the total energy

$$\langle H\rangle = \sum_{k=1}^{N}\left(\frac{|p_k|^2}{2m_k} + \frac{\varepsilon d}{2m_k}\,\frac{\alpha_k^2 + \beta_k^2}{\beta_k}\right) + \langle V\rangle. \tag{1.6}$$

The L^2 norm of the approximation, $\|u\| = \sqrt{\langle\, u \,|\, 1 \,|\, u\,\rangle}$, is also preserved.

Our interest in this paper is the structure-preserving time integration of the Gaussian wavepacket equations (1.5). We propose and study a method that is based on the splitting of (1.5) into the kinetic and potential energy parts, similarly as is done in classical molecular dynamics in the Störmer-Verlet method [9, 18]. Since it turns out that the kinetic and potential energy subsystems can be solved explicitly, this splitting yields a simple time-stepping algorithm for computing approximations $q_k^n, p_k^n, a_k^n = \alpha_k^n + i\beta_k^n, c_k^n = \gamma_k^n + i\delta_k^n$, and ϕ^n to the corresponding parameters at time $t^n = n\Delta t$. We denote the so obtained approximation to $u(t^n)$ of (1.2) by u^n, and averages at u^n by

$$\langle A \rangle^n = \langle\, u^n \,|\, A \,|\, u^n \,\rangle / \|\, u^n \,\|^2.$$

Algorithm. A step from time t^n to t^{n+1} proceeds as follows:

1. With the averages determined from (1.4), compute

$$\begin{aligned} p_k^{n+1/2} &= p_k^n - \frac{\Delta t}{2}\, \langle \nabla_{x_k} V \rangle^n \\ \alpha_k^{n,+} &= \alpha_k^n - \frac{\Delta t}{4d}\, \langle \Delta_{x_k} V \rangle^n \\ \gamma_k^{n,+} &= \gamma_k^n + \frac{\Delta t\,\varepsilon}{16\beta_k^n}\, \langle \Delta_{x_k} V \rangle^n. \end{aligned} \tag{1.7}$$

2. From the values $p_k^{n+1/2}$, $a_k^{n,+} = \alpha_k^{n,+} + i\beta_k^n$ and $c_k^{n,+} = \gamma_k^{n,+} + i\delta_k^n$ compute q_k^{n+1}, $a_k^{n+1,-} = \alpha_k^{n+1,-} + i\beta_k^{n+1}$, and $c_k^{n+1,-} = \gamma_k^{n+1,-} + i\delta_k^{n+1}$ via

$$\begin{aligned} q_k^{n+1} &= q_k^n + \frac{\Delta t}{m_k}\, p_k^{n+1/2} \\ a_k^{n+1,-} &= a_k^{n,+} \Big/ \Big(1 + 2\frac{\Delta t}{m_k} a_k^{n,+}\Big) \\ c_k^{n+1,-} &= c_k^{n,+} + \frac{i\varepsilon d}{2} \log\Big(1 + 2\frac{\Delta t}{m_k} a_k^{n,+}\Big). \end{aligned} \tag{1.8}$$

3. Compute $p_k^{n+1}, \alpha_k^{n+1}, \gamma_k^{n+1}$ from

$$\begin{aligned} p_k^{n+1} &= p_k^{n+1/2} - \frac{\Delta t}{2}\, \langle \nabla_{x_k} V \rangle^{n+1} \\ \alpha_k^{n+1} &= \alpha_k^{n+1,-} - \frac{\Delta t}{4d}\, \langle \Delta_{x_k} V \rangle^{n+1} \\ \gamma_k^{n+1} &= \gamma_k^{n+1,-} + \frac{\Delta t\,\varepsilon}{16\beta_k^{n+1}}\, \langle \Delta_{x_k} V \rangle^{n+1}. \end{aligned} \tag{1.9}$$

Finally, the phase is updated as

$$\phi^{n+1} = \phi^n + \Delta t \sum_{k=1}^{N} \frac{|p_k^{n+1/2}|^2}{2m_k} - \frac{\Delta t}{2}\Big(\langle V \rangle^n + \langle V \rangle^{n+1}\Big). \tag{1.10}$$

Note that in Step 3. the averages $\langle A \rangle^{n+1}$ depend, in view of (1.4), only on the parameters $q_k^{n+1}, \beta_k^{n+1}, \delta_k^{n+1}$ which are computed already in Step 2. These averages can be reused in the subsequent time step.

We show in Section 3 that this method is a second-order, time-reversible Poisson integrator for (1.5), which preserves the total energy $\langle H \rangle^n$ up to $O(\Delta t^2)$ over time intervals that are exponentially long in Δt, i.e., of size $O(e^{c/\Delta t})$ with a constant $c > 0$, uniformly in ε. The norms $\|u^n\|$ are conserved exactly along the numerical solutions. If the potential has a rotational symmetry so that the angular momentum is conserved in the full quantum dynamics, then also the numerical integrator preserves the angular momentum.

In the classical limit $\varepsilon \to 0$, the position and momentum approximations q_k^n, p_k^n converge to the position and momentum aproximations obtained by applying the Störmer-Verlet method to the classical limit system $\dot{q}_k = p_k/m_k$, $\dot{p}_k = -\nabla_{q_k} V(q)$.

In Section 5 we present the results of numerical experiments with the Hénon-Heiles and Kepler Hamiltonians. Extensions of the algorithm to higher-order approximations, to non-spherical Gaussian wavepackets, and to splittings other than between the kinetic and potential energy are briefly addressed in Section 6.

2 The Poisson structure of variational Gaussian wavepacket dynamics

In this section we discuss the non-canonical symplectic structure that the Gaussian wavepacket equations (1.5) inherit from the Schrödinger equation by the variational approximation.

2.1 Poisson structure of the Schrödinger equation

We consider now the Schödinger equation (1.1). Let us write $\psi = v + iw$ the real and imaginary parts of $\psi \in L^2(\mathbb{R}^D, \mathbb{C})$, where $D = N \times d$. The functions v and w are thus functions in the *real* Hilbert space $L^2(\mathbb{R}^D, \mathbb{R})$. We denote the complex inner product by $\langle \cdot, \cdot \rangle$ and the real inner product by $(\cdot, \cdot)$.

As H is a real operator, Eq. (1.1) can be written

$$\begin{aligned} \varepsilon \dot{v} &= Hw, \\ \varepsilon \dot{w} &= -Hv, \end{aligned} \tag{2.1}$$

or equivalently, with the canonical structure matrix

$$J = \begin{pmatrix} 0 & -1 \\ 1 & 0 \end{pmatrix}$$

and the Hamiltonian

$$H(v, w) = \tfrac{1}{2}\langle \psi, H\psi \rangle = \tfrac{1}{2}(v, Hv) + \tfrac{1}{2}(w, Hw)$$

for $\psi = v + iw$ (we use the same symbol H as for the operator), this becomes the canonical Hamiltonian system

$$\begin{pmatrix} \dot{v} \\ \dot{w} \end{pmatrix} = \varepsilon^{-1} J^{-1} \nabla H(v, w).$$

Note that the real multiplication with J corresponds to the complex multiplication with the imaginary unit i.

The flow of this system preserves the canonical symplectic 2-form

$$\omega(\xi, \eta) = (\xi, J\eta), \qquad \xi, \eta \in L^2(\mathbb{R}^D, \mathbb{R})^2. \tag{2.2}$$

The associated Poisson bracket is

$$\{F, G\}_{\mathrm{can}} = (\nabla F, J^{-1} \nabla G) \tag{2.3}$$

for differentiable functions $F, G : L^2(\mathbb{R}^D, \mathbb{R})^2 \to \mathbb{R}$.

2.2 Poisson structure of variational approximations

Consider a (finite-dimensional, smooth) submanifold $\mathcal{M}$ of the complex Hilbert space $\mathcal{H}_{\mathbb{C}} = L^2(\mathbb{R}^D, \mathbb{C})$ with the property that at every $u \in \mathcal{M}$, the tangent space $T_u\mathcal{M}$ is a *complex* linear space, i.e., with $\delta u \in T_u\mathcal{M}$ also $i\delta u \in T_u\mathcal{M}$.

The Dirac-Frenkel-McLachlan variational principle [5, 6, 17] (see also [15] and references therein) determines an approximation $u(t) \in \mathcal{M}$ to the Schrödinger equation (1.1) by requiring that

$$\langle\, \delta u,\, i\varepsilon\dot{u} - Hu \,\rangle = 0 \quad \text{ for all } \quad \delta u \in T_u\mathcal{M}. \tag{2.4}$$

Since $T_u\mathcal{M}$ is assumed complex linear, this condition is equivalent to requiring the analogous condition only for the real part or alternatively only for the imaginary part.

To understand the Poisson structure of this system, we return to the real formulation, where taking the real part of (2.4) becomes, upon identifying $u = v + iw$ with the real pair $u = (v, w)^T$,

$$\big(\,\delta u,\, \varepsilon J\dot{u} - \nabla_u H(u)\big) = 0 \quad \text{ for all } \quad \delta u \in T_u\mathcal{M}.$$

We choose a (local) parametrization $u = \chi(y)$ of $\mathcal{M}$ for y in an open subset $\Omega \subset \mathbb{R}^m$ and denote its derivative $X_{\mathbb{C}}(y) = V(y) + iW(y) = \chi'(y)$ or in the real setting as $X = \begin{pmatrix} V \\ W \end{pmatrix}$. Denoting by X^T the adjoint of X with respect to the real inner product $(\cdot,\cdot)$, we thus obtain

$$\varepsilon X(y)^T J X(y)\, \dot{y} = X(y)^T \nabla_u H(\chi(y)).$$

With $X_{\mathbb{C}}^*$ denoting the adjoint of $X_{\mathbb{C}}$ with respect to the complex inner product $\langle\cdot,\cdot\rangle$, we note $X_{\mathbb{C}}^* X_{\mathbb{C}} = (V^TV + W^TW) + i(V^TW - W^TV) = X^TX - iX^TJX$ and hence

$$X^TJX = -\mathrm{Im}\, X_{\mathbb{C}}^* X_{\mathbb{C}}. \tag{2.5}$$

This skew-symmetric matrix is invertible, as the following lemma shows.

Lemma 2.1 *If $T_u\mathcal{M}$ is a complex linear space for every $u \in \mathcal{M}$, then $\mathcal{M}$ is a symplectic submanifold of $L^2(\mathbb{R}^D, \mathbb{R})^2$, that is, the symplectic two-form (2.2) is non-degenerate on $T_u\mathcal{M}$ for all $u \in \mathcal{M}$. Expressed in coordinates,*

$$X(y)^T J X(y) \quad \textit{is invertible for all } y.$$

Proof. We fix $u = \chi(y) \in \mathcal{M}$ and omit the argument $y \in \Omega$ in the following. Since $T_u\mathcal{M} = \mathrm{Range}(X_{\mathbb{C}})$ is complex linear by assumption, there exists a real linear mapping $L : \mathbb{R}^m \to \mathbb{R}^m$ such that $iX_{\mathbb{C}}\eta = X_{\mathbb{C}}L\eta$ for all $\eta \in \mathbb{R}^m$. This implies

$$JX = XL \quad \text{ and } \quad L^2 = -\mathrm{Id}$$

and hence $X^TJX = X^TXL$, which is invertible, since X is of full rank. ∎

We denote the inverse, which is again skew-symmetric, by

$$B(y) = \varepsilon^{-1}\big(X(y)^T J X(y)\big)^{-1}. \tag{2.6}$$

Introducing the Hamiltonian function on the manifold $\mathcal{M}$ in the coordinates y as

$$K(y) = H(\chi(y)),$$

we note $X(y)^T \nabla_u H(\chi(y)) = \nabla_y K(y)$. Hence the differential equation for y becomes

$$\dot{y} = B(y) \nabla_y K(y). \tag{2.7}$$

We then have the following result; for related material on Poisson systems see also [8, Ch. VII.2] and [16, Ch. 8.5].

Theorem 2.2 *The system* (2.7) *is a non-degenerate Poisson system, i.e., the structure matrix $B(y)$ is invertible and generates a bracket*

$$\{F, G\}(y) = \nabla F(y)^T B(y) \nabla G(y) \tag{2.8}$$

on smooth functions $F, G : \Omega \to \mathbb{R}$, which is skew-symmetric ($\{G, F\} = -\{F, G\}$) *and satisfies the Jacobi identity* ($\{E, \{F, G\}\} + \{F, \{G, E\}\} + \{G, \{E, F\}\} = 0$) *and the Leibniz rule* ($\{E \cdot F, G\} = E \cdot \{F, G\} + F \cdot \{E, G\}$).

Proof. Since ε plays no role here, we let $B(y) = \big(X(y)^T J X(y)\big)^{-1}$ (without the factor ε^{-1}) in this proof. For points $u \in \mathcal{M}$ we introduce the symplectic projector $P(u)$ from the Hilbert space $\mathcal{H} = L^2(\mathbb{R}^D, \mathbb{R})^2$ onto the tangent space $T_u\mathcal{M}$ as

$$P(u) = X(y)B(y)X(y)^T J, \qquad u = \chi(y) \in \mathcal{M},$$

and the complementary projector $Q(u) = I - P(u)$. From the induced decomposition $\mathcal{H} = P(u)\mathcal{H} \oplus Q(u)\mathcal{H}$ we obtain, by the implicit function theorem, a corresponding splitting in a neighbourhood of the manifold $\mathcal{M}$ in $\mathcal{H}$,

$$\psi = u + v \quad \text{ with } u \in \mathcal{M},\ P(u)v = 0.$$

This permits us to extend functions F to a neighbourhood of $\mathcal{M}$ by setting

$$\widehat{F}(\psi) = F(y) \quad \text{ for } \ \psi = u + v \ \text{ with } u = \chi(y),\ P(u)v = 0.$$

We then have for the derivative $D\widehat{F}(u) = D\widehat{F}(u)P(u)$ and hence for its adjoint, the gradient, $\nabla\widehat{F}(u) = P(u)^T \nabla\widehat{F}(u)$. Moreover, $\nabla F(y) = X(y)^T \nabla\widehat{F}(u)$ for $u = \chi(y)$. For the canonical bracket this gives at $u = \chi(y)$

$$\{\widehat{F}, \widehat{G}\}_{\mathrm{can}}(u) = \nabla\widehat{F}(u)^T P(u) J^{-1} P(u)^T \nabla\widehat{G}(u) = \nabla F(y)^T B(y) \nabla G(y) = \{F, G\}(y),$$

and hence the stated properties follow from the corresponding properties of the canonical bracket. ■

As a consequence, the flow $\varphi_t : y(0) \mapsto y(t)$ is a Poisson map, i.e., its derivative φ_t' with respect to the initial value satisfies (see [8, Ch. VII.2])

$$\varphi_t'(y) B(y) \varphi_t'(y)^T = B(\varphi_t(y)) \qquad \text{for all } \ y \in \Omega. \tag{2.9}$$

On taking the inverse on both sides, this is seen to be equivalent to

$$\varphi_t'(y)^T X(\varphi_t(y))^T J X(\varphi_t(y)) \varphi_t'(y) = X(y)^T J X(y),$$

which says that the symplectic two-form (2.2) restricted to the tangent space is conserved along the flow of the variational equations of motion (2.4).

2.3 Poisson structure of Gaussian wavepacket dynamics

Heller's variational Gaussian wavepacket dynamics is obtained by choosing the manifold $\mathcal{M}$ as consisting of complex Gaussians. For the one-particle case $N = 1$, to which we restrict our attention in the following for ease of presentation (the extension to $N > 1$ being straightforward), we have

$$\mathcal{M} = \{u = \chi(y) \in L^2(\mathbb{R}^d) : y = (p, q, \alpha, \beta, \gamma, \delta) \in \mathbb{R}^{2d+4} \text{ with } \beta > 0\} \tag{2.10}$$

with[1]

$$\Big(\chi(y)\Big)(x) = \exp\Big(\frac{i}{\varepsilon}\big((\alpha + i\beta)\,|x - q|^2 + p \cdot (x - q) + \gamma + i\delta\big)\Big). \tag{2.11}$$

The tangent space $T_u\mathcal{M} \subset L^2(\mathbb{R}^d)$ at a given point $u = \chi(y) \in \mathcal{M}$ is $(2d + 4)$-dimensional and is made of the elements of $L^2(\mathbb{R}^d)$ written as

$$\frac{i}{\varepsilon}\Big((A + iB)\,|x - q|^2 + (P - 2(\alpha + i\beta)Q) \cdot (x - q) - p \cdot Q + C + iD\Big)u \tag{2.12}$$

with arbitrary $(P, Q, A, B, C, D)^T \in \mathbb{R}^{2d+4}$. It is worth noticing that the tangent space $T_u\mathcal{M}$ is invariant under multiplication by i (note $\beta > 0$). Moreover, we have $u \in T_u\mathcal{M}$. By choosing $\delta u = u$ in (2.4) and taking the imaginary part, this yields $(d/dt)\|u\|^2 = 2\,\mathrm{Re}\,\langle u, \dot{u}\rangle = 0$ and hence the preservation of the squared L^2 norm of $u = \chi(y)$, which is given by (see (1.4))

$$I(y) := \|\chi(y)\|^2 = \exp\left(-\frac{2\delta}{\varepsilon}\right)\left(\frac{\pi\varepsilon}{2\beta}\right)^{d/2}. \tag{2.13}$$

With these preparations we have the following result.

Theorem 2.3 *The variational approximation on the Gaussian wavepacket manifold $\mathcal{M}$ of (2.10)-(2.11) yields the Poisson system*

$$\dot{y} = B(y)\nabla_y K(y) \tag{2.14}$$

where, for $y = (p, q, \alpha, \beta, \gamma, \delta) \in \mathbb{R}^{2d+4}$ with $\beta > 0$, and with 1_d denoting the d-dimensional identity,

$$B(y) = \frac{1}{I(y)}\begin{pmatrix} 0 & -1_d & 0 & 0 & -p & 0 \\ 1_d & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & \frac{4\beta^2}{\varepsilon d} & 0 & -\beta \\ 0 & 0 & -\frac{4\beta^2}{\varepsilon d} & 0 & \beta & 0 \\ p^T & 0 & 0 & -\beta & 0 & \frac{d+2}{4}\varepsilon \\ 0 & 0 & \beta & 0 & -\frac{d+2}{4}\varepsilon & 0 \end{pmatrix} \tag{2.15}$$

defines a Poisson structure, and for $u = \chi(y)$,

$$K(y) = \langle\, u \,|\, H \,|\, u \,\rangle = K_T(y) + K_V(y) \tag{2.16}$$

is the total energy, with kinetic and potential parts

$$K_T(y) = I(y)\left(\frac{|p|^2}{2m} + \frac{\varepsilon d}{2m}\,\frac{\alpha^2 + \beta^2}{\beta}\right) = \langle\, u \,|\, T \,|\, u \,\rangle$$

and

$$K_V(y) = \int_{\mathbb{R}^d} V(x) \exp\Big(-\frac{2}{\varepsilon}\big(\beta|x - q|^2 + \delta\big)\Big)\,\mathrm{d}x = \langle\, u \,|\, V \,|\, u \,\rangle.$$

Both $K(y)$ and $I(y)$ are conserved quantities of the system.

[1] In contrast to (1.2), we here include the real phase ϕ in γ.

Proof. In view of Eq. (2.12), the matrix $X_{\mathbb{C}}(y) = \chi'(y) : T_u\mathcal{M} \to L^2$ is written

$$X_{\mathbb{C}}(y) = \frac{i}{\varepsilon}\left(x-q\,,\, -2a(x-q)-p\,,\, |x-q|^2\,,\, i|x-q|^2\,,\, 1\,,\, i\right) u$$

where $a = \alpha + i\beta$ and $u = \chi(y)$ in the complex setting. Using the exact calculcus of Gaussian integrals, we compute directly (see (2.5)) that

$$\varepsilon X^T(y)JX(y) = \frac{1}{2}I(y)\begin{pmatrix} 0 & 1_d & 0 & 0 & 0 & 0 \\ -1_d & 0 & 0 & \frac{dp}{2\beta} & 0 & \frac{2p}{\varepsilon} \\ 0 & 0 & 0 & -\frac{\varepsilon d(d+2)}{8\beta^2} & 0 & -\frac{d}{2\beta} \\ 0 & -\frac{dp^T}{2\beta} & \frac{\varepsilon d(d+2)}{8\beta^2} & 0 & \frac{d}{2\beta} & 0 \\ 0 & 0 & 0 & -\frac{d}{2\beta} & 0 & -\frac{2}{\varepsilon} \\ 0 & -\frac{2p^T}{\varepsilon} & \frac{d}{2\beta} & 0 & \frac{2}{\varepsilon} & 0 \end{pmatrix},$$

and this yields the result (up to the change $B \mapsto \frac{1}{2}B$ and $K \mapsto 2K$). ■

In the following it will be useful to use scaled variables

$$\widehat{y} = (p, q, \alpha, \widehat{\beta}, \gamma, \widehat{\delta}) \in \mathbb{R}^{2d+4} \qquad \text{with} \quad \widehat{\beta} = \frac{\beta}{\varepsilon},\ \widehat{\delta} = \frac{\delta}{\varepsilon}, \tag{2.17}$$

for which the structure matrix becomes independent of ε. Here we have

$$\dot{\widehat{y}} = \widehat{B}(\widehat{y})\nabla_{\widehat{y}}\widehat{K}(\widehat{y}) \tag{2.18}$$

with

$$\widehat{B}(\widehat{y}) = \frac{1}{\widehat{I}(\widehat{y})}\begin{pmatrix} 0 & -1_d & 0 & 0 & -p & 0 \\ 1_d & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & \frac{4\widehat{\beta}^2}{d} & 0 & -\widehat{\beta} \\ 0 & 0 & -\frac{4\widehat{\beta}^2}{d} & 0 & \widehat{\beta} & 0 \\ p^T & 0 & 0 & -\widehat{\beta} & 0 & \frac{d+2}{4} \\ 0 & 0 & \widehat{\beta} & 0 & -\frac{d+2}{4} & 0 \end{pmatrix} \tag{2.19}$$

and

$$\widehat{I}(\widehat{y}) = e^{-2\widehat{\delta}}\left(\frac{\pi}{2\widehat{\beta}}\right)^{d/2} \tag{2.20}$$

and

$$\widehat{K}(\widehat{y}) = \widehat{I}(\widehat{y})\left(\frac{|p|^2}{2m} + \frac{d}{2m}\,\frac{\alpha^2+\varepsilon^2\widehat{\beta}^2}{\widehat{\beta}}\right) + \int_{\mathbb{R}^d} V(x)\, e^{-2\widehat{\beta}|x-q|^2-2\widehat{\delta}}\,\mathrm{d}x. \tag{2.21}$$

3 The numerical integrator

In this section we derive the numerical integrator of the Algorithm of Section 1 and study its properties.

3.1 Variational splitting

We consider the splitting induced by the natural decomposition $H = T + V$ in the variational approximation to the Schrödinger equation on a manifold $\mathcal{M}$. Let

$u^n = \chi(y^n)$ be on the approximation manifold $\mathcal{M}$ and $\Delta t > 0$ the time step size. We define the approximation u^{n+1} to $u(t^{n+1})$ by variational splitting as given in [14].

1. We define $u^{n,+}$ in $\mathcal{M}$ as the solution at time $\frac{1}{2}\Delta t$ of the equation for u,
$$\langle\, \delta u,\, i\varepsilon\dot{u} - Vu \,\rangle = 0 \quad \text{for all} \ \ \delta u \in T_u\mathcal{M} \tag{3.1}$$
with initial value $u(0) = u^n$.
2. We define $u^{n+1,-}$ as the solution at time Δt of
$$\langle\, \delta u,\, i\varepsilon\dot{u} - Tu \,\rangle = 0 \quad \text{for all} \ \ \delta u \in T_u\mathcal{M} \tag{3.2}$$
with initial value $u(0) = u^{n,+}$.
3. Then u^{n+1} is the solution at time $\frac{1}{2}\Delta t$ of (3.1) with initial value $u(0) = u^{n+1,-}$.

Notice that u^{n+1} does not depend on the choice of coordinates on $\mathcal{M}$. Let now $\mathcal{M}$ be parametrized by $u = \chi(y)$ with coordinates y in a domain Ω. To any $u = \chi(y) \in \mathcal{M}$ there corresponds thus a (locally) unique point $y \in \Omega$. The scheme $u^n \mapsto u^{n+1}$ can therefore be seen as a scheme $y^n \mapsto y^{n+1}$, which defines a one-step map
$$y^{n+1} = \Phi_{\Delta t}(y^n) \qquad \text{with} \quad \Phi_{\Delta t} : \Omega \to \Omega.$$
The substeps in the definition of this splitting method are the exact flows $\varphi^V_{\Delta t/2}$ and $\varphi^T_{\Delta t}$ of the Poisson systems
$$\dot{y} = B(y)\nabla_y K_V(y) \quad \text{and} \quad \dot{y} = B(y)\nabla_y K_T(y),$$
where for $u = \chi(y)$, we have $K_V(y) = \langle u \,|\, V \,|\, u\rangle$ and $K_T(y) = \langle u \,|\, T \,|\, u\rangle$, respectively. Therefore, the composed map $\Phi_{\Delta t} = \varphi^V_{\Delta t/2} \circ \varphi^T_{\Delta t} \circ \varphi^V_{\Delta t/2}$ is a Poisson map, i.e., its derivative $\Phi'_{\Delta t}$ with respect to y satisfies
$$\Phi'_{\Delta t}(y)B(y)\Phi'_{\Delta t}(y)^T = B(\Phi_{\Delta t}(y)) \qquad \text{for all} \ \ y \in \Omega, \tag{3.3}$$
since the exact flows $\varphi^V_{\Delta t/2}$ and $\varphi^T_{\Delta t}$ are known to be Poisson maps from (2.9).

3.2 Variational splitting for Gaussian wavepacket dynamics

Most remarkably, the one-step map $\Phi_{\Delta t}$ of variational splitting for Gaussian wavepacket dynamics can be computed explicitly.

Let us consider first the equations (3.2). They are written, for $a = \alpha + i\beta$ and $c = \gamma + i\delta$, as
$$\left\{ \begin{array}{rcl} \dot{q} &=& p/m, \\ \dot{p} &=& 0, \end{array} \right. \qquad \left\{ \begin{array}{rcl} \dot{a} &=& -2a^2/m, \\ \dot{c} &=& \big(\frac{1}{2}|p|^2 + i\varepsilon d a\big)/m, \end{array} \right. \tag{3.4}$$
with initial values $y^0 = (p^0, q^0, a^0, c^0)$ corresponding to $u^0 = \chi(y^0)$. They correspond to the free Gaussian wavepacket equations and can be solved explicitly (see [10]):
$$q(t) = q^0 + \frac{t}{m}\,p^0, \qquad p(t) = p^0, \qquad a(t) = \frac{a^0}{1 + 2a^0 t/m},$$

and

$$c(t) = c^0 + t\,\frac{|p^0|^2}{2m} + \frac{i\varepsilon d}{2}\log\left(1 + \frac{2a^0 t}{m}\right).$$

Let us consider now the equations (3.1). Taking into account the fact that the potential V is real, these equations are written

$$\begin{aligned} \dot p &= -\langle \nabla_x V\rangle, & \dot q &= 0, \\ \dot\alpha &= -\tfrac{1}{2d}\langle \Delta_x V\rangle, & \dot\beta &= 0, \\ \dot\gamma &= -\langle V\rangle + \tfrac{\varepsilon}{8\beta}\langle \Delta_x V\rangle, & \dot\delta &= 0. \end{aligned} \tag{3.5}$$

As the averages in the equations for p, α, γ depend, by (1.4), only on q, β, δ which are constant along this trajectory, these equations can be solved trivially, requiring only the computation of the averages at the initial value. We thus see that the splitting scheme $\Phi_{\Delta t} = \varphi^V_{\Delta t/2}\circ\varphi^T_{\Delta t}\circ\varphi^V_{\Delta t/2}$ can be computed explicitly. This gives the algorithm of Section 1 for the case $N = 1$ after decomposing the real phase $\gamma \to \phi + \gamma$, and the extension to the general case is now obvious.

Since the substeps in the algorithm preserve the L^2 norm $\|u\|^2 = I(y)$, so does the composed method. We summarize these properties in a theorem.

Theorem 3.1 *The splitting scheme described above is an explicit, time-reversible, second-order integrator that preserves the L^2 norm. Its one-step map $\Phi_{\Delta t}$ is a Poisson map (3.3) for the structure matrix (2.15).*

In view of the small parameter ε, the statement on the order of the method requires additional attention. Here it is useful to consider the integrator in the scaled variables $\widehat{y} = (p, q, \alpha, \beta/\varepsilon, \gamma, \delta/\varepsilon)$ of (2.17). Since the differential equation (2.18) contains ε only as a regular perturbation parameter, after n steps of the splitting integrator we have the ε-uniform error bound

$$\widehat{y}^n - \widehat{y}(t^n) = O(\Delta t^2),$$

where the constants symbolized by the O-notation are independent of ε and of n and Δt with $n\Delta t \le$ Const. This yields, in particular, for the approximation of the absolute values of the Gaussian wavepackets,

$$\big\| |u^n|^2 - |u(t^n)|^2 \big\| = O(\Delta t^2), \tag{3.6}$$

but the approximation of the phases is only such that

$$\|u^n - u(t^n)\| = O(\Delta t^2/\varepsilon). \tag{3.7}$$

3.3 Long-time near-conservation of energy

With the Poisson structure of the problem, we can prove the long-time energy conservation of the numerical integrator, even uniformly in ε.

Theorem 3.2 *Let the potential V be real-analytic and bounded (by M, say) on a complex domain $D \subset \mathbb{C}^d$, and locally integrable on $\mathbb{R}^d$ with at most exponential growth at infinity. Suppose that the numerical trajectory $y^{n+1} = \Phi_{\Delta t}(y^n)$ for $n = 0, 1, 2, \ldots, n^*$ remains in a subset of $\mathbb{R}^{2d+4}$ in which the total energy $\langle H\rangle$ at $u = \chi(y)$ is bounded (by E, say), in which the position variables q remain in* *a closed subset Q of the domain D, and in which $\beta \geq c_0\varepsilon$ with $c_0 > 0$. Then the energy error of $u^n = \chi(y^n)$ is bounded by*

$$\Big| \langle H \rangle^n - \langle H \rangle^0 \Big| \leq C\Delta t^2 \qquad \textit{for} \;\; n\Delta t \leq \exp(c/\Delta t) \tag{3.8}$$

and $n \leq n^*$, where c and C are positive constants that are independent of n, Δt, and ε. (They depend only on E, on M and $r = \mathrm{dist}(Q, \partial D)$, and on c_0.)

Note the condition $\beta^n \geq c_0\varepsilon$ on the width parameter along the numerical solution. If the Gaussian wavepacket becomes too wide (β too small), then energy conservation is no longer guaranteed; see the numerical example on the Kepler problem in Section 5. On the other hand, very wide Gaussians cannot be expected to carry physical significance.

Proof. To obtain the uniformity in ε, it is appropriate to work with the scaled variables $\widehat{y}$ of (2.17), so that the scheme is considered as $\widehat{y}^{n+1} = \widehat{\Phi}_{\Delta t}(\widehat{y}^n)$. Since $\widehat{\Phi}_{\Delta t}$ is a Poisson map with respect to the (ε-independent) structure matrix $\widehat{B}(\widehat{y})$ of (2.19), and since the method is symmetric, there exists a modified Hamiltonian

$$\widehat{K}_{\Delta t}(\widehat{y}) = \widehat{K}(\widehat{y}) + \Delta t^2 \widehat{K}_3(\widehat{y}) + \Delta t^4 \widehat{K}_5(\widehat{y}) + \cdots + \Delta t^N \widehat{K}_{N+1}(\widehat{y})$$

where the functions $\widehat{K}_j(\widehat{y})$ are analytic functions defined on all of Ω (see Theorems 2.2 and 3.6 of Chapter IX in [8]). This modified Hamiltonian is such that for a suitable truncation index $N \sim \Delta t^{-1}$, the exact flow $\widehat{\varphi}_{\Delta t}$ at time Δt of the modified Poisson system

$$\dot{\widehat{y}} = \widehat{B}(\widehat{y}) \nabla_{\widehat{y}} \widehat{K}_{\Delta t}(\widehat{y})$$

is exponentially close to the one-step map $\widehat{\Phi}_{\Delta t}$,

$$\|\widehat{\varphi}_{\Delta t}(\widehat{y}) - \widehat{\Phi}_{\Delta t}(\widehat{y})\| \leq C_1 e^{-c_1/\Delta t},$$

see Theorem IX.7.6 in [8]. Under the stated conditions, $\widehat{B}(\widehat{y})$ and $\widehat{K}(\widehat{y})$ are analytic and uniformly bounded in an ε-independent domain that contains the numerical solution; see Lemma 3.3 below. The above exponentially small estimate is therefore true along the numerical solution, with constants independent of Δt and ε. As in Theorem IX.8.1 of [8], we then obtain the estimate (3.8) for the Hamiltonian $\widehat{K}(\widehat{y}^n) = \langle\, u^n \,|\, H \,|\, u^n \,\rangle$. ■

The following lemma ensures that the Hamiltonian $\widehat{K}(\widehat{y}) = \langle u|H|u\rangle$ is analytic and bounded uniformly in ε as is required in the preceding proof. For simplicity we state it for the one-dimensional case $d = 1$ and omit the obvious extension to $d > 1$.

Lemma 3.3 *Let the potential V be locally integrable on $\mathbb{R}$, with at most exponential growth at infinity. Assume that V is analytic and bounded by M in a complex disk of radius r centered at $q_0 \in \mathbb{R}$. Then, the function defined by*

$$F(q,b,s) := \int_{\mathbb{R}} V(x)\, e^{-b(x-q)^2 - s}\, \mathrm{d}x$$

is analytic and bounded by $2M$ on the domain (with sufficiently large $c_0 > 0$)

$$\Big\{ (q,b,s) \in \mathbb{C}^3 : |q - q_0| < r/2,\ \mathrm{Re}\, b > c_0,\ |\mathrm{Im}\, b| < 1,\ \big| e^{-s} \sqrt{\pi/\mathrm{Re}\, b} \big| < 3/2 \Big\}.$$

Proof. The analyticity is obvious, and it remains to show the uniform boundedness on the stated domain. Let q be a complex number with $|q - q_0| < r/2$. We deform the integration contour from the real line to a complex step contour which consists of the horizontal line segment through q in the disk of analyticity of V (of radius r around q_0), of two vertical connections to the real line, and which remains on the real line outside the disk. The contribution from the horizontal line segment Γ_q through q is bounded, for $q = \xi + i\eta$, by

$$\Big| \int_{\Gamma_q} V(x)\, e^{-b(x-q)^2} dx \Big| \le M \int_{\mathbb{R}} e^{-\mathrm{Re}\, b\, (x-\xi)^2} dx = M \sqrt{\frac{\pi}{\mathrm{Re}\, b}}.$$

The contributions from the other pieces of the contour are exponentially small in $\mathrm{Re}\, b$. This yields the result. ■

We remark that Lemma 3.3 and Theorem 3.2 extend in an obvious way to the higher-dimensional situation $N \ge 1$, $d \ge 1$ considered in Section 1.

3.4 The Störmer-Verlet method as the classical limit

For classical molecular dynamics, i.e.,

$$\dot q = p/m, \qquad \dot p = -\nabla V(q),$$

the standard numerical integrator is the Störmer-Verlet method (see, e.g., [1, 7, 9]),

$$\begin{aligned} p^{n+1/2} &= p^n - \frac{\Delta t}{2}\, \nabla V(q^n) \\ q^{n+1} &= q^n + \frac{\Delta t}{m}\, p^{n+1/2} \\ p^{n+1} &= p^{n+1/2} - \frac{\Delta t}{2}\, \nabla V(q^{n+1}), \end{aligned}$$

which is also based on a splitting between kinetic and potential energy, now in the classical mechanical setting. Comparing this scheme with the method formulas as given in the Algorithm of Section 1, we obtain the following.

Theorem 3.4 *In the limit $\varepsilon \to 0$, the position and momentum approximations q^n, p^n of the variational splitting integrator for Gaussian wavepacket dynamics tend to those given by the Störmer-Verlet method applied to the associated classical mechanical system.*

4 Conservation of linear and angular momentum

In the case of translational or rotational symmetries in the system, the exact wave function, the Gaussian wavepacket approximation and its time-discrete approximation all conserve linear and angular momentum, respectively.

4.1 Conserved quantities in variational approximations

For general variational approximations (2.4) we notice the following.

Lemma 4.1 *Let the self-adjoint operator A commute with the Hamiltonian H, $[H, A] = 0$. If*

$$Au \in T_u\mathcal{M} \qquad \text{for all } u \in \mathcal{M}, \tag{4.1}$$

then the average of A along variational approximations $u(t) \in \mathcal{M}$ is conserved:

$$\frac{d}{dt}\langle A \rangle = 0.$$

Proof. We have

$$\frac{d}{dt}\langle u \,|\, A \,|\, u \rangle = 2\,\mathrm{Re}\,\langle Au, \dot{u} \rangle = 2\,\mathrm{Re}\,\langle Au, \frac{1}{i\varepsilon}\,Hu \rangle = \langle u \,|\, \frac{1}{i\varepsilon}\,[H, A] \,|\, u \rangle = 0$$

on using (2.4) and (4.1) in the second equality. ■

4.2 Conservation of linear and angular momentum in Gaussian wavepacket dynamics

The quantum-mechanical *total linear momentum* and *total angular momentum*,

$$P = \sum_{k=1}^{N}(-i\varepsilon\nabla_{x_k}) \quad \text{and} \quad L = \sum_{k=1}^{N} x_k \times (-i\varepsilon\nabla_{x_k}), \tag{4.2}$$

both commute with the kinetic energy operator T. (We consider here the case $d = 3$, i.e., $x_k \in \mathbb{R}^3$.) For potentials that are invariant under translations, i.e.,

$$V(x_1, \dots, x_N) = V(x_1 + q, \dots, x_N + q) \quad \text{for all } q \in \mathbb{R}^3,$$

P commutes also with V, and for potentials invariant under rotations, i.e.,

$$V(x_1, \dots, x_N) = V(Qx_1, \dots, Qx_N) \quad \text{for all } Q \in SO(3),$$

L commutes with V, and hence also with the Hamiltonian $H = T + V$. For example, potentials with only distance-dependent pair interactions $V_{jk}\left(|x_j - x_k|\right)$ are invariant under both translations and rotations.

For Gaussian wavepackets (1.2) with (1.3) with $d = 3$ it is verified by direct calculation that condition (4.1) is indeed satisfied by the components of P and L. Hence we have the following.

Lemma 4.2 *For Gaussian wavepacket dynamics in a translation- or rotation-invariant potential, the total linear momentum $\langle P \rangle$ and the total angular momentum $\langle L \rangle$, respectively, are conserved quantities.*

We note that for Gaussian wavepackets,

$$\langle P \rangle = \sum_{k=1}^{N} p_k \quad \text{and} \quad \langle L \rangle = \sum_{k=1}^{N} q_k \times p_k$$

equal the *classical* total linear and angular momentum.

4.3 Conservation of linear and angular momentum by the numerical integrator

Theorem 4.3 *For a translation- or rotation-invariant potential, the Gaussian wavepacket integrator preserves the total linear and angular momentum, respectively: for all n,*

$$\langle P \rangle^n = \langle P \rangle^0 \quad \textit{and} \quad \langle L \rangle^n = \langle L \rangle^0.$$

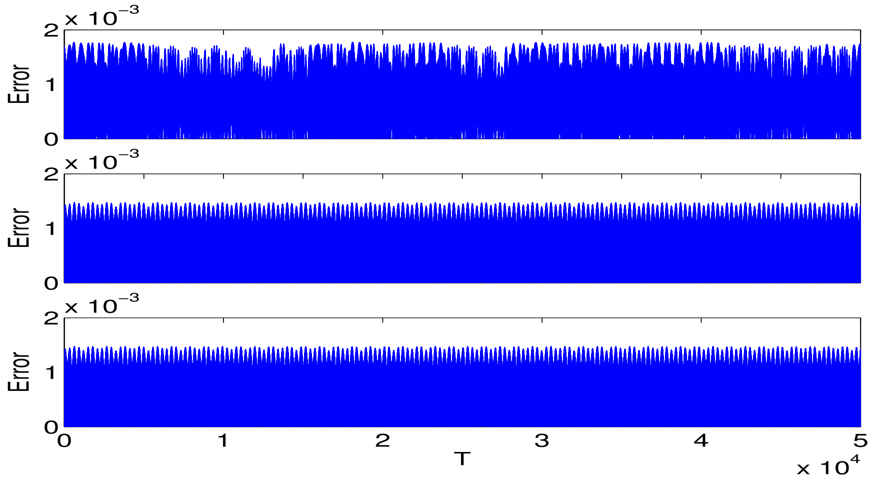

Figure 1: Hénon-Heiles. $\Delta t = 0.1$, $\varepsilon = 10^{-2}$ (top), 10^{-3} (middle) and 10^{-4} (bottom)

Proof. Since P or L commutes with both T and V, the considerations of the previous subsection show that the total linear or angular momentum is conserved exactly in each of the substeps of the variational splitting, and hence also in their composition $\Phi_{\Delta t}$. ∎

5 Numerical experiments

5.1 Generalized Hénon-Heiles Hamiltonian

We consider the potential functions ($N = 1$, $d = 2$)

$$V(x_1, x_2) = \frac{A}{2}x_1^2 + \frac{B}{2}x_2^2 + Dx_1x_2^2 - C\frac{x_1^3}{3}, \tag{5.1}$$

where A, B, C and D are real parameters. In this case, the inner products (1.4) can be computed explicitly: For the average at a Gaussian wavepacket $u = \chi(y)$, we have

$$\langle V \rangle = V(q_1, q_2) + (A + B)\frac{\varepsilon}{8\beta} - q_1(C - D)\frac{\varepsilon}{4\beta},$$

and similarly,

$$\begin{aligned} \langle \partial_{x_1} V \rangle &= Aq_1 + Dq_2^2 - Cq_1^2 - (C - D)\frac{\varepsilon}{4\beta}, \\ \langle \partial_{x_2} V \rangle &= Bq_2 + 2Dq_1q_2 \quad \text{and} \\ \langle \Delta_x V \rangle &= A + B - 2(C - D)q_1. \end{aligned}$$

To see the real effect of the Gaussian wavepacket equations, we need to take $C \neq D$. We made numerical experiments with $A = B = D = 1$ and $C = 2$. We took the initial conditions $p_1^0 = 0$, $p_2^0 = 1$, $q_1^0 = 0$, $q_2^0 = 1$, $\alpha^0 = 1$, $\beta^0 = 1$, $\gamma^0 = 0$, $\phi^0 = 0$, and $\delta^0 = -\frac{1}{2}\varepsilon \log(2\beta^0\pi^{-1}\varepsilon^{-1})$ to ensure $\|u^0\| = 1$.

In Figure 1 we plot the energy error for $\Delta t = 0.1$ and $\varepsilon = 10^{-2}$, 10^{-3} and 10^{-4} over a long time interval $0 \le t \le 5 \cdot 10^4$. In accordance with Theorem 3.2, we see that the energy $\langle H \rangle$ is well conserved independently of ε.

5.2 Three-dimensional Kepler problem

We consider now the case where $N = 1$, $d = 3$ and the potential is $V(q) = -1/|q|$. In this case, we compute the L^2 inner products using Gauss-Hermite integration points, except for the Laplacian of V where we compute directly that

$$\langle \Delta V \rangle = 4\pi \exp\left(-\frac{2}{\varepsilon}(\beta |q|^2 + \delta) \right).$$

We used the initial conditions: $p^0 = (1, 0, 0)^T$, $q^0 = (0, 1, 0)^T$, $\alpha^0 = 1$, $\beta^0 = 1$, $\gamma^0 = 0$, $\phi^0 = 0$, and $\delta^0 = -\frac{3}{4}\varepsilon \log(2\beta^0 \pi^{-1} \varepsilon^{-1})$ to ensure unit norm of the wavepacket.

In Figures 2, 3 and 4 we plot the relative energy error and β for $\varepsilon = 10^{-3}$, 10^{-4} and 10^{-5}. We see that the energy is well conserved until β is of the order of magnitude of ε where a drift appears. This indicates that the condition $\beta \ge c_0 \varepsilon$ in Theorem 3.2 is optimal to ensure the energy conservation.

Finally, we illustrate the convergence of the integrator toward the Störmer-Verlet scheme. In Figure 5, we used the same data as before, and plotted 500 steps with step size $\Delta t = 0.1$ in the (q_1, q_2)-plane of the Störmer-Verlet scheme and of our Gaussian wavepacket integrator for $\varepsilon = 0.1$, 0.01, and 0.001. As ε goes to zero, the numerical solution tends to the solution given by the Störmer-Verlet integrator. For $\varepsilon = 0.001$ the result cannot be distinguished in the figure from that of the Störmer-Verlet scheme.

6 Extensions

The method studied in this paper admits extensions in various directions: to higher order of approximation, to non-spherical Gaussian wavepackets, and to splittings other than between the kinetic and potential energy.

6.1 Higher-order methods

Higher-order methods with the same conservation properties can be constructed in a standard way by composing several steps of our algorithm with different time steps; see, e.g., the composition methods and references in [8, Sect. V.3.2] and [18].

6.2 Non-spherical Gaussian wavepackets

Instead of (1.3) we may look for an approximation (1.2) with

$$\varphi_k(x_k, t) = \exp\Big(\frac{i}{\varepsilon} \big((x_k - q_k(t))^T A_k(t)(x_k - q_k(t)) + p_k(t) \cdot (x_k - q_k(t)) + c_k(t) \big) \Big)$$

with a $d_k \times d_k$ complex matrix A_k with symmetric positive definite imaginary part; cf. [3, 13]. Denoting by M_k the mass matrix for the (one or several) particles with the coordinates $x_k \in \mathbb{R}^{d_k}$, the equations of motion for A_k and c_k become

$$\begin{aligned} \dot{A}_k &= -2A_k M_k^{-1} A_k - \frac{1}{2} \langle \nabla^2_{x_k} V \rangle \\ \dot{c}_k &= i\varepsilon \, \mathrm{tr}(M_k^{-1} A_k) + \frac{\varepsilon}{8} \langle \, \mathrm{tr}\,(B_k^{-1} \nabla^2_{x_k} V) \, \rangle \end{aligned}$$

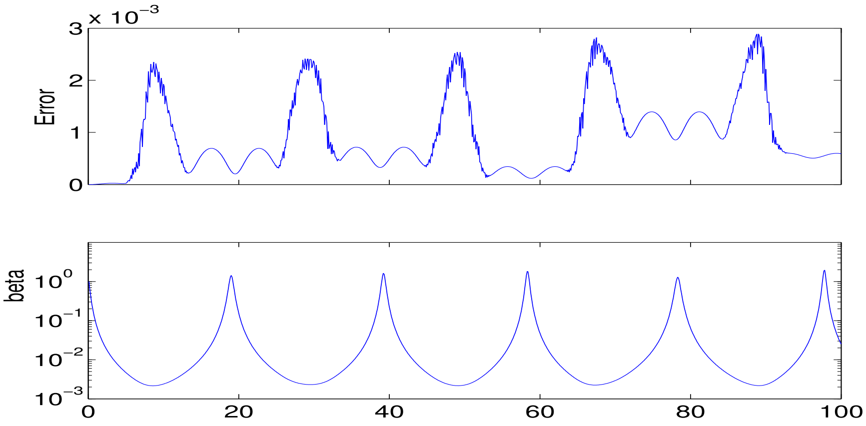

Figure 2: 3D Kepler, $\Delta t = 0.1$, $\varepsilon = 10^{-3}$. Energy error (top) and β (bottom) vs. time.

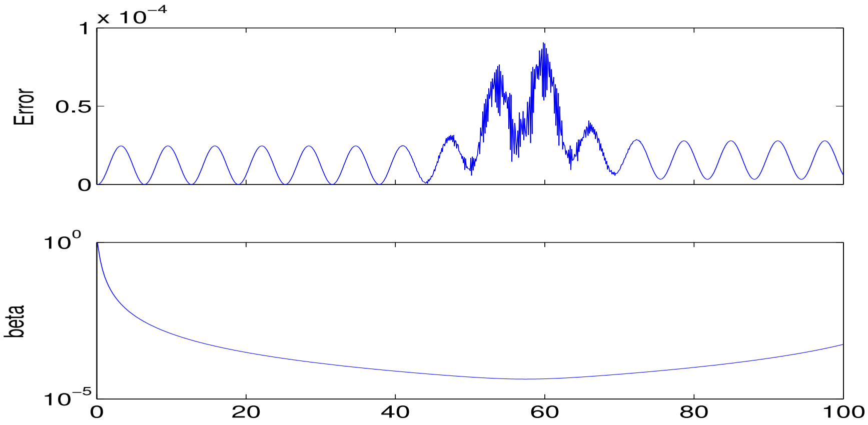

Figure 3: 3D Kepler, $\Delta t = 0.1$, $\varepsilon = 10^{-4}$. Energy error (top) and β (bottom)

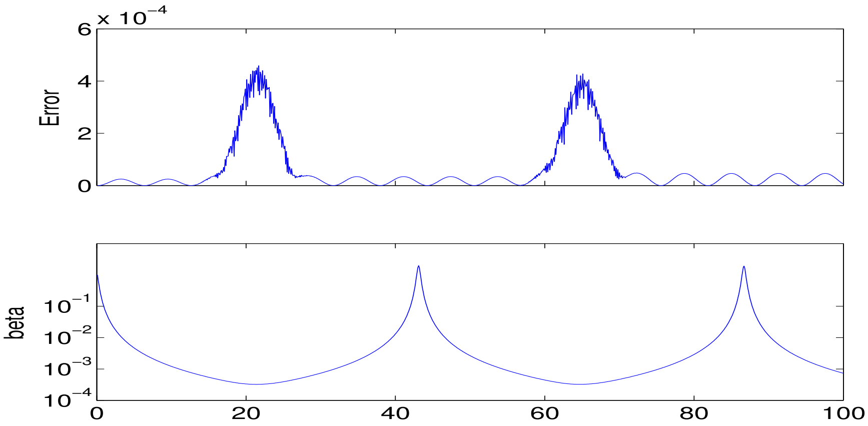

Figure 4: 3D Kepler, $\Delta t = 0.1$, $\varepsilon = 10^{-5}$. Energy error (top) and β (bottom)

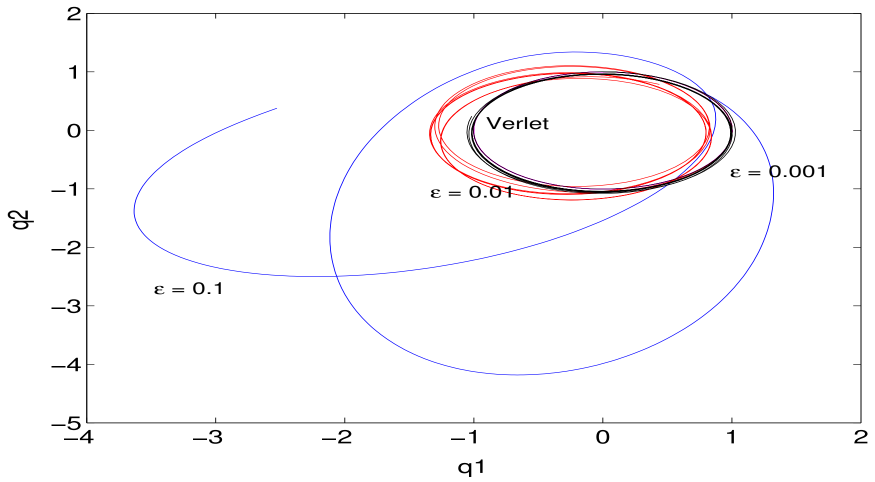

Figure 5: 3D Kepler, $\Delta t = 0.1$, classical limit toward the Verlet integrator.

where $B_k = \operatorname{Im} A_k$ and $\nabla^2_{x_k} V$ is the Hessian matrix of V with respect to the variables x_k. These equations admit the same splitting as previously, which still can be solved explicitly by a straightforward adaptation of the previous formulas. (Note that with $\widehat{A}_k = M_k^{-1/2} A_k M_k^{-1/2}$, the equation $\dot{\widehat{A}}_k = -2\widehat{A}_k^2$ has the solution $\widehat{A}_k(\Delta t) = \widehat{A}_k(0)\big(1 + 2\Delta t \widehat{A}_k(0)\big)^{-1}$.)

6.3 Modified splitting

Instead of decomposing the Hamiltonian as $H = T + V$ it may be preferable in some situations to decompose H as

$$H = \left(T + \sum_{k=1}^{N} Q_k(x_k) \right) + W(x_1, \ldots, x_N),$$

where Q_k is a positive semi-definite quadratic function of x_k. In this case the equations from the variational splitting can still be solved exactly, cf. [10]. We do not write down the full algorithm, but note that in the classical limit $\varepsilon \to 0$ the position coordinates q_k tend to those obtained by applying Deuflhard's method [4]

$$q_k^{n+1} - 2\cos(\Delta t\, \Omega_k)\, q_k^n + q_k^{n-1} = -\frac{\Delta t^2}{m_k} \operatorname{sinc}(\Delta t\, \Omega_k) \nabla_{q_k} W(q^n),$$

with $\Omega_k = (\nabla^2_{x_k} Q_k)^{1/2}$ and $\operatorname{sinc}(\xi) = \sin(\xi)/\xi$, to the classical limit equations $m_k \ddot{q}_k + \Omega_k^2 q_k = -\nabla_{q_k} W(q)$.

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