Computing semiclassical quantum dynamics using Hagedorn wavepackets

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joint work with

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Outline

The Schrödinger equation in the semi-classical regime

Hagedorn wavepackets

A splitting method for time integration
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Schrödinger equation in semi-classical scaling

\[ i\varepsilon \frac{\partial \psi}{\partial t}(x, t) = -\frac{\varepsilon^2}{2m} \Delta_x \psi(x, t) + V(x, t)\psi(x, t) \]

for the wavefunction \( \psi = \psi(x, t) \), \( x = (x_1, \ldots, x_N) \in \mathbb{R}^N \), \( t \geq 0 \)

initial value problem: \( \psi \) specified at time \( t = 0 \)

SE for the nuclei in a molecule \( 0 < \varepsilon \ll 1 \)
Computational challenges

- high dimension: $N = 3 \cdot n_{particles}$

- solutions are highly oscillatory with wavelengths $\sim \varepsilon$
- localized with width $\sim \sqrt{\varepsilon}$, with velocity $\sim 1$

no grids! (neither full nor sparse)
Rescue?

wavefunction is well approximated by

complex Gaussian $\times$ polynomial

$\rightarrow$ Hagedorn wavepackets
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Complex Gaussians in Hagedorn’s parametrization

\[ \varphi_0[q, p, Q, P](x) = (\pi \varepsilon)^{-N/4} (\det Q)^{-1/2} \times \]
\[ \exp\left( \frac{i}{2\varepsilon} (x - q)^T P Q^{-1} (x - q) + \frac{i}{\varepsilon} p^T (x - q) \right), \]

\( q \in \mathbb{R}^N \) position, \( p \in \mathbb{R}^N \) momentum

\( Q, P \) complex \( N \times N \) matrices such that

\( Y = \begin{pmatrix} \text{Re} Q & \text{Im} Q \\ \text{Re} P & \text{Im} P \end{pmatrix} \) is symplectic: \( Y^T J Y = J \) for \( J = \begin{pmatrix} 0 & -I \\ I & 0 \end{pmatrix} \)

Consequence: \( PQ^{-1} \) is complex symmetric with positive definite imaginary part

\[ \text{Hagedorn 1980} \]
Hagedorn wavepackets

$L^2$-orthonormal set of functions $\varphi_k(x) = \varphi_k[q, p, Q, P](x)$
for multi-indices $k = (k_1, \ldots, k_N)$, constructed recursively:
define the raising operator

$$\mathcal{R} = (\mathcal{R}_j) = \frac{1}{\sqrt{2\varepsilon}} \left( P^*(x - q) + Q^*(-i\varepsilon\nabla_x - p) \right)$$

With $\langle j \rangle = (0 \ldots 1 \ldots 0)$ the $j$th unit vector, set

$$\varphi_{k+\langle j \rangle} = \frac{1}{\sqrt{k_j + 1}} \mathcal{R}_j \varphi_k .$$

$\varphi_k$ are polynomials of degree $k_1 + \cdots + k_N$ multiplied with the Gaussian $\varphi_0$ ($N = 1$: Hermite functions).
Recursive evaluation

\[
Q \left( \sqrt{k_j + 1} \varphi_{k+\langle j \rangle}(x) \right)^N_{j=1} = \sqrt{\frac{2}{\varepsilon}} (x-q) \varphi_k(x) - \overline{Q} \left( \sqrt{k_j} \varphi_{k-\langle j \rangle}(x) \right)^N_{j=1}
\]

\( k_1 = 3, \; k_2 = 2 \)

\( k_1 = 4, \; k_2 = 2 \)
Approximate wavefunction by Hagedorn wavepacket

\[
\psi(x, t) \approx e^{iS(t)/\epsilon} \sum_{k \in K} c_k(t) \varphi_k[q(t), p(t), Q(t), P(t)](x)
\]

over multi-index set \( K \)

- in low dimensions, full cube: \( k_j \leq K \) (\( j = 1, \ldots, N \))
- in moderate dimensions, hyperbolic cross:
  \[
  (1 + k_1) \cdot \ldots \cdot (1 + k_N) \leq K
  \]
- in high dimensions, axes: \( k_j > 0 \) only for a single component \( j \)
  in each \( k \) (Hartree-type approximation in a moving frame)

problem-adapted moving basis functions
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Recap: Schrödinger equation

\[ i\varepsilon \frac{\partial \psi}{\partial t} = H\psi \]

with the Hamiltonian

\[ H = T + V \]

composed of the kinetic energy operator

\[ T = -\frac{\varepsilon^2}{2m}\Delta_x \]

and a smooth potential

\[ V = V(x). \]


\[ H = T + U_q(t) + W_q(t) \]

- We can solve exactly the **free Schrödinger equation**, with the wavefunction remaining in the Hagedorn wavepacket form with unaltered coefficients \( c_k \).
- For a **quadratic potential**, we can solve exactly the potential equation with the wavefunction remaining in the Hagedorn wavepacket form with the same coefficients \( c_k \).
- For the **non-quadratic remainder**, we compute the variational approximation of the potential equation on the linear space spanned by the functions \( \varphi_k \) with fixed parameters \( q, p, Q, P \), letting the coefficients \( c_k \) vary.
Free Schrödinger equation

\[ i\varepsilon \frac{\partial \psi}{\partial t} = -\frac{\varepsilon^2}{2m} \Delta \psi \]

A time-dependent Hagedorn wavepacket solves the free Schrödinger equation with modified positions

\[ q(t) = q(0) + \frac{t}{m} p(0) \]
\[ Q(t) = Q(0) + \frac{t}{m} P(0) \]

and unchanged momenta \( p, P \) and unchanged coefficients \( c_k \).

change only position \( q \) and \( Q \) and phase \( S \)
Quadratic potential

\[ i\varepsilon \frac{\partial \psi}{\partial t} = U \psi \]

For a quadratic potential \( U(x) \), a time-dependent Hagedorn wavepacket solves the equation with modified momenta

\[
\begin{align*}
p(t) &= p(0) - t \nabla U(q(0)) \\
P(t) &= P(0) - t \nabla^2 U(q(0)) Q(0)
\end{align*}
\]

and unchanged positions \( q \) and \( Q \) and unchanged coefficients \( c_k \).

change only momentum \( p \) and \( P \) and phase \( S \)
Galerkin approximation for the remainder

\[ i\varepsilon \frac{\partial \psi}{\partial t} = W\psi, \quad W = W(x) \]

fix Gauss parameters \( q, p, Q, P \) in \( \varphi_k(x) = \varphi_k[q, p, Q, P](x) \)

Galerkin condition: determine \( u(x, t) = \sum_{k \in K} c_k(t)\varphi_k(x) \) from

\[ \langle \varphi_k , i\varepsilon \partial_t u - Wu \rangle = 0 \quad \forall \, k \in K \]
Galerkin approximation for the remainder (ctd.)

Galerkin condition determines the coefficient vector $c = (c_k)$ as

$$c(t) = \exp\left(-\frac{it}{\varepsilon} F\right) c(0)$$

with the Hermitian matrix

$$F = (f_{k\ell}), \quad f_{k\ell} = \int_{\mathbb{R}^N} W(x) \overline{\varphi}_k(x) \varphi_\ell(x) \, dx$$

- The integrals are non-oscillatory, approximated by sparse Gauss–Hermite quadrature.
- $F = O(\varepsilon^{3/2})$ if the quadratic Taylor polynomial of $W$ at $q$ vanishes. Therefore, $\exp\left(-\frac{it}{\varepsilon} F\right) c(0)$ is computed efficiently using just a few Lanczos iterations with $F$. 

change only coefficients $c_k$
Time-stepping algorithm

start from position $q^0$, momentum $p^0$, phase $S^0$, width matrices $Q^0$, $P^0$ satisfying the symplecticity condition, and coefficients $c_k^0$

$$\psi(x, t^0) \approx u^0(x) = e^{iS^0/\varepsilon} \sum_{k \in \mathcal{K}} c_k^0 \varphi_k[q^0, p^0, Q^0, P^0](x)$$

determine approximation $u^1(x)$ of the same form after time step $\Delta t$ using a splitting algorithm
Splitting algorithm

1. **Half-step of kinetic part**: updates \( q^{1/2}, Q^{1/2}, S^{1/2}, \). 

2. **Full step of potential part**: split the potential

\[
V(x) = U^{1/2}(x) + W^{1/2}(x)
\]

into its quadratic Taylor polynomial \( U^{1/2}(x) \) at \( q^{1/2} \) and the remainder

- solve with quadratic potential \( U^{1/2} \): updates \( p^1, P^1, S^{1/2},+ \)
- Galerkin approximation for the non-quadratic remainder \( W^{1/2} \):
  update coefficients \( c_k^1 \)

3. **Half-step of kinetic part**: updates \( q^1, Q^1, S^1 \).
Properties

- time-reversible method
- preserves the symplecticity relation of the matrices $Q$ and $P$
- preserves the $L^2$ norm of the wavepacket
- for position $q$ and momentum $p$: Störmer-Verlet method for the corresponding classical Hamiltonian system
- limit of taking the full basis set $\varphi_k$ with all $k \in \mathbb{N}^N$: Strang splitting of the Schrödinger equation
- robust in the semi-classical limit $\varepsilon \to 0$: approximation in the potential part becomes exact for $\varepsilon \to 0$, while the kinetic part is solved exactly for all $\varepsilon$. 
Error behaviour in a numerical example

Maximum error vs. number of basis functions at $t = 1$ and $t = 5$. 
Flying carpet

Squared absolute values of the approximate wave function evaluated on the flying carpet of quadrature points.
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