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

initial value problem: ψ specified at time $t = 0$

SE for the nuclei in a molecule $\quad 0 < \varepsilon \ll 1$

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 ~ 1

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no grids! (neither full nor sparse)

Rescue?

wavefunction is well approximated by

$$\text{complex Gaussian} \times \text{polynomial}$$

$\longrightarrow$ Hagedorn wavepackets

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Complex Gaussians in Hagedorn's parametrization

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

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

Q, P complex $N \times N$ matrices such that

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

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

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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*

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

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

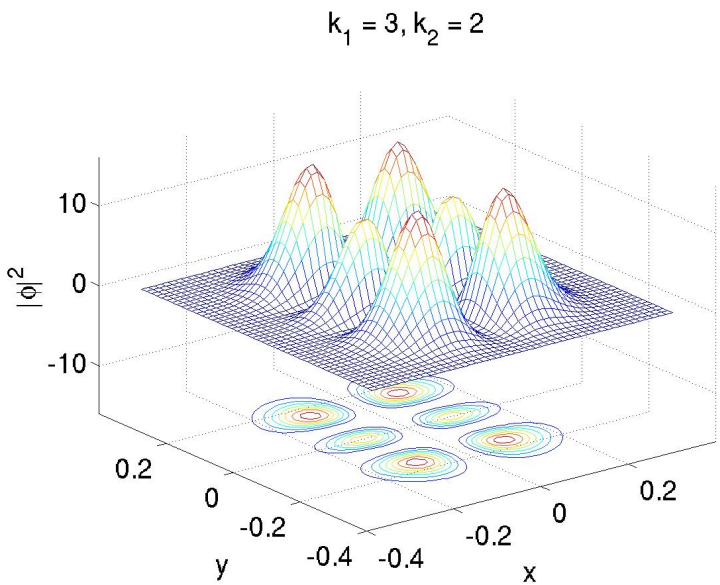
$$\varphi_{k+\langle j \rangle} = \frac{1}{\sqrt{k_j + 1}} \mathcal{R}_j \varphi_k \,.$$

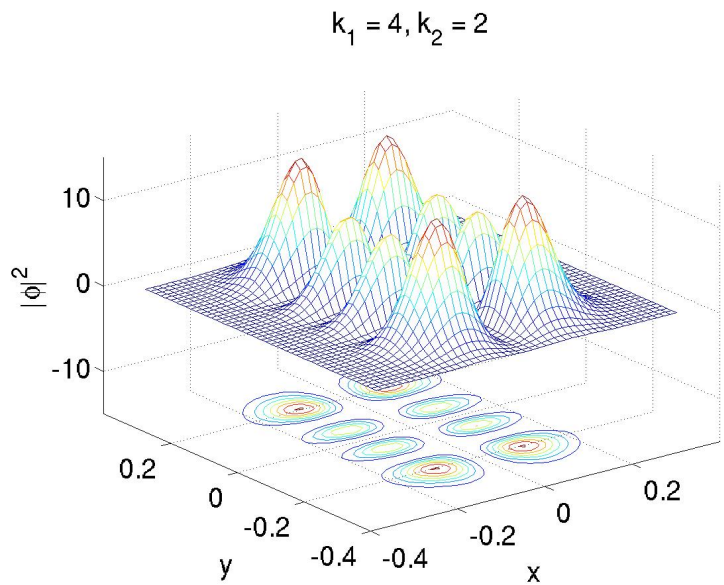
φ_k are polynomials of degree $k_1 + \cdots + k_N$ multiplied with the Gaussian φ_0 ($N = 1$: Hermite functions).

Hagedorn 1998

Recursive evaluation

$$Q\left(\sqrt{k_j+1}\,\varphi_{k+\langle j\rangle}(x)\right)_{j=1}^{N} = \sqrt{\frac{2}{\varepsilon}}\,(x-q)\varphi_k(x) - \overline{Q}\left(\sqrt{k_j}\,\varphi_{k-\langle j\rangle}(x)\right)_{j=1}^{N}$$





Approximate wavefunction by Hagedorn wavepacket

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

over multi-index set $\mathcal{K}$

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

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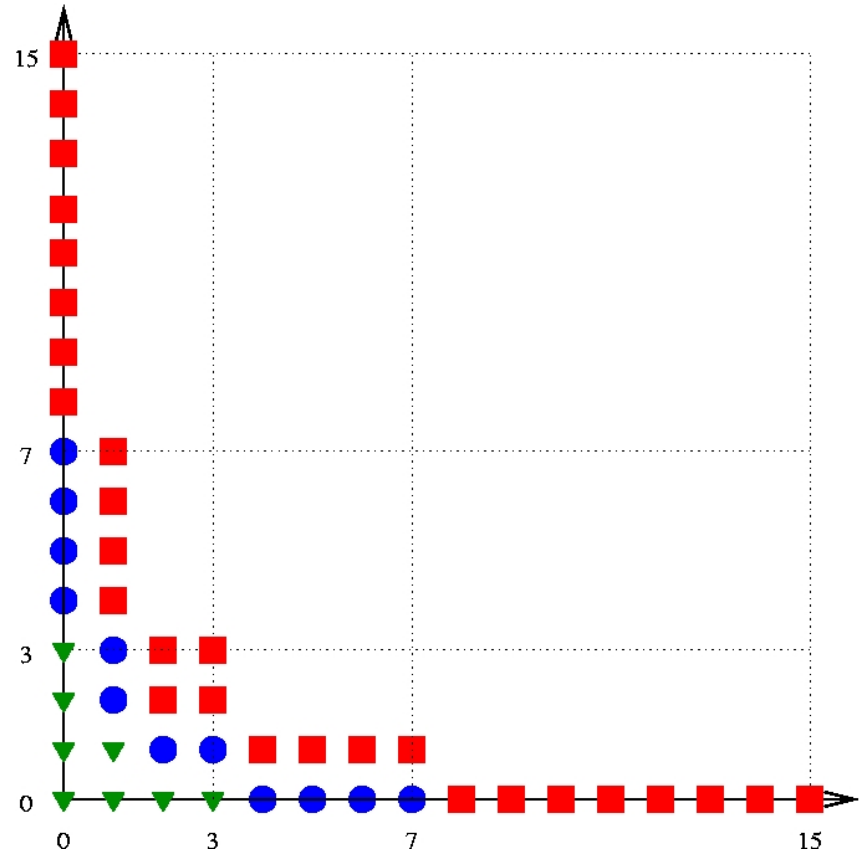
problem-adapted moving basis functions


15
7
3
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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

$$V = V(x).$$

Bits and pieces

$$H = T + U_{q(t)} + W_{q(t)}$$

- 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 φ_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

$$\begin{aligned} q(t) &= q(0) + \frac{t}{m} p(0) \\ Q(t) &= Q(0) + \frac{t}{m} P(0) \end{aligned}$$

and unchanged momenta p, P and unchanged coefficients c_k.

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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

$$\begin{aligned} p(t) &= p(0) - t\,\nabla U(q(0)) \\ P(t) &= P(0) - t\,\nabla^2 U(q(0))Q(0) \end{aligned}$$

and unchanged positions q and Q and unchanged coefficients c_k.

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change only momentum p and P and phase S

Galerkin approximation for the remainder

$$i\varepsilon \frac{\partial \psi}{\partial t} = W\psi\,, \qquad W = W(x)$$

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

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

$$\langle \varphi_k\,,\; i\varepsilon\partial_t u - Wu\rangle = 0 \qquad \forall\, k \in \mathcal{K}$$

Galerkin approximation for the remainder (ctd.)

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

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

with the Hermitian matrix

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

- 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, $\exp\big(-\frac{it}{\varepsilon}\, F\big)\, c(0)$ is computed efficiently using just a few Lanczos iterations with F.

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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 Δt 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

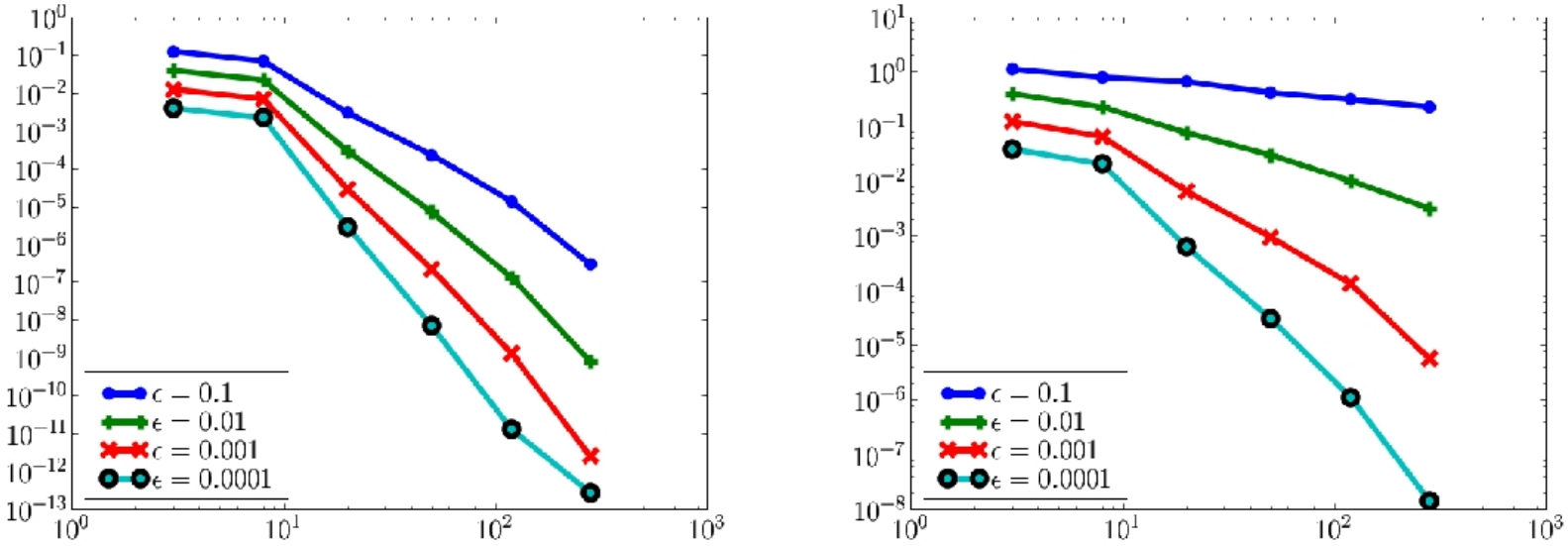
   - solve with quadratic potential $U^{1/2}$: updates p^1, P^1, $S^{1/2,+}$
   - Galerkin approximation for the non-quadratic remainder $W^{1/2}$: update coefficients c_k^1

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

Properties

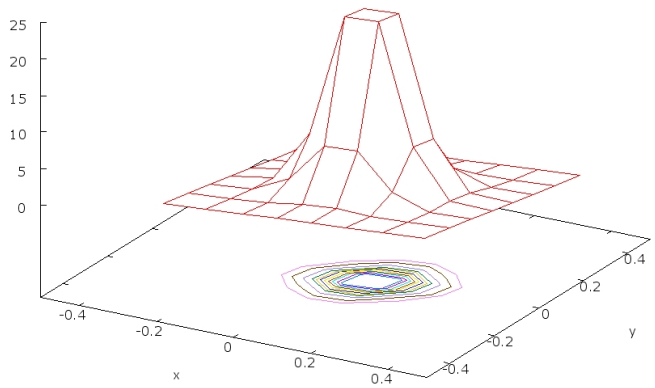
- time-reversible method
- preserves the symplecticity relation of the matrices Q and P
- preserves the L^2 norm of the wavepacket
- for position q and momentum p: Störmer-Verlet method for the corresponding classical Hamiltonian system
- limit of taking the full basis set φ_k with all $k \in \mathbb{N}^N$: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

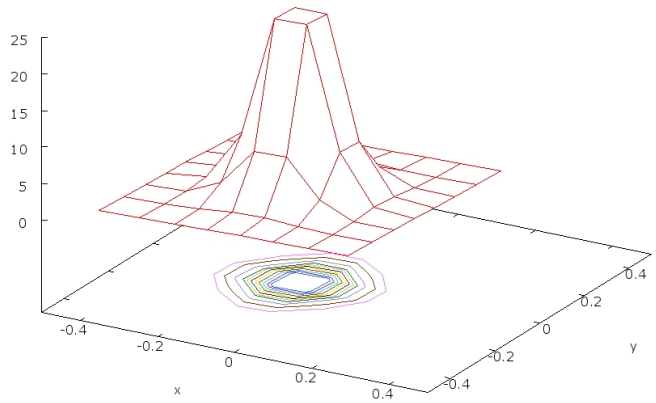
Error behaviour in a numerical example



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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