Chebychev Propagator for Inhomogeneous Schrödinger Equations

Michael Goerz

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Solving the Schrödinger Equation

Schrödinger Equation

$$\frac{\partial}{\partial t}\left|\Psi(t)\right\rangle = \hat{\mathbf{H}}\left|\Psi(t)\right\rangle; \qquad \text{e.g. } \hat{\mathbf{H}} = \begin{pmatrix} V_1(R) & \mu\epsilon(t) \\ \mu\epsilon(t) & V_2(R) \end{pmatrix}$$

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Solution

$$\left|\Psi(t)\right\rangle = e^{-i\hat{\mathbf{H}}t}\left|\Psi_0\right\rangle \qquad \text{if } \hat{\mathbf{H}} \text{ not time dependent}$$

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$$|\Psi(t+\Delta t)\rangle = e^{-i\hat{\mathbf{H}}\Delta t} |\Psi(t)\rangle \quad \rightarrow \text{piecewise constant pulses}$$

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Evaluation of the Time Evolution Operator

$$\text{Expand into series:} \quad e^{-i\hat{\mathbf{H}}t} \longrightarrow \sum_{k=1}^{N} a_n P_n(\hat{\mathbf{H}})$$

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cf. Runge-Kutta: solving the differential equation, instead of evaluating the analytical solution

Chebychev Polynomials

Properties of Chebychev polynomials

- $P_0 = 1,\ P_1(x) = x,\ P_n(x) = 2xP_{n-1}(x) - P_{n-2}(x)$
- Defined over range $[-1, 1] \rightarrow$ normalize Hamiltonian

$$\hat{\mathbf{H}}_{\text{norm}} = 2\frac{\hat{\mathbf{H}} - E_{\text{min}}\mathbb{1}}{\Delta E} - \mathbb{1}$$

- Fastest converging polynomial expansion
- $P_n(x) = \cos(n\theta)$ with $\theta = \arccos(x) \rightarrow$ Cosine transform for coefficients

Chebychev coefficients

- Expansion coefficients a_n for function $f(x)$:

$$a_n = \frac{2 - \delta_{n0}}{\pi} \int_{-1}^{+1} \frac{f(x)P_n(x)}{\sqrt{1 - x^2}}\,\mathrm{d}x$$

- For $f(\hat{\mathbf{H}}_{\text{norm}}) = e^{-i\hat{\mathbf{H}}_{\text{norm}}t}$: $a_n \rightarrow$ Bessel functions.

Inhomogeneous Schrödinger Equation

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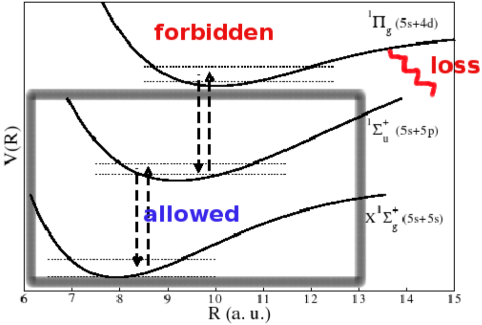
Note: not the same as *nonlinear* SE:

$$\text{e.g.} \quad \frac{\partial}{\partial t} |\Psi(t)\rangle = \left(\hat{\mathbf{H}} + |\Psi(t)|^2\right) |\Psi(t)\rangle$$

OCT with State-Dependent Costs



Optimization Functional

$$J = -F[\Psi_t] + \int g_a[\epsilon(t)]\,\mathrm{d}t + \int g_b[\Psi(t)]\,\mathrm{d}t; \qquad g_b[\Psi] = \lambda_b \left\langle \Psi(t) \middle| \hat{\mathbf{P}}_{\text{allow}} \middle| \Psi(t) \right\rangle$$

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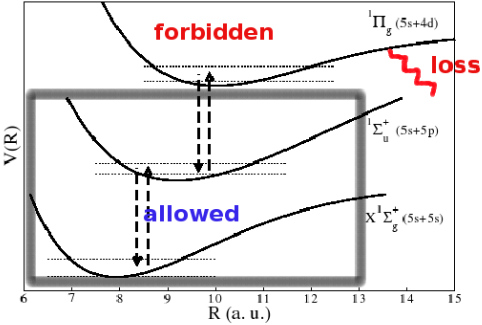

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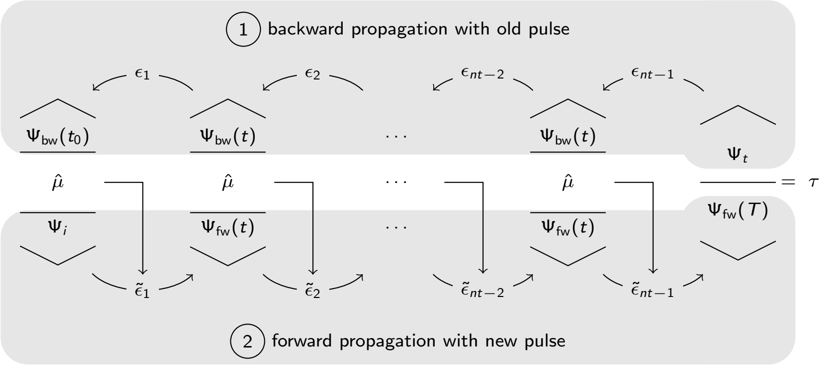
also: time-dependent targets

Reminder: Krotov



Pulse update by matching forward- and backward-propagated states

Inhomogeneous Backward-Propagation

Daniel: Second Order Krotov Preprint (arXiv:1008.5126v1)

Optimization Functional

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Pulse Update

$$\Delta\epsilon \propto -\mathfrak{Im} \left\langle \chi^{(0)}(t) \,|\hat{\boldsymbol{\mu}}|\, \phi^{(1)}(t) \right\rangle$$

Backward Propagation

$$\begin{aligned} \frac{d}{dt}|\chi^{(0)}(t)\rangle &= -\frac{i}{\hbar}\hat{\mathbf{H}}^{\dagger}[\varphi^{(0)}, \epsilon^{(0)}]|\chi^{(0)}(t)\rangle + \nabla_{\langle\varphi|} g_b\Big|_{\varphi^{(0)}(t)} \\ |\chi^{(0)}(T)\rangle &= -\nabla_{\langle\varphi|} J_T\Big|_{\varphi^{(0)}(T)} \end{aligned}$$

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|\chi^{(0)}(T)\rangle &= -\nabla_{\langle\varphi|} J_T \Big|_{\varphi^{(0)}(T)}
\end{aligned}$$

Solving the Inhomogeneous Schrödinger Equation Numerically

References

[1] JCP 130, 124108 (2009)

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A Chebychev propagator for inhomogeneous Schrödinger equations

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[2] JCP 132, 064105 (2010)

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A Chebychev propagator with iterative time ordering for explicitly time-dependent Hamiltonians

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Treating the Inhomogeneity in Order m

Inhomogeneous SE

$$\frac{\partial}{\partial t} |\Psi(t)\rangle = \hat{\mathbf{H}} |\Psi(t)\rangle + |\Phi(t)\rangle$$

Expansion of $\Phi(t)$

$$|\Phi(t)\rangle_m = \sum_{j=0}^{m-1} |\bar{\Phi}_j\rangle P_j(\bar{t})$$

- Expand inhomogeneous term in Chebychev series

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Expansion of $\Phi(t)$

$$|\Phi(t)\rangle_m = \sum_{j=0}^{m-1} \left|\bar{\Phi}_j\right\rangle P_j(\bar{t}) \equiv \sum_{j=0}^{m-1} \frac{t^j}{j!} \left|\Phi^{(j)}\right\rangle$$

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- Reorder into power series (or use Taylor to begin with)

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- Expand inhomogeneous term in Chebychev series
- Reorder into power series (or use Taylor to begin with)
- Decide on which order to solve: 1, 2, 3, maybe 4
- The smaller the order, the smaller Δt has to be

The Analytical Solution

Inhomogeneous SE (Φ to order m)

$$\frac{\partial}{\partial t}\left|\Psi(t)\right\rangle = \hat{\mathbf{H}}\left|\Psi(t)\right\rangle + \sum_{j=0}^{m-1}\frac{t^j}{j!}\left|\Phi^{(j)}\right\rangle$$

Solution

$$\left|\Psi(t)\right\rangle_{(m)} = \sum_{j=0}^{m-1}\frac{t^j}{j!}\left|\lambda^{(j)}\right\rangle + f_m(\hat{\mathbf{H}})\left|\lambda^{(m)}\right\rangle$$

$$f_m = (-i\hat{\mathbf{H}})^{-m}\left(e^{-i\hat{\mathbf{H}}t} - \sum_{j=0}^{m-1}\frac{(-i\hat{\mathbf{H}}t)^j}{j!}\right)$$

$$\lambda^{(0)} = \left|\Psi_0\right\rangle$$

$$\lambda^{(j)} = -i\hat{\mathbf{H}}\left|\lambda^{(j-1)}\right\rangle + \left|\Phi^{(j-1)}\right\rangle$$

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e.g. $\left|\Psi(t)\right\rangle_{(3)} = \left|\Psi_0\right\rangle + t\left|\lambda^{(1)}\right\rangle + \frac{t^2}{2}\left|\lambda^{(2)}\right\rangle + f_3(\hat{\mathbf{H}})\left|\lambda^3\right\rangle$, with
$f_3(\hat{\mathbf{H}}) = \left(-i\hat{\mathbf{H}}\right)^{-3}\left(e^{-i\hat{\mathbf{H}}t} - 1 - i\hat{\mathbf{H}}t\right)$

The Analytical Solution

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$$\frac{\partial}{\partial t} |\Psi(t)\rangle = \hat{\mathbf{H}} |\Psi(t)\rangle + \sum_{j=0}^{m-1} \frac{t^j}{j!} \left|\Phi^{(j)}\right\rangle$$

Solution

$$|\Psi(t)\rangle_{(m)} = \sum_{j=0}^{m-1} \frac{t^j}{j!} \left|\lambda^{(j)}\right\rangle + f_m(\hat{\mathbf{H}}) \left|\lambda^{(m)}\right\rangle$$

$$f_m = (-i\hat{\mathbf{H}})^{-m} \left(e^{-i\hat{\mathbf{H}}t} - \sum_{j=0}^{m-1} \frac{(-i\hat{\mathbf{H}}t)^j}{j!} \right)$$

$$\lambda^{(0)} = |\Psi_0\rangle$$

$$\lambda^{(j)} = -i\hat{\mathbf{H}} \left|\lambda^{(j-1)}\right\rangle + \left|\Phi^{(j-1)}\right\rangle$$

e.g. $\quad |\Psi(t)\rangle_{(0)} = e^{-i\hat{\mathbf{H}}t} |\Psi_0\rangle \quad \rightarrow \quad$ homogeneous propagation

Chebychev Propagation

Solution

$$|\Psi(t)\rangle_{(m)} = \sum_{j=0}^{m-1} \frac{t^j}{j!} \left|\lambda^{(j)}\right\rangle + f_m(\hat{\mathbf{H}}) \left|\lambda^{(m)}\right\rangle ; \qquad |\lambda\rangle \sim \left\{ \left|\Phi^{(j)}\right\rangle \right\}$$

Idea

Evaluate $f_m(\hat{\mathbf{H}})$ by expanding it into Chebychev Polynomials
(just like for the "standard" Chebychev propagator with $f_0(\hat{\mathbf{H}}) = e^{i\hat{\mathbf{H}}t}$)

Chebychev Propagation

Solution

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Evaluate $f_m(\hat{\mathbf{H}})$ by expanding it into Chebychev Polynomials
(just like for the "standard" Chebychev propagator with $f_0(\hat{\mathbf{H}}) = e^{i\hat{\mathbf{H}}t}$)

Algorithm Outline (for fixed m)

For each time step:

- determine $\left\{\left|\Phi^{(j)}\right\rangle\right\}$ and from that $\left\{\lambda^{(j)}\right\}$
- run through the Chebychev series for f_m
- sum everything up, yielding $|\Psi(t)\rangle_{(m)}$

Details of the Algorithm

Global Initialization (before any actual propagation)

Calculate Chebychev Coefficients

Calculate the Chebychev expansion coefficients a_n for $f_m(\hat{\mathbf{H}})$, for the chosen order m.

Global Initialization (before any actual propagation)

Calculate Chebychev Coefficients

Calculate the Chebychev expansion coefficients a_n for $f_m(\hat{\mathbf{H}})$, for the chosen order m.

- a_n *cannot* be calculated analytically (like for the standard Chebychev)
- Calculation of a_n is done through a fast cosine-transform:

$$a_n = \frac{2 - \delta_{n0}}{N} \sum_{k=0}^{N-1} f_m(\theta_k) \cos(n\theta_k)$$

- $\hat{\mathbf{H}}$ needs to be normalized $\rightarrow$ a_n might have to be re-calculated if spectral radius changes (after each OCT iteration)
- On a non-equidistant time grid, the a_n would have to be re-calculated at every time step
- *For small $\hat{\mathbf{H}}$, the term $(-i\hat{\mathbf{H}})^{-m}$ might lead to numerical instability. We could use Taylor instead. ...?*

Local Initialization (at every time step)

Calculate Expansion of Inhomogeneous Term

Calculate all necessary $\left|\Phi^{(j)}\right\rangle$ (i.e. up to order m) to approximate the local $\Phi(t_i)$, either by an intermediate Chebychev expansion, fallowed by calculation of coefficients in the power series, or by a direct Taylor series.

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Calculation via Taylor:

- Calculate derivatives through FFT

Calculation via Chebychev:

- Sample $\Phi(t)$ at intermediate points around t by interpolation (splining should be fine)
- Calculate Chebychev coefficients $\left|\bar{\Phi}_j\right\rangle$ by cosine transform
- Calculate $\left|\Phi^{(j)}\right\rangle$ from $\left|\bar{\Phi}_j\right\rangle$ by formulas in References (just collect the powers)

Propagation Step

Solution

$$|\Psi(t)\rangle_{(m)} = \sum_{j=0}^{m-1} \frac{t^j}{j!} \left|\lambda^{(j)}\right\rangle + f_m(\hat{\mathbf{H}}) \left|\lambda^{(m)}\right\rangle$$

- Calculate $\left|\lambda^{(j)}\right\rangle$, $j = 0 \dots m-1$

$$\lambda^{(j)} = -i\hat{\mathbf{H}} \left|\lambda^{(j-1)}\right\rangle + \left|\Phi^{(j-1)}\right\rangle$$

- Calculate $f_m(\hat{\mathbf{H}}) \left|\lambda^{(m)}\right\rangle$ by Chebychev recursion (Choose N to reach machine precision)

$$f_m(\hat{\mathbf{H}}) \left|\lambda^{(m)}\right\rangle = \sum_{n=1}^{N} a_n P_n(\hat{\mathbf{H}}) \left|\lambda^{(m)}\right\rangle$$

$$P_n(\hat{\mathbf{H}}) = 2\hat{\mathbf{H}} P_{n-1}(\hat{\mathbf{H}}) - P_{n-2}(\hat{\mathbf{H}})$$

- Calculate $|\Psi(t)\rangle_{(m)}$

Inhomogeneous Chebychev in QDYN

Propagation Routines (prop.f90)

prop

```
subroutine prop(psi, grid, ham, work, para, from_ti, to_ti, bw, &
&               info_hook, pulses, alt_pulse, upd_hook, storage)
```

prop_step

```
subroutine prop_step(psi, grid, ham, work, para, ti, bw, &
&                    pulses, alt_pulses, alt_pulse, upd_hook)
```

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```
subroutine prop(psi, grid, ham, work, para, from_ti, to_ti, bw, &
&               info_hook, pulses, alt_pulse, upd_hook, storage)
```

prop with optional inhomogeneous term

```
subroutine prop(psi, grid, ham, work, para, from_ti, to_ti, bw, inh_psi, &
&               get_inh_phi, info_hook, pulses, alt_pulse, upd_hook, storage)
```

- `inh_psi`: stored array of forward-propagated states
- `get_inh_phi`: function that calculates $|\Phi\rangle$ from $|\Psi\rangle$ (e.g. $\hat{\mathbf{P}}_{\text{allow}} |\Psi\rangle$).

prop_step

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Calculation of Chebychev Coefficients

Calculate Chebychev Coefficients

Calculate the Chebychev expansion coefficients a_n for $f_m(\hat{\mathbf{H}})$, for the chosen order m.

in `inhom_cheby.f90`:
`subroutine init_inh_cheby(ham, wcheby, order, para)`

- Use the same work array (`wcheby`) as for the homogeneous Chebychev propagation.
- Sufficient a_n are calculated and stored in `wcheby` to reach machine precision
- Watch out for numerical instability ($\rightarrow$ Taylor)

Expansion of Inhomogeneous Term

Calculate Expansion of Inhomogeneous Term

Calculate all necessary $\left|\Phi^{(j)}\right\rangle$ (i.e. up to order m) to approximate the local $\Phi(t_i)$, either by an intermediate Chebychev expansion, fallowed by calculation of coefficients in the power series, or by a direct Taylor series.

in `inhom_cheby.f90`:

```
subroutine expand_inh_phi(inh_psi, get_inh_phi, order, phi_coeffs)
```

- phi_coeffs stores the $\left|\Phi^{(j)}\right\rangle$ (power series)
- invoke `monic_transf` to calculate $\left|\Phi^{(j)}\right\rangle$ from $\left|\bar{\Phi}_j\right\rangle$

Continue with calculation of $\left|\lambda^{(j)}\right\rangle$:

```
subroutine get_inh_lambda(lambda, phi, ham, ...)
```

Performing the Propagation Step

Solution

$$|\Psi(t)\rangle_{(m)} = \sum_{j=0}^{m-1} \frac{t^j}{j!} \left|\lambda^{(j)}\right\rangle + f_m(\hat{\mathbf{H}}) \left|\lambda^{(m)}\right\rangle$$

in `inhom_cheby.f90`:

```
subroutine inhom_cheby(psi, work, ham, grid, lambda, para, dt, ti, &
&                      alt_pulses, alt_pulse)
```

- Identical interface to `cheby`, except for λ
- Inhomogeneous Chebychev coefficients are implicit in `work`