# Supporting Information: Functional Tensor-Train Chebyshev Method for Multidimensional Quantum Dynamics Simulations

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# 1 Computational Performance

The favorable computational performance of the tensor-train Chebyshev (TTC) method is demonstrated through analysis of a system whose time evolution is known exactly, a Gaussian coherent state in the harmonic oscillator potential

$$V(x) = \sum_{i=1}^{D} \frac{1}{2} m\omega^2 x_i^2 \tag{1}$$

of dimension $D = 2$, mass $m = 1\,\text{au}$, and frequency $\omega = 1\,\text{au}$.

The benchmark analytic time evolution of the Gaussian state is determined recognizing that a coherent state $|\alpha\rangle$ can be expanded in terms of the harmonic oscillator eigenstates states $|n\rangle$ of energy $E_n = (n + 1/2)\hbar\omega$ as follows

$$|\alpha\rangle = e^{-\frac{1}{2}|\alpha|^2} \sum_{n=0}^{\infty} \frac{\alpha^n}{\sqrt{n!}} |n\rangle \tag{2}$$

where $\alpha$ is a complex number that indicates the displacement of the coherent state from the origin in phase space, such that the coherent state is an eigenstate of the annihilation operator

$$\hat{a}\,|\alpha\rangle = \alpha\,|\alpha\rangle \tag{3}$$

where

$$\hat{a}\,|n\rangle = \sqrt{n}\,|n-1\rangle \tag{4}$$

and an eigenstate of the time-evolved annihilation operator in the Heisenberg picture

$$\hat{a}\left(t\right)\left|\alpha\right\rangle = e^{\frac{i}{\hbar}\hat{H}t}\hat{a}e^{-\frac{i}{\hbar}\hat{H}t}\left|\alpha\right\rangle \tag{5}$$

$$= \alpha e^{-\frac{1}{2}|\alpha|^2}\sum_{n=1}^{\infty}\frac{\alpha^{n-1}}{\sqrt{n-1!}}e^{-\frac{i}{\hbar}(E_n-E_{n-1})t}\left|n-1\right\rangle \tag{6}$$

$$= \alpha e^{-i\omega t}e^{-\frac{1}{2}|\alpha|^2}\sum_{n=0}^{\infty}\frac{\alpha^{n}}{\sqrt{n!}}\left|n\right\rangle \tag{7}$$

$$= \alpha e^{-i\omega t}\left|\alpha\right\rangle \tag{8}$$

The time evolution of the coherent state in the Schrödinger picture is then determined by the action of the annihilation operator

$$\hat{a}\left|\alpha\left(t\right)\right\rangle = \alpha(t)\left|\alpha\left(t\right)\right\rangle \tag{9}$$

with the time-dependent displacement of the coherent state

$$\alpha\left(t\right) = e^{-i\omega t}\alpha\left(0\right) \tag{10}$$

which yields the position-space representation of the time-evolved coherent state as a coherent state of the form

$$\Psi\left(x,t\right) = \langle x|\alpha\rangle = \left(\frac{\omega}{\pi\hbar}\right)^{1/4}\exp\left(-\frac{1}{2}\left|\alpha\left(0\right)\right|^2 + \frac{\omega}{2\hbar}x^2 - \left(\sqrt{\frac{\omega}{\hbar}}x - \frac{e^{-i\omega t}\alpha\left(0\right)}{\sqrt{2}}\right)^2\right) \tag{11}$$

We find the wavepacket determined by tensor-train Chebyshev (TTC) quantum dynamics is significantly more accurate than the short-time Tensor-Train Split Operator Fourier Transform (TT-SOFT) approach for long time steps, as shown in Fig. 1. Whereas the error of the $L^2$-norm of the wavefunction is nearly equivalent for both methods for short time steps, TTC produces the wavepacket with several orders of magnitude lower $L^2$-norm error for time steps on the order of 100 to 1000 times longer than those required by TT-SOFT

given a sufficient number of Chebyshev polynomials in the expansion of the propagator. This suggests TTC can require fewer costly Fourier transform computations than TT-SOFT for long time steps.

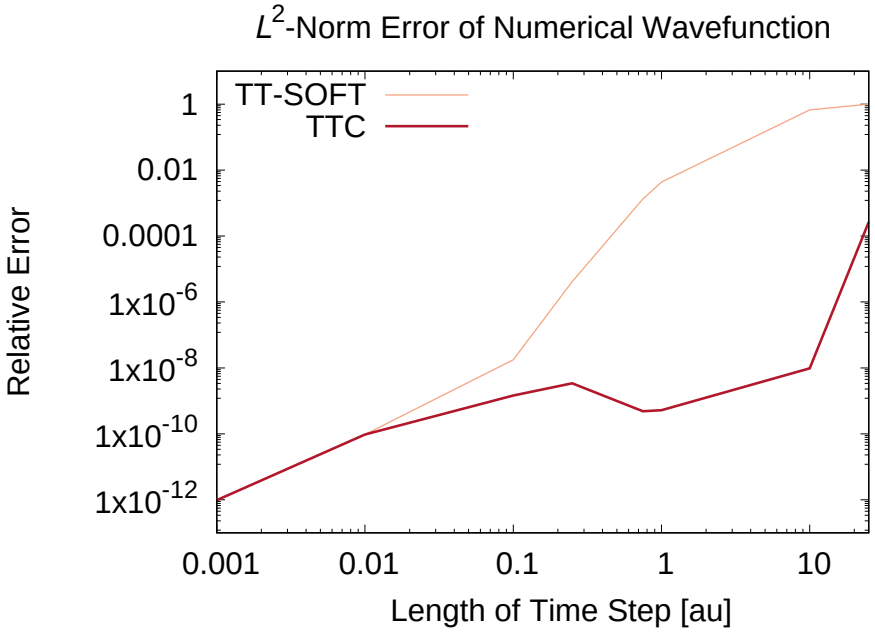


Figure 1: The $L^2$-norm error of the numerical wavefunction is significantly lower for TTC (red line, $N = 750$ Chebyshev polynomials) than TT-SOFT (orange line) for long time steps.

Likewise, TTC molecular dynamics is found to be more accurate relative to TT-SOFT for computation of the autocorrelation function over long time steps (see Fig. 2). For short time steps, TTC and TT-SOFT closely agree, and TTC reduces the relative error of the autocorrelation function by orders of magnitude at the longest time steps considered. We find TTC yields the autocorrelation function with only one percent error with a time step 100 times longer than the maximum time step that can be used to accurately simulate the dynamics with TT-SOFT. The TTC molecular dynamics method therefore successfully maintains accuracy and, where desired, avoids calculation of the wavefunction at intermediate time steps for calculation of the autocorrelation function at the final time.

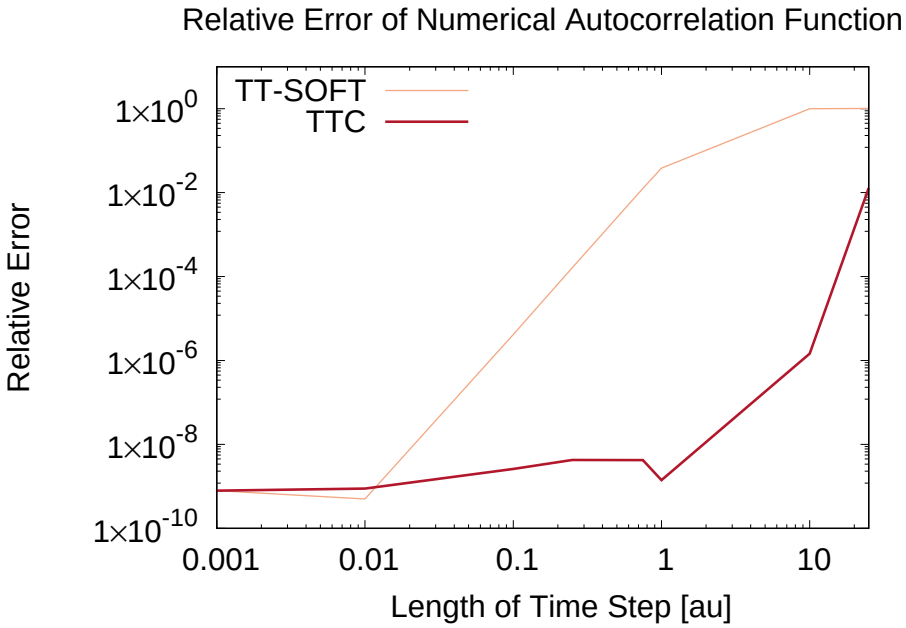


Figure 2: TTC (red line, $N = 750$ Chebyshev polynomials) significantly reduces the relative error associated with determination of the autocorrelation function of the benchmark system relative to the state-of-the-art short-time TT-SOFT method (orange line) for long time steps.

Examination of the relative error of the $L^2$-norm of the wavefunction as a function of the number of terms in the Chebyshev expansion confirms the ability of the TTC algorithm to achieve high accuracy solutions for long time steps given a sufficient number of Chebyshev polynomials, as shown in Fig. 3. The TTC approach requires fewer than 200 Chebyshev polynomials to accurately determine the propagator for a final time of 1 au, such that the method is efficient for short-time propagation, which is beneficial for computation of the wavefunction at intermediate time steps. As expected, the number of polynomials required for accurate simulation of the benchmark system's dynamics increases for direct propagation of the initial wavepacket for larger final times. The expansion is shown to converge for final times up to 6 au for fewer than 500 polynomials, which demonstrates the robustness of TTC for simulation of long-time dynamics in agreement with the measure of Chebyshev accuracy previously observed in the one-dimensional case.

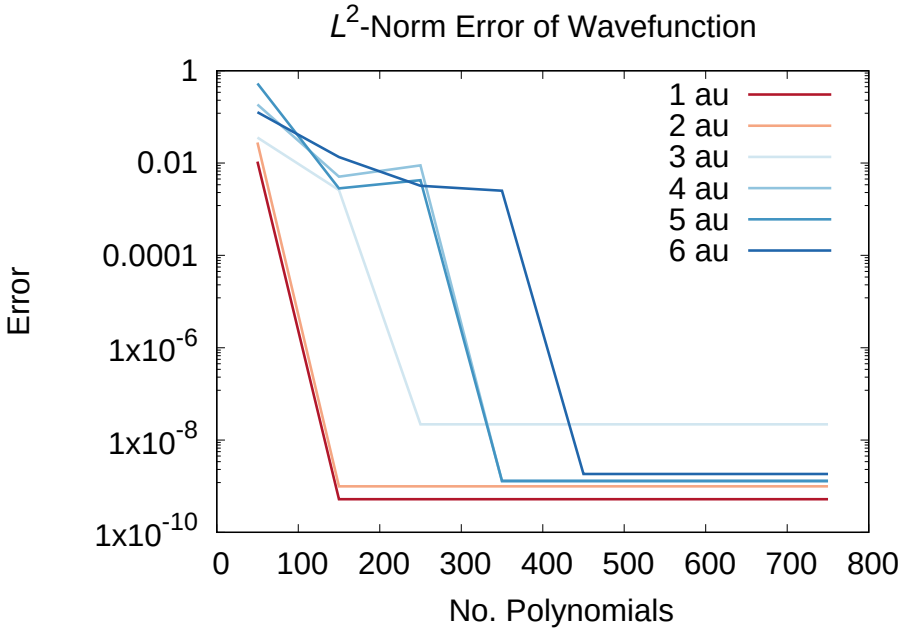


Figure 3: $L^2$-norm error of the wavefunction in TTC as a function of the number of Chebyshev polynomials for varying final times.