Multiscale quantum propagation using compact-support wavelets in space and time

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Abstract

Orthogonal compact-support Daubechies wavelets are employed as bases for both space and time variables in the solution of the time-dependent Schrödinger equation. Initial value conditions are enforced using special early-time wavelets analogous to edge wavelets used in boundary-value problems. It is shown that the quantum equations may be solved directly and accurately in the discrete wavelet representation, an important finding for the eventual goal of highly-adaptive multiresolution Schrödinger equation solvers. While the temporal part of the basis is not sharp in either time or frequency, the Chebyshev method used for pure time-domain propagations is adapted to use in the mixed domain and is able to take advantage of Hamiltonian matrix sparseness. The orthogonal separation into different time scales is determined...
theoretically to persist throughout the evolution and is demonstrated numerically in a partially-adaptive treatment of scattering from an asymmetric Eckart barrier.

I. Introduction

Many problems in molecular and chemical physics have benefited in recent decades from the ability to accurately solve the time-dependent Schrödinger equation (TDSE). Accordingly, a number of different basis-function and grid methods have been developed for calculating the full quantum dynamics of systems with a few degrees of freedom. One application of interest to our own group is in the use of the time-domain description of Dissociative Resonance Raman Spectroscopy for polyatomic molecules, where computational analysis generally requires handling of both multimode large-amplitude vibrational excitation and dissociation dynamics. Ultimately, it is to be expected that highly-adaptive TDSE solvers will emerge, based on one or more robust algorithms. One flexible approach currently being pursued in a variety of quantum calculations is the use of wavelet bases, which can provide locally customizable resolution according to problem requirements. Of particular interest is the availability of orthogonal compact support bases, e.g., those due to Daubechies and other generalizations, for which fast transform methods have already been extensively employed in signal processing and other applications.

For time-independent quantum problems, recent work in our laboratory has focused on systematic and accurate orthogonal wavelet calculations for general potential functions in Cartesian coordinates and, with suitable adaptation, in curvilinear coordinates. The extension to curvilinear coordinates, important for large-amplitude descriptions of nuclear motion, relies on the construction of special edge wavelets near the boundaries of coordinate domains ("wavelets
on the interval") in order to correctly satisfy quantum boundary conditions in prototypical problems. Wavelet bases are capable of democratically representing spatial coordinates in a wide variety of systems, with particular advantages in systems characterized by large dynamic scale variation. This characteristic carries over to the spatial representation in time-dependent calculations as well, for which some initial investigations have already been performed.\textsuperscript{10,11} In the present paper, a new extension is explored, namely the wavelet transformation of both space and time coordinates. In analogy to boundary value wavelet methods,\textsuperscript{8} edge wavelets in the time variable are utilized to allow proper satisfaction of initial value problems. A fundamental goal of this initial investigation is to establish that it is possible to solve the TDSE directly (and accurately) in the wavelet representation. Since the temporal wavelets are not sharp in either time or frequency, this constitutes a joint time-and-frequency approach (technically, "time-and-scale" is more appropriate).

Different wavelet families are available for use, e.g., orthogonal and biorthogonal, single and multiple, exponentially and strictly localized, smooth and rough, etc. The wavelet families due to Daubechies, which started the huge interest in the digital application arena, are orthogonal and strictly localized, with different smoothness properties from one family to the next.\textsuperscript{13} This provides an ability to systematically study the effects of basis set smoothness in the wavelet propagation. For these reasons, Daubechies wavelets are employed below. It is expected that other (present or future) wavelet choices will eventually be determined to offer superior performance in applications, but the choice is not critical here. In any compact support wavelet basis, the strictly-localized nature of the functions gives operator matrices sparse character of one form or another depending on the number of resolution levels chosen. This suggests the use of Krylov basis or related methods that can exploit fast matrix-vector multiplication steps. The
susceptibility of Krylov methods to roundoff error in finite precision is well-known, and this is shown to be of concern as well in the particular case of wavelets. It is found that the degree of smoothness of the basis is a significant factor in the buildup of roundoff error, a fact which is utilized in starting the propagation within the span of the edge wavelets.

For subsequent regions in time, we show that one may use a short-time propagator to determine the wavelet coefficients sequentially (only time-independent Hamiltonians are considered in this paper). While neighboring time wavelets share support, their orthogonality permits adaptation of more usual sharp-time propagation methods to the present purposes. The Chebyshev method familiar from pure time-domain calculations is chosen for use in two 1D examples, periodic motion of a displaced harmonic oscillator and scattering of a particle from an asymmetric Eckart potential.

Remarkably, it is found that, quite generally, multiscale wavelet decomposition along the time coordinate is preserved at all times. In other words, once the early-time wave packet is resolved into orthogonal contributions on different time scales, these components can be propagated independently. This result, which was derived theoretically on the basis of the orthonormality of the wavelet families used, was further verified numerically.

II. Time-dependent Schrödinger Equation

A. Space and time wavelet bases

With \( \hat{H} \) a 1D time-independent Hamiltonian, the TDSE takes the form

\[
 i\hbar \frac{\partial}{\partial t} y(x,t) = \hat{H} y(x,t), \quad (1)
\]

\[
 y(x,0) = f(x), \quad (2)
\]
with \( f(x) \) a known function representing the initial values of \( y \). The solution may be pursued by expansion in a wavelet basis, a prototypical example being the Daubechies compact support families generated from one father wavelet, or scaling function, \( \phi(x) \), and one mother wavelet, \( \psi(x) \). Each of these is nonzero only on a compact interval \([0, L-1]\) with \( L \) even. They satisfy two-scale relations of the form

\[
\phi(x) = \sum_{k=0}^{L-1} c_k \phi(2x - k), \tag{3}
\]

\[
\psi(x) = \sum_{k=0}^{L-1} d_k \phi(2x - k). \tag{4}
\]

The parental functions are constructed such that the first \( L/2 \) moments of \( \psi(x) \) vanish:

\[
\int x^p \psi(x) dx = 0, \quad 0 \leq p < L/2. \]

Roughly, \( \psi(x) \) contains higher-order detail of the finer-scale functions \( \phi(2x - k) \), while the orthogonal scaling function \( \phi(x) \) represents averaged lower-order detail. Normalized functions on different octaves are obtained by dilating and translating the father and mother wavelets,

\[
\phi_{jk}^\lambda(x) = \lambda_j^{-1/2} \phi(x/\lambda_j - k), \tag{5}
\]

\[
\psi_{jk}^\lambda(x) = \lambda_j^{-1/2} \psi(x/\lambda_j - k), \tag{6}
\]

with \( \lambda_j = \lambda / 2^j \). Both \( \phi_{jk}^\lambda \) and \( \psi_{jk}^\lambda \) exist on the supports \([k/\lambda_j, (k + L - 1)/\lambda_j]\) where \( \lambda_j \) is the spacing between basis functions and the parameter \( \lambda \), not necessarily integer, can be chosen according to the physical problem considered. The \( L = 8 \) functions are shown in Fig. 1 as an example.
Figure 1. The Daubechies wavelet family with $L = 8$ and $\lambda = 1$. The father wavelet or scaling function (solid line) is shown in the center while the mother wavelet (solid line) is shown at the top. These functions and their translated copies (dashed lines) are all mutually orthogonal. The father and mother wavelets are both linear combinations of eight normalized scaling functions (solid lines) on the finer scale shown at the bottom. The wavelet subspace at the top exactly spans the difference in detail between the two scaling functions subspaces at different resolutions.

The initial value function $f(x)$ can be expanded in an array of scaling functions on a level $j = J$,

$$f(x) = \sum_k f_{jk}^* \phi_{jk}^*(x),$$

(7)

$$f_{jk}^* = \int \phi_{jk}^*(x) f(x) dx,$$

(8)
where the accuracy of the expansion increases systematically with increasing $J$ and, for localized $f(x)$, the sum over $k$ is finite. This single-scale representation may be replaced by a multiresolution expansion

$$f(x) = \sum_k f_{j_0k}^\lambda \phi_{j_0k}^\lambda(x) + \sum_{j=j_0}^{J-1} \sum_k g_{jk}^\lambda \psi_{jk}^\lambda(x), \quad (9)$$

$$g_{jk}^\lambda = \int \psi_{jk}^\lambda(x) f(x) dx, \quad (10)$$

using the relations derived from Eqs. (3)-(6),

$$\phi_{jk}^\lambda(x) = 2^{-1/2} \sum_{k_0}^{L-1} c_{k_0}^\lambda \phi_{j1,2k+k_0}^\lambda(x), \quad (11)$$

$$\psi_{jk}^\lambda(x) = 2^{-1/2} \sum_{k_0}^{L-1} d_{k_0}^\lambda \phi_{j1,2k+k_0}^\lambda(x), \quad (12)$$

and the corresponding relations between the projections of $f(x)$ of the basis functions defined in Eqs. (8) and (10). Obtaining the coefficients $f_{j_0k}^\lambda$ and $g_{jk}^\lambda$ for $j_0 \leq j < J$ corresponds to taking the Wavelet Transform (WT) of the function $f(x)$ in the (orthogonal) basis of the $\phi_{jk}^\lambda(x)$ and $\psi_{jk}^\lambda(x)$. The wavelets cover the difference in detail between scaling function expansions of $f(x)$ at adjacent scales $j$ and $j+1$, and are especially important in locations where $f(x)$ possesses significant variability. Conversely, they are unimportant in regions where $f(x)$ is smooth on level $j$, manifested by tiny magnitudes of the wavelet coefficients $g_{jk}^\lambda$. This forms the basis of compression methods in signal and image processing,\(^{13,14}\) where small $g_{jk}^\lambda$ are set exactly to zero and the wavelet representation can be shrunk adaptively scale-by-scale and location-by-location.

The same compressional advantage can in principle be applied to continuous functions and pursuit of solutions of differential equations, though the latter generally require significantly increased accuracy, e.g., even in the determination of the initial projection coefficients $f_{j_0k}^\lambda$. This
problem is addressed in a systematically improvable manner by previously-developed wavelet quadrature methods.\textsuperscript{7,16}

The wave function at other times can be expanded in the spatial wavelets and the time-dependent coefficients determined by integration of coupled equations\textsuperscript{11} or other means.\textsuperscript{10} In principle, however, it would be advantageous to be able to focus on the important regions and scale behavior of wave packets in both space and time, i.e., where they traveled and when they were there. Even if for no other reason, it can be expected that this will allow greater compression of the information gained in the full propagation in the same sense that video compression rates can be significantly higher than still image compression rates. The time variable in the following is therefore also subjected to a wavelet transform. For simplicity, we first consider only a single scale in each variable. An appropriate expansion of the solution to the TDSE therefore takes the form

$$y(x,t) = \sum_{n_r=0}^{L/2-1} \sum_{k_x} y^e_{J_x,n_x,J_x,k_x} \Phi^e_{J_x,n_x}(t) \phi^e_{J_x,k_x}(x) + \sum_{k_x} y^\tau_{J_x,k_x} \phi^\tau_{J_x,k_x}(t) \phi^\tau_{J_x,k_x}(x).$$  (13)

The second term uses the same type of scaling functions (same $L$) for both variables, though this is not necessary and the time- and space-functions are scaled separately by $\tau_{J_x} = \tau / 2^{J_x}$ and $\lambda_{J_x} = \lambda / 2^{J_x}$, respectively. The first term involves edge scaling functions ($e = \text{edge}$) which are correspondingly scaled versions

$$\Phi^e_{J_x,n_x}(t) = \tau^{-1/2}_{J_x} \Phi^e_{n_x} \left( t / \tau_{J_x} \right).$$  (14)

of functions $\Phi^e_{n_x}(t)$ orthogonal both to each other and to all $\phi^e_{k_x}(t)$ for $k_x \geq 1$. These types of edge functions are introduced to terminate the train of positive-$t$ functions at the particular point $t = 0$, in direct analogy to the use of left- and right-hand spatial edge functions to terminate wavelet bases for boundary value problems.\textsuperscript{8,9}
The explicit construction of the edge functions, beyond the scope of the present paper, is
developed from the literature on "intervalizing" compact support wavelets. Essentially, these
functions are constructed as orthonormal linear combinations of \( \phi_0(t) \) and the positive-\( t \) tails of
the \( \phi_{k_j}(t) \), \( k_j = 2^{-L}, 3^{-L}, \ldots, -1 \), that straddle \( t = 0 \). The combined scaling functions \( \Phi_{n_t}(t) \) and
\( \phi_{k_{j,x}}(t) \) provide an exact expansion basis for all polynomials up to order \( L/2-1 \) for all \( t \geq 0 \). They
also allow multiscale decomposition over the same domain, thereby requiring also the
construction of edge wavelets \( \Psi_{n_t}(t) \). Our particular choice of edge functions is detailed for a
multiwavelet basis (multiple father and mother wavelets) elsewhere, and leads near \( t = 0^+ \) to the
behaviors \( \Phi_{n_t}, \Psi_{n_t} \sim t^{n_t} \), which is convenient for satisfaction of the initial value condition.

While all positive \( t \) scaling functions satisfy the usual two-scale relations, the edge functions
satisfy the modified relations,

\[
\Phi_{n_t}(t) = \sum_{n'_t = n_t}^{L/2-1} A_{n_t,n'_t} \Phi_{n'_t}(2t) + \sum_{k_t=1}^{L-1} B_{n_t,k_t} \phi(2t - k_t), \tag{15}
\]

\[
\Psi_{n_t}(t) = \sum_{n'_t = n_t}^{L/2-1} F_{n_t,n'_t} \Phi_{n'_t}(2t) + \sum_{k_t=1}^{L-1} G_{n_t,k_t} \phi(2t - k_t). \tag{16}
\]

Examples are shown in Figs. 2 and 3 for \( L = 8 \). A C++ program named MultiWavePack which is
capable of generating these functions and the associated constant coefficient matrices \( A, B, F, G \)
is available electronically.
It is important to have the ability to calculate the coefficients in the expansion of Eq. (13) directly in the wavelet representation rather than simply transforming the output from other methods of propagation. Such a direct method would fall under the general heading of joint time-frequency methods, and would be directed at the eventual goal of performing computations...
within compressed bases. One time-frequency method for solving the TDSE is already well-known, i.e., expansion of wave functions in complex nonstationary gaussians. In the present approach, however, the parameters to be determined in the wave function are all linear (expansion coefficients). Furthermore, all basis functions are inherently orthogonal and rigorously, rather than exponentially, localized.

B. Short-time propagator

We avoid notational clutter in the rest of this Section by making implicit the indices \( \tau, \lambda, J_t \) and \( J_x \) that do not change (one could alternatively just rescale \( x \) and \( t \)). Since only the \( n_t = 0 \) edge functions are nonzero at \( t = 0 \), we could in principle solve the initial value problem by choosing

\[
y_0(x) = y(x,0) = \sum_{k_x} y^c_{0k_x} \Phi_0(0) \phi_{k_x}(x).
\]

The approximate equality naturally improves with higher resolution of the basis (assuming that \( y_0 \) does not happen to be exactly a linear combination of scaling functions in \( x \) on some scale). However, the \( y^c_{0k_x} \) are actually 2D expansion coefficients of a complex function in the neighborhood of \( t = 0 \), and therefore will have at least small imaginary components. The wave packet has the formal solution

\[
y(x,t) = \hat{K}(t)y_0(x), \quad \hat{K}(t) = \exp(-it\hat{H}/\hbar),
\]

so that the simultaneous projections on the levels \( J_t \) and \( J_x \) are given by

\[
y^e_{n_t k_x} = \mathcal{J} \Phi_{n_t}(t) \phi_{k_x}(x) \hat{K}(t)y_0(x)dx \, dt, \quad \hat{K}(t) = \exp(-it\hat{H}/\hbar), \quad \hat{K}(t) = \exp(-it\hat{H}/\hbar),
\]

\[
y^e_{k_t k_x} = \mathcal{J} \phi_{k_t}(t) \phi_{k_x}(x) \hat{K}(t)y_0(x)dx \, dt.
\]
These are all complex-valued integrals, though the imaginary components of the $n_t = 0$ edge coefficients are small and shrink systematically as $J_t$ grows (i.e., as the edge region shrinks).

We consider the short-time propagator

$$\hat{K}(\Delta t) = \exp(-i\Delta t \hat{H}/\hbar).$$

(22)

Let us apply this to the wave packet as expanded in Eq. (13). Then

$$y(x,t+\Delta t) = \hat{K}(\Delta t)y(x,t)$$

$$= \sum_{n_t=0}^{L/2-1} \sum_{k_x} y_{n_t,k_x}^e \Phi_{n_t}(t) \hat{K}(\Delta t) \phi_{k_x}(x) + \sum_{k_t=1}^{L/2-1} \sum_{k_x} y_{k_t,k_x} \phi_{k_t}(t) \hat{K}(\Delta t) \phi_{k_x}(x).$$

(23)

At the same time, Eq. (13) with $t \to t + \Delta t$ gives

$$y(x,t+\Delta t)$$

$$= \sum_{n_t=0}^{L/2-1} \sum_{k_x} y_{n_t,k_x}^e \Phi_{n_t}(t+\Delta t) \phi_{k_x}(x) + \sum_{k_t=1}^{L/2-1} \sum_{k_x} y_{k_t,k_x} \phi_{k_t}(t+\Delta t) \phi_{k_x}(x).$$

(24)

Equating both right-hand sides and projecting on a particular $\phi_{k_x}(x)$, the orthonormality of the $x$ basis gives

$$\sum_{n_t=0}^{L/2-1} \sum_{k_x} y_{n_t,k_x}^e \Phi_{n_t}(t) K_{k_x,k_x}(\Delta t) + \sum_{k_t=1}^{L/2-1} \sum_{k_x} y_{k_t,k_x} \phi_{k_t}(t) K_{k_x,k_x}(\Delta t)$$

$$= \sum_{n_t=0}^{L/2-1} y_{n_t,k_x}^e \Phi_{n_t}(t+\Delta t) + \sum_{k_t=1}^{L/2-1} y_{k_t,k_x} \phi_{k_t}(t+\Delta t),$$

(25)

where

$$K_{k_x,k_x}(\Delta t) = \int \phi_{k_x}(x) \hat{K}(\Delta t) \phi_{k_x}(x) dx.$$  

(26)

If we now multiply both sides of Eq. (25) by a particular $\phi_{k_t}(t + \Delta t)$ and integrate over $t$, the basis orthonormality gives us
\[ y_{k, k_x} = \sum_{n_i=0}^{L/2-1} \sum_{k_x} y_{n_i, k_x}^e Y_{k, n_i}^e (\Delta t) K_{k_x, k_x} (\Delta t) \]

\[ + \sum_{k_i=1}^{k} \sum_{k_x} y_{k_i, k_x}^e Y_{k, k_i}^e (\Delta t) K_{k_x, k_x} (\Delta t), \]  

where

\[ Y_{k, n_i}^e (\Delta t) = \int \phi_{k, i} (t + \Delta t) \Phi_{n_i} (t) dt, \]  

\[ Y_{k, k_i}^e (\Delta t) = \int \phi_{k, i} (t + \Delta t) \phi_{k_i} (t) dt, \]

are correlation functions between different time basis elements. A simplification arises if we specify that \( \Delta t = \tau_{J_i} \), the basis function spacing. The time analog of Eq. (5) yields that \( \phi_{k, i} (t + \tau_{J_i}) = \phi_{k, i-1} (t) \) and therefore

\[ Y_{k, n_i}^e (\tau_{J_i}) = \int \phi_{k, i} (t + \tau_{J_i}) \Phi_{n_i} (t) dt \]

\[ = \delta_{k_i, 1} \int \phi_0 (t) \Phi_{n_i} (t) dt, \]  

\[ Y_{k, k_i}^e (\tau_{J_i}) = \int \phi_{k, i} (t + \tau_{J_i}) \phi_{k_i} (t) dt \]

\[ = \delta_{k_i, 1, k_i}. \]  

For \( k_i = 1 \), Eq. (27) becomes

\[ y_{1, k_x} = \sum_{k_x} K_{k_x, k_x} (\tau_{J_i}) b_{k_x}^e, \]  

where \( b^e \) is a vector composed of edge coefficients

\[ b_{k_x}^e = \sum_{n_i=0}^{L/2-1} y_{n_i, k_x}^e \int \phi_0 (t) \Phi_{n_i} (t) dt \]  

and the easily-calculated components of \( \phi_0(t) \) in each of the edge functions. For \( k_i' > 1 \), on the other hand, Eq. (27) becomes
which gives the \( k_i \) coefficients directly in terms of those for \( k_i - 1 \). We thus obtain a marching algorithm in the wavelet coefficients and may work entirely in the wavelet representation during propagation. Larger jumps in the marching algorithm are also allowed by using multiples of \( \tau_{J_i} \).

In the time domain, the unitary nature of the evolution ensures that the \( x \)-norm of a wave packet is preserved. This condition becomes quite complicated under direct wavelet transformation, becoming spread over a number of neighboring values of \( k \). Instead, the appropriate analog follows from Eqs. (32)–(34) which involve only two values at a time. Since \( \mathbf{K} \) is the matrix of a unitary operator, we obtain that the \( x \)-wavelet norm is maintained at all steps \( k_i \) and is equal to that of the initial \( \mathbf{b}^e \) vector (not unity). Letting \( \mathbf{y}_{k_i} \) be the vector of coefficients for a single value of \( k \), we have

\[
|\mathbf{y}_{k_i}|^2 = |\mathbf{y}_{k_i-1}|^2 = ... = |\mathbf{b}^e|^2. \tag{35}
\]

This normalization will not be strictly conserved in using finite matrix approximations to the effect of the short-time propagator. It is, however, easy to monitor and serves as a diagnostic to buildup of error.

C. Initiation of the propagation

To initiate the numerical propagation, the edge coefficients can be approximated by truncated local Taylor expansion of the propagator,

\[
y^e_{n_t k_x} = \sum_{p=0}^{p_{\text{max}}} (-i/\hbar)^p m^e_{p,n_t} W_{p k_x}, \tag{36}
\]

where the edge time moments,
\[ m_{p_n_t}^{e} = \int t^{P} \Phi_{n_t}(t) dt, \]  

(37)

can be evaluated exactly by the methods used in Johnson, et al.\textsuperscript{8} If one makes the indices \( \tau \) and \( J \) explicit, these quantities are found to scale as \( \tau_{J}^{P+1/2} \). The spatial integrals are

\[ W_{p} = \frac{1}{p!} \int \phi_{k_x}(x) \hat{H}^{p} y_0(x) dx. \]  

(38)

In a truncated basis using matrix-vector notation, one has

\[ \mathbf{W}_{p} = \mathbf{H}^{p} \cdot \mathbf{y}_0 = \frac{1}{p} \mathbf{H} \cdot \mathbf{W}_{p-1}. \]  

(39)

The components of the vector \( y_0 \) may be calculated by local quadrature based on Lagrange interpolation polynomials. Quadrature on finer levels than \( J_x \), followed by use of the two-scale recursion relations back to \( J_x \), provides exponential reduction of error. Thus, the component integrals can be calculated to any desired accuracy. The remaining errors then lie only in the incompleteness of the scaling function basis, i.e., truncation of the basis both in length and in scale, which provides a finite resolution representation of \( y_0(x) \). For a given scale, one may easily monitor truncation error at the boundaries by focusing on the magnitudes of those coefficients. The errors due to the finite scale behave as \( 2^{-J_x L/2} \), so that \( J_x \) may be chosen such that increase of \( J_x \) makes changes to the total representation error only below a pre-set threshold.

The Hamiltonian matrix \( \mathbf{H} \) is banded with \( L - 2 \) subdiagonals and \( L - 2 \) superdiagonals since the basis functions only share support if \( |k_x - k_x'| \leq L - 2 \). The computational requirements of multiplication of the Hamiltonian matrix into the vector \( \mathbf{y}_0 \) thus scale linearly with the number of basis functions. Calculation of the vectors in Eq. (39) up to \( p = p_{\text{max}} \) generates a Krylov vector subspace in a series of \( O(N) \) matrix-vector multiplications. If the multiresolution representation is used, the simple banded nature of the Hamiltonian matrix is changed to a more complicated
and less sparse structure, but the Beylkin-Coifman-Rokhlin "nonstandard representation" can
still be used to implement each matrix-vector multiply with $N \log_2 N$ scaling complexity.\textsuperscript{19}

Despite the low computational order of Krylov basis calculations, they are well-known to
be subject to roundoff error in finite precision calculations.\textsuperscript{15} There is a problem-dependent limit
to the number of times one can apply the Hamiltonian matrix to a vector before the effects of
roundoff error build up exponentially. Since the time moments scale as $\tau_{n+1/2}$, one can always
choose $J_t$ such that convergence of Eq. (36) occurs before roundoff errors grow.

\textit{D. Wavelet-Chebyshev propagation}

For the later time basis functions, another method must be employed. The structure of
Eqs. (32)-(34) allows one to borrow from pure-time-domain approaches. Of the several different
methods available for evaluating the effects of an exponential matrix on a vector, the Chebyshev
propagation method, introduced into quantum dynamics by Tal-Ezer and Kosloff,\textsuperscript{20} is chosen
here for its inherent stability. For a finite Hamiltonian matrix $H$, the maximum energy $E_{\text{max}}$ and
minimum energy $E_{\text{min}}$ may be calculated or estimated, allowing definition of a modified matrix
with spectral range $[-1, +1]$,

$$
H' = \frac{2H - E_{\text{max}} - E_{\text{min}}}{E_{\text{max}} - E_{\text{min}}}. \quad (40)
$$

One expands the propagator in terms of Chebyshev polynomials of the operator $H'$, with the
action of the propagator on the initial vector $b$ taking the form

$$
K \cdot b \equiv \exp\left(-i \frac{E_{\text{max}} + E_{\text{min}}}{2\hbar} \tau_{J_t}\right) \sum_{n=0}^{n_{\text{max}}} \left(2 - \delta_{n,0}\right) \left(-i\hbar\right)^n J_n \left(\frac{E_{\text{max}} + E_{\text{min}}}{2\hbar} \tau_{J_t}\right) X_n, \quad (41)
$$

where $J_n$ is a Bessel function and the vectors $X_n$ are generated by a series of sparse matrix-vector
multiplications
\[
\begin{align*}
X_0 &= b, \quad X_1 = \mathbf{H}^\prime \cdot X_0, \quad X_{n>1} = 2\mathbf{H}^\prime \cdot X_{n-1} - X_{n-2}. 
\end{align*}
\] (42)

The Bessel function coefficients decay exponentially as the index \( n \) exceeds the argument, with longer times requiring higher \( n_{\text{max}} \) to achieve convergence. Nevertheless, the stable nature of the recursion allows us to reduce the algorithm error toward machine precision, so that residual errors arise only from incompleteness of the dual wavelet expansion basis. The starting vector \( b \) is, in turn, \( b', y_1, y_2, \) etc.

### III. Displaced Harmonic Oscillator

As a first example, consider a harmonic oscillator displaced from equilibrium at \( t = 0 \), a frequently-used model for activity of a vibrational mode in an electronic transition. For \( t < 0 \), the molecule has the vibrational Hamiltonian

\[
\hat{H}_0 = -\frac{\hbar^2}{2m} \frac{d^2}{dx^2} + \frac{k}{2} x^2
\] (43)

with corresponding ground state eigenfunction

\[
y_0(x) = (\alpha/\pi)^{1/4} e^{-\alpha x^2/2}, \quad \alpha = m \omega / \hbar = \sqrt{k m / \hbar}.
\] (44)

At \( t = 0 \), the evolution becomes governed by the displaced potential of the other electronic state (ignoring a constant term representing the adiabatic electronic energy gap)

\[
\hat{H} = -\frac{\hbar^2}{2m} \frac{d^2}{dx^2} + \frac{k}{2} (x - x_0)^2.
\] (45)

The solution to the time-dependent Schrödinger equation which reduces to \( y_0(x) \) at \( t = 0 \) has a known form following the corresponding classical trajectory,

\[
y(x,t) = (\alpha/\pi)^{-1/4} \exp \left[ -\frac{\alpha}{2} (x - x_{cl})^2 + i \alpha x_0 \sin(\omega t)(x - x_{cl}) \right]
\]
\[-i \frac{\alpha}{2} x_0^2 \cos(\omega t) \sin(\omega t) - i \frac{\omega t}{2} \], \quad (46)

\[x_{ct} = x_0 - x_0 \cos(\omega t). \quad (47)\]

Choosing \( h = m = 1 \) and \( x_0 = 1/2 \), the oscillatory motion of the exact wave packet is shown in Fig. 4.

![Figure 4. Absolute value of evolving oscillator wave function, \( |y(x,t)| \).](image)

Scaling function expansions of \( y_0(x) \) as in Eq. (7) are calculated by 1D wavelet quadrature as discussed above. The 2D projection integrals of \( y(x, t) \) as in Eq. (13) are calculated by a product quadrature. Evaluation of the Hamiltonian matrix is trivial for this example since matrix elements of powers of \( x \) or \( d/dx \) can be obtained exactly as the solutions of linear systems of equations.\(^8,21\)

For \( L = 8 \) and \( 10 \) and \( J_x = 3 \) and \( 4 \), Fig. 5 shows the maximum magnitude component of each \( W_{\lambda x}^{\lambda} \) vector generated using the recursion in Eq. (39). The calculations run with 16 and
30 decimals of precision (in Mathematica 5.0) start to disagree after \( p = 11 \) for \( J_x = 3 \) and after \( p = 8 \) for \( J_x = 4 \) (more or less independently of \( L \)). The roundoff error is accentuated by the more rapid \( W^{pJ_x}_p \) oscillations possible in a basis with finer spacings. For a fixed precision, there is thus a practical limit on how high the order \( p \) should be taken. This is consistent with results found for the Short Iterative Lanczos propagation procedure as well.\(^2\)

![Figure 5. Logarithms of the maximum magnitudes (over \( k_x \)) of the coefficient vectors \( W_p \) for different matrix-vector multiply order \( p \), Daubechies family \( L \) and scaling function expansion scale \( j_{x0} \). Numerical precisions: (squares) 16 decimals, (plus signs) 30 decimals.](image)
Figure 6. Maximum error in $y_{n_t,k_x}^e$ (as a function of $n_t$ and $k_x$) calculated from Eq. (36) for (a) $J_t = 6$, (b) $J_t = 7$, (c) $J_t = 8$. The fixed parameters are $\lambda = 1$, $\tau = 2\pi$, $J_x = 3$, while the parameters varying in each graph are $L = 8–16$ and $p_{\text{max}} = 1–15$. The errors are dominated in different regimes by different influences, as discussed in the text.

Using these vectors, the edge coefficients can be calculated as in Eq. (33) and compared to numerically converged values of the expansion coefficients from the exact wave packet using 16 decimal precision. For $\lambda = 1$, $\tau = 2\pi$ and $J_x = 3$ and for a number of cases of $p_{\text{max}}$, $J_t$ and $L$, Fig. 6 plots the logarithm of the worst absolute error as a function of the indices $n_t$ and $k_x$. The
errors are dominated by different causes in different general parameter regions: (1) For small $p_{\text{max}}$, the error is chiefly due to premature truncation of the propagator expansion and decrease exponentially with $p_{\text{max}}$. (2) For intermediate $p_{\text{max}}$, the error is dominated by the representation capabilities of finite bases in $x$ and is flat from one value of $p_{\text{max}}$ to the next. (3) For higher $p_{\text{max}}$, the roundoff error in the $W_{pJ_x}^\lambda$ for $p > \sim 11$ grows and eventually dominates. This switchover is delayed more for higher $J_t$ since the coefficients of the $W_{pJ_x}^\lambda$ decrease exponentially with both $p$ and $J_t$. One can therefore always choose $J_t$ and $p_{\text{max}}$ so as to avoid domination by roundoff error. For $J_x = 3$, $J_t = 7$ or $8$, and $p_{\text{max}} = 11$, these results show us that the choice $L = 16$ gives us quite low errors, though we could reduce these further with even higher $L$ at the expense of increased lengths of the basis functions.

Propagation to $k_t \geq 1$ can be accomplished in $k_t$ sequential unit steps, in a single $k_t$–fold jump, or else in a combination of intermediate-size sub-steps. The global nature of Chebyshev error minimization\(^2\) allows accuracy to be achieved at large $k_n$, though at the price of increased $n_{\text{max}}$. Division into smaller substeps allows smaller maximum orders, though at the price of error accumulation depending on the number of substeps. The optimal choice of substep size may vary according to circumstances. In Fig. 7 the single-step limit is considered in order to assess the best-possible accuracy for 16-decimal precision. For each value of $k_n$, the maximum absolute error in $y_{k_n k_x}$ as a function of $k_x$ is plotted, the reference values being determined by fully converging the 2D projection coefficients of the exact wave packet. For $J_x = 3$ and $J_t = 6–9$, all errors are less than $10^{-12}$, establishing that satisfactory accuracy can indeed be obtained using Daubechies wavelets. Investigation of optimal balancing of efficiency and accuracy is reserved for future investigation.
IV. Multiscale Propagation

Propagation across multiple simultaneous scales in $x$ and $t$ can be investigated by use of the two-scale relations satisfied by each wavelet basis. We consider explicitly the case of single filtering steps to levels $j_x = J_x - 1$ and/or $j_t = J_t - 1$, thus maintaining well-localized basis functions. The scale indices are once again explicit in the following.

Consider first the transformation of the spatial functions,

$$
\begin{align*}
\begin{bmatrix}
\phi_{J_x-1,k_x}^\lambda(x) \\
\psi_{J_x-1,k_x}^\lambda(x)
\end{bmatrix}
&= 2^{-1/2} \sum_{k_x',=0}^{L-1} \left[c_{k_x'}\right] \begin{bmatrix}
\phi_{J_x,2k_x'+k_x}^\lambda(x) \\
\psi_{J_x,2k_x'+k_x}^\lambda(x)
\end{bmatrix}.
\end{align*}
$$

Letting the new basis be the conjoined scaling function and wavelet functions,
we again have an orthogonal basis. The Hamiltonian and short-time propagator matrices in this representation may be obtained similarly by using the two-scale relations on both bra and ket sides. While the overlap matrix remains diagonal, the operator matrices now have nonzero elements in both diagonal and off-diagonal sub-blocks. A general way to handle matrix-vector multiplications for multiresolution representations (any number of scales) is use of the Non-Standard Representations (NSR) of the vectors and operators in intermediate calculations, as described elsewhere by Beylkin, et al.\textsuperscript{19}. For many scales, the multiplication computations approach $N \log_2 N$ scaling rather than $N$, but this is a mild additional computational demand.

For the time functions, the non-edge basis functions transform in the same manner as the spatial ones in Eq. (48). In addition, however, the edge functions near $t = 0$ must be transformed,

$$\left\{ \Phi_{J t, -1, n_t}^r(t) \right\} = 2^{-1/2} \sum_{n_i = n_t}^{L/2 - 1} \left\{ A_{n_t, n_i} \right\} \Phi_{J t, n_t}^r(t) + 2^{-1/2} \sum_{k_t = 1}^{L - 1} \left\{ B_{n_t, k_t} \right\} \phi_{J t, k_t}^r(t).$$

(50)

The new time basis vectors are ordered as

$$u_{J t, -1}(t) = \left[ \ldots, \Phi_{J t, -1, n_t}^r(t), \ldots, \phi_{J t, -1, k_t}^r(t), \ldots, \Psi_{J t, -1, n_t}^r(t), \ldots, \psi_{J t, -1, k_t}^r(t), \ldots \right]^T.$$  

(51)

An analysis may be made of the resulting coupled equations similar to that for the single resolution level $J_t$ given earlier. Either the new or old $x$ basis may be used. Choosing the latter for simplicity, the expansion of the time-dependent wave function takes the form

$$y(x, t) = \sum_{n_t = 0}^{L/2 - 1} \sum_{k_x} y_{J t, -1, n_t, k_x}^{\tau \lambda} \Phi_{J t, -1, n_t}^r(t) \phi_{J t, k_x}^r(x) + \sum_{k_t = 1}^{L - 1} \sum_{k_x} y_{J t, -1, k_t, k_x}^{\tau \lambda} \phi_{J t, -1, k_t}^r(t) \psi_{J t, -1, k_x}^r(x).$$

(52)
One again uses the short time propagator for time $\Delta t$, projects onto different $\phi_{J_x,k_x}^\tau(x)$, then projects onto different $\phi_{J_x-1,k_x}^\tau(t+\Delta t)$ and $\psi_{J_x-1,k_x}^\tau(t+\Delta t)$. The resulting set of equations in this case largely decouples for $\Delta t$ twice as large as before, i.e., the basis function spacing on the $J_x-1$ level. Specifically, we obtain for $k_x = 1$,

$$y_{J_x-1,1}^{\tau} = \sum_{k_x} K_{k_x,k_x} \left( \tau_{J_x-1} \right) b_{J_x-1,1}^{\tau}$$

and for $k_x > 1$,

$$y_{J_x-1,k_x}^{\tau} = \sum_{k_x} K_{k_x,k_x} \left( \tau_{J_x-1} \right) y_{J_x-1,k_x-1}^{\tau}$$

and

$$z_{J_x-1,k_x}^{\tau} = \sum_{k_x} K_{k_x,k_x} \left( \tau_{J_x-1} \right) z_{J_x-1,k_x-1}^{\tau}$$

The b vectors used for $k_x = 1$ are again linear combinations of edge contributions,

$$b_{J_x-1,1}^{\tau} = \sum_{n_{t=1}}^{L/2-1} \left[ \int \phi_{J_x-1,0}^\tau(t) \Phi_{J_x-1,n_{t}}^\tau(t) dt \right] y_{J_x-1,n_{t}}^{\tau}$$

and

$$b_{J_x-1,k_x}^{\tau} = \sum_{n_{t=1}}^{L/2-1} \left[ \int \psi_{J_x-1,0}^\tau(t) \Psi_{J_x-1,n_{t}}^\tau(t) dt \right] z_{J_x-1,n_{t}}^{\tau}$$

The (unanticipated) result is that the scaling function ($y$) and wavelet ($z$) time channels propagate independently after the first step. The only coupling is at the initial step in Eq. (57), where the b vector for the $y$ channel contains a component from the $z$ channel. However, this is due only to the details of the construction of the edge functions.$^{8,17}$ While all the $\Phi_{n_{t}}(t)$ are automatically
made orthogonal to $\psi_0(t)$, the $\Psi_{n_i}(t)$ are not explicitly made orthogonal to $\phi_0(t)$. In principle this latter point could be corrected, in which case the initial coupling would also vanish. In practice, it is very small and easily taken into account, so no further action is required here.

It is in any case a consequence of the subsequent complete decoupling in Eqs. (55) and (56) that the partial norms $\sum_{k_x} |\gamma_{J_{t-1},J_x,k_x}|^2$ and $\sum_{k_x} |\gamma_{J_{t-1},J_x,k_x}|^2$ are independent of $k_x$. By extension of the above arguments, a time wavelet decomposition carried out over more scales will have each subspace evolving independently with constant partial norm. At the same time, these components must all be obtainable from repeated use of two-scale relations on the wave function calculated using only time scaling functions on the single resolution level $J_t$. To verify this, therefore, a single time scale is used for the numerical calculations of the next Section and a multiscale analysis of the results is performed.

V. Eckart Barrier Scattering

We consider the scattering of an initially free particle from an asymmetric Eckart barrier

$$V(x) = \frac{A\xi}{\xi - 1} - \frac{B\xi}{(\xi - 1)^2},$$

(59)

$$\xi = -\frac{A + B}{B - A} \exp\left(\frac{2\pi x}{l}\right).$$

(60)

The free particle wave packet given long ago by Kennard and Darwin can be written in the form,

$$y(x,t) = (2\pi)^{-1/4} \left(\sigma + \frac{iht}{2m\sigma}\right)^{-1/2} \exp \left[-\frac{(x-x_c)^2}{4\sigma^2 + 2iht/m} + \frac{ip_c x}{h} - \frac{ip_c x^2}{2h}\right].$$

(61)
where the classical position is $x_c = x_0 + v_0 t$, the classical momentum is $p_c = m v_0$, and $\sigma$ is an initial width parameter. In Fig. 8 is shown the modulus of the initial wave function ($\sigma = 1/2, m = 16, x_0 = -6, v_0 = 1$) and the asymmetric Eckart barrier ($A = 3, B = 24, l = 2$) toward which it moves. As time evolves, the packet will develop into transmitted and reflected waves, spreading all the while.

Figure 8. Asymmetric Eckart potential (solid line) and incident free particle wave packet (dashed line).

Fixed-grid or fixed-basis dynamics calculations in principle require foreknowledge of the entire region of space visited during the propagation time. There are various means by which adaptive algorithms can be implemented in order to focus only on the regions where the quantum amplitude is important, e.g., propagation of multiple phase-space gaussians, adaptation of Fourier grid methods, dynamically-modified finite element bases, moving grids, etc. For spatial coordinates, wavelet expansion allows use of a fixed grid with different regions emphasized by the magnitude of the local basis functions. Bacry, et al.,25 developed an adaptive algorithm for
this scenario based on the ease with which individual members of an orthonormal basis may be added or deleted (for example, there is no regridding as can be required in finite element algorithms). Here the method is extended and applied to the space-and-time wavelet expansion of the wave function for the Eckart barrier as a prototypical scattering problem.

Since one of our primary goals is to maintain numerical accuracy while working strictly within the wavelet representation, we maintain buffer zones of size $n_{buf}$ beyond the values of $k_x$ on left and right hand sides where the coefficient magnitudes cross a specified numerical threshold. As the value of $k_t$ increases, $k_x$ basis functions are added to or deleted from the outside in order to maintain constant buffer size past the threshold crossing. When points are added, the corresponding coefficients are initially taken as zero for simplicity, although they could in principle be approximated by extrapolation. The important point is that the edge errors are kept on the order of the threshold, and may always be improved by tightening of the latter.

We use $\lambda = \tau = 1$ and $J_x = J_t = 4$. For the $x$–basis, the equivalent scaling functions and wavelets of the $j_x = 3$ level are used as described in the last section, and the square root of the sums of the scaling function and wavelet coefficients for a given $k_x$ are used in comparisons with a threshold value taken as $3 \times 10^{-13}$. (Propagations using $j_t = 3$ have also been performed, but we focus on $j_t = J_t = 4$ here.) The buffer size is taken as $n_{buf} = 16$ in these calculations. The resulting $y(x,t)$ is shown in Fig. 9. For space-time regions where the magnitude is greater than 1% of maximum, both the magnitude and phase of $y(x,t)$ are shown. (The Eckart phase here is associated with wavefront trajectories in the hydrodynamical picture of quantum mechanics, see Fig. 3 of Bittner and Wyatt.26) The flat gray regions surrounding the incident, reflected and transmitted wave packets represent the areas where the magnitude is still greater than the threshold used for adaptive basis selection.
Figure 9. Absolute value of $y(x, t)$ calculated for free particle scattering by an asymmetric Eckart barrier. The phase of the wave function is mapped on the surface in grayscale, while the underlying flat gray region corresponds to all coefficients above threshold in the adaptive selection of space–time basis elements.

The wavelet decomposition of the coefficients is performed for both coordinates to scales $j_{t_0} = j_{x_0} = 1$, yielding four distinct subspaces $s_1, w_1, w_2$ and $w_3$ for each. The multiresolution filtering for $x$ is not required to address mixing or non-mixing between different time-scale subspaces, but provides additional insight. For each joint time and space subspace, partial norms are formed by summing the absolute squares of the coefficients over $k_x$. The results starting from
$k_t = 1$ (i.e., neglecting the edge coefficients) are shown in Fig. 10. Each subplot corresponds to one $t$ subspace (the first symbol $s_1$, etc.) and the full range of $x$ subspaces (the second symbol). It is seen that every partial norm is individually conserved during free propagation, but mixes strongly with the others within each plot during the scattering event. Nevertheless, the sums of the partial norms within a plot are completely conserved for all values of $k_t$. This provides verification that the different time wavelet subspaces do not mix, i.e., the wavelet separation into different time scales is determined only by the initial stages of the propagation.

The largest "populations" are found in the $j_x = 1$ $t$–wavelet subspace $w_1(t)$, Fig. 10(b). Their dominance must reflect only the velocity and phase behavior of the initial wave packet, independent of the subsequent scattering event. The $j_x = 2$ $x$–wavelet components in $w_2(x)$ are initially the most significant, but shrink significantly while those in both $s_1(x)$ and $w_1(x)$ grow during the scattering. This indicates the importance of slightly larger length scales induced by the Eckart potential. Subsequently, free evolution is gained as the reflected and transmitted packets exit the scattering region, with $w_2(x)$ smaller than and $w_1(x)$ larger than before. This same basic pattern is obtained in all of the plots of Fig. 10, though the mixing of functions on different $x$–scales is clearly dependent on which $t$–scale is examined. Differences in the left-vs-right positions of the peaks between traces and subplots in coefficient space primarily stem from the details of the shapes and extents of the basis functions.
Figure 10. Partial norms of Eckart barrier wave packet in different scaling function ($s$) and wavelet ($w$) subspaces $w_{j_1t} (t) w_{j_1x} (x)$, etc., past the $t = 0$ edge functions. Figures are grouped according to time subspaces: (a) $s_1$, (b) $w_1$, (c) $w_2$, (d) $w_3$.

The conclusions of Sect. IV are thereby verified. The full wave packet can be decomposed into different time-scale components via orthogonal wavelet filtering, and each component propagates independently. The effects of the scattering potential are restricted to mixing among the spatial components for each time subspace, and therefore the proper analysis of such results is to compare how spatial mixing varies with different time scales. Analogous derivations will also hold for other choices of wavelets, potentials and spatial dimensions, so the present results are of a general nature. Accordingly, it is to be expected that a more general derivation will eventually be found. The explicit analysis in terms of localized time functions appears to be fairly new, the only other example of which we are aware being a finite element analysis by Jensen and Linderberg.27

On the computational side, as discussed earlier, it is of long-term interest to examine compressibility of the wave function at run-time. Efforts were made to automatically divide the wave packet into reflected and transmitted components via simple monitoring of extended space-time wavelet coefficients. While the wave packet bifurcation is evident visually in Fig. 9, the fact is that components whose coefficients are very small but still much larger than the adaptive threshold linger in the neighborhood of the barrier (see, for example, Ryabov and Moiseyev28 for a discussion of resonances in the symmetric and asymmetric Eckart barrier systems). Using a less accurate outside threshold, on the other hand, produces commensurate errors in the inner region. While the wavelet basis is in principle easily able to handle wave packet bifurcation,
further work is needed to determine the best way to implement this in the context of multiresolution representations. Other issues for examination in the future include Hamiltonians with explicit time dependence, for which the exact separation of time scales is not expected, and methods for reduction of large phase components, for which phase space wavelets\textsuperscript{12} offer an alternative basis possessing natural advantages. Finally, the combined use of edge wavelets for the initial value specification with spatial edge wavelets for boundary value specifications should provide the capability to perform direct wave packet propagations in curvilinear coordinate problems.

VI. Summary

We have demonstrated the use of orthogonal compact-support wavelets as localized bases in space and time for solutions of the TDSE. The initial value specification has been allowed by use of special edge wavelets for the earliest times. Direct marching in the wavelet representation for later times was shown to be possible with systematic control of accuracy afforded by a Chebyshev implementation. A primitive adaptive algorithm was added which takes advantage of the easy addition and deletion of wavelet basis functions during dynamic evolution. Last, but not least, it was determined that the multiscale time components propagate independently of each other and that their total spatial norms are conserved quantities.

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