Comparison of Numerical Approaches to the Time-Dependent Schrödinger Solutions in One Dimension

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We examine the performance of various time propagation schemes using a one-dimensional model of the hydrogen atom. In this model, the exact coulomb potential is replaced by a soft-core interaction. The model has been shown to be a reasonable representation of what occurs in the fully three-dimensional hydrogen atom. Our results show that while many numerically simple (low order) propagation schemes work, they often require quite small time steps. Comparing them against more accurate methods, which may require more work per time step but allow much larger time steps, can be illuminating. We show that at least in this problem, the compute time for a number of the more accurate methods is actually less than lower order schemes. Finally, we make some remarks on what to expect in generalizing our findings to more than one dimension.

I. INTRODUCTION

Numerical solutions to the time-dependent Schrödinger equation (TDSE) for a Hamiltonian that explicitly depends on time requires some approximation to the time-ordered exponential. The usual procedure is to:

1. Make a short-time approximation to the exponential by essentially ignoring time-ordering.
2. Introduce some discretization in space via a finite-difference or basis set expansion.
3. Develop a time evolution scheme for what is now a large matrix exponential.

Converging the solution to sufficient accuracy is achieved by increasing the number of spatial basis functions - or grid points- and decreasing time-step size so that the propagated wavefunction is essentially unchanged. Short-time propagation is the action of the exponentiated matrix on a known vector. Here we investigate a number of methods that perform this propagation with the goal of quantifying their accuracy and computational efficiency. The results indicate that higher-order methods often perform better than lower-order approaches if both high accuracy and efficiency are demanded. We also give some estimates of what to expect when the methods are generalized to more than one spatial dimension.

This paper is organized as follows. In section II we discuss the general properties of a quantum mechanical Hamiltonian that involves the interaction of an electromagnetic field with an atom and emphasize on the question of gauges. Readers familiar with the details of Maxwell’s equations and gauges can skip this section but it has been included for completeness. In section III we explain the 1-D model of hydrogen atom that we used as a test case for propagating the TDSE. Here, we also detail the length and velocity gauge forms of the model; discuss its discretization scheme; and mention the computational parameters used throughout the paper. In section IV we examine the computational methods used for propagating the solutions to the TDSE. In section V the methods are compared to one another in terms of their relative performance. The comparisons and results are shown for both bound-state excitations as well as above threshold ionization (ATI). In section VI we present some general remarks on the extension of the methods to higher dimensions and more general discretization. Finally, we present our conclusions in section VII.

II. GENERAL HAMILTONIAN

The Hamiltonian (atomic units \( e = \hbar = m = 1 \) are used throughout the article) of a particle under the influence of an external classical electromagnetic field (EM) is

\[ H = \frac{1}{2} \left[ \mathbf{p} - \frac{1}{c} \mathbf{A}(x, t) \right]^2 + \phi(x, t) + \mathbf{V}(x), \quad (1) \]

where \( \mathbf{A} \) and \( \phi \) are respectively the vector and scalar potential, \( \mathbf{p} = i \nabla \) is the momentum operator and \( \mathbf{V}(x) \) is a local potential. The magnetic and electric fields in Maxwell’s equations are related to the scalar and vector potentials by,

\[ B = \nabla \times \mathbf{A}, \quad (2a) \]

\[ E = -\nabla \phi - \frac{1}{c} \frac{\partial \mathbf{A}}{\partial t}. \quad (2b) \]
It can be demonstrated that $B$ and $E$ remain unchanged in Eqs. [2] under the transformation

$$A \rightarrow A' = A + \nabla \chi, \quad (3a)$$

$$\phi \rightarrow \phi' = \phi - \frac{1}{c} \frac{\partial \chi}{\partial t}, \quad (3b)$$

where $\chi$ is any twice-differentiable function. The equation satisfied by $\chi$ depends on the condition imposed on $A$. In chemistry and atomic and molecular physics, the most commonly used gauge is the Coulomb gauge, where we impose the condition,

$$\nabla \cdot A = 0. \quad (4)$$

If we impose the additional condition that,

$$\phi = 0, \quad (5)$$

it then follows that,

$$E = -\frac{1}{c} \frac{\partial A}{\partial t}, \quad (6)$$

$$H = \frac{p^2}{2} - \frac{1}{c} A \cdot p + \frac{1}{2c^2} A^2 + V(x). \quad (7)$$

This is commonly called the velocity gauge, which is a consequence of imposing $\phi = 0$ in addition to Eq. [2]. Alternatively, we may choose,

$$A = 0, \quad (8a)$$

$$\phi = -x \cdot E, \quad (8b)$$

which still satisfies Eq. [4], leading to,

$$E = -\nabla \phi, \quad (9a)$$

$$H = \frac{p^2}{2} - E \cdot x + V(x). \quad (9b)$$

The last set of equations is commonly called the length gauge for the Schrödinger equation. We can freely transform from length to velocity gauge by choosing

$$\chi = x \cdot A', \quad (10)$$

and from velocity gauge to length gauge via

$$\chi = -x \cdot A. \quad (11)$$

In principle, for an exact solution, the two representations are equivalent. Numerically, this need not be the case. In our numerical studies, we have confirmed that our results for the two gauges agree, giving us added confidence in the data presented for the different methods of solution and eliminating that from clouding our conclusions.

For low intensity light (≤ $10^{15}$ W/cm²), $A^2$ in Eq. [7] may be neglected. Care is required to ensure this approximation is valid when dealing with very high intensity laser fields. In the dipole approximation the size of the atomic system is much smaller than the wavelength of the radiation, so one may replace $e^{ik_0 x - i\omega t}$ by $e^{-i\omega t}$ (see, for example, Peskin and Moiseyev [2]).

### III. THE ONE DIMENSIONAL HYDROGEN ATOM

#### A. Outline in Length Gauge

The Hamiltonian for the one-dimensional model of the hydrogen atom in an external EM field is given in the length form as,

$$H(t) = H_0 - x \cdot E(t) \sin(\omega t + \phi), \quad (12)$$

with the time-independent part

$$H_0 = -\frac{1}{2} \frac{d^2}{dx^2} + V(x), \quad (13a)$$

$$V(x) = -\frac{1}{\sqrt{1 + x^2}}. \quad (13b)$$

The soft-core potential in Eq. (13b) is identical to that used by the authors of Refs [3, 4]. The virtue of this rather simple model is that it preserves the Rydberg spectrum of the hydrogen atom but eliminates the singularity of the potential at $x = 0$. Its failing is that it does not contain the angular momentum coupling inherent in the 3-D hydrogen atom and as a consequence one should be cautious in drawing certain conclusions from the calculations. The bound-state energies of this model do not agree with the actual 3-D hydrogen’s, but the 1-D model has been used in applications ranging from high magnetic field interaction with hydrogen to semiconductor quantum wires, carbon nanotubes, and polymers [5].

In what follows we consider laser field interaction terms with both a smooth pulse envelope,

$$E(t) = \begin{cases} 
E_0 \sin^2 \left[\frac{\pi t}{T}\right], & 0 \leq t \leq T \\
0 & \text{otherwise},
\end{cases} \quad (14)$$

as well as a square pulse,

$$E(t) = \begin{cases} 
E_0, & 0 \leq t \leq T \\
0 & \text{otherwise},
\end{cases} \quad (15)$$

over a time interval $T$. There is a detailed discussion about the use of each of these laser pulse types in Javanainen et al. [3]. In order to study the convergence of the excitation of low-lying bound states with respect to the size of the time-step for various methods, we employed a smooth pulse. For the above threshold ionization (ATI) continuous spectrum, we used a square pulse to compare with Javanainen et al. [3]. We emphasize that our findings concerning the relative timings of the methods and their convergence with step size are independent of the type of pulse employed. This will be discussed in more detail in section [V].
B. The Velocity Gauge

Javanainen et al. suggest that the dipole length form of the time-dependent interaction could cause problems at large distances. In reality, the interaction is turned off at \( T \), so there is no formal problem. However, if the computational region is large, as it may need to be in order to study ionization, there could be numerical issues.

To ensure the accuracy of the numerical results, we have performed most of the calculations in both length and velocity gauge; we have found no differences in the converged results. Of course, in the length gauge, the interaction with the external EM field is local, producing a Hamiltonian which is a real operator and that certainly makes the calculation more straightforward. In the velocity gauge, the Hamiltonian is Hermitian and the interaction is non-local. In more realistic problems, the choice of gauge can make significant differences depending on the wavelength of the radiation. This is a consequence of the presence of high angular momenta in the coupling of the radiation field to the electrons. A detailed discussion of this remains outside the scope of the present study, but has been commented on in the literature by others, but was not pursued in this paper.

Finally, it is worthwhile pointing out that the Numerov method, which also produces tridiagonal matrices, is two orders of magnitude more accurate than the simple three point finite difference method. This has been noted in the literature by others, but was not pursued in this paper.

C. Discretization

To solve Eq. (12), we divide space into a set of \((2N+1)\) equidistant points \((x_n = n \delta x)\) centered at the origin and apply the lowest order central difference formula to discretize the first and second derivatives of the Hamiltonian. In the length gauge this yields,

\[
(H\psi)_n = -\frac{1}{2(\delta x)^2}(\psi_{n+1} - 2\psi_n + \psi_{n-1}) + V(x_n)\psi_n
- x_n E(t) \sin(\omega t + \phi)\psi_n,
\]

and in the velocity gauge,

\[
(H\psi)_n = -\frac{1}{2(\delta x)^2}(\psi_{n+1} - 2\psi_n + \psi_{n-1}) + V(x_n)\psi_n
+ \frac{iA(t)}{2c\delta x}(\psi_{n+1} - \psi_{n-1}).
\]

In both gauges the discretization of the Hamiltonian gives rise to a tridiagonal matrix. The length gauge produces a real symmetric-tridiagonal matrix, the velocity-gauge a Hermitian-tridiagonal matrix. The boundary conditions employed set the wavefunction to zero outside the computational region. Consequently it is important to ensure that this region is sufficiently large enough to be able to extract the probabilities of excitation and ionization without any spurious reflections that would compromise the numerical results. We did not resort to any more sophisticated methods such as absorbing potentials or complex rotation although that would be interesting as a separate study.

D. Computational Implementation and Parameters

For the purposes of this study we began by examining the convergence of the probabilities of excitation to a few of the lower lying bound states. It is sufficient in this case to employ a box of \( \approx 200 \) a.u. on each side of the origin. However, to obtain converged results for the low lying continuum states, a box of \( \approx 800 \) a.u. on either side of the origin is necessary. A spatial step of \( \delta x = 0.1 \) was employed, resulting in matrix sizes of \( n = 4001 \) to \( n = 16001 \). Laser interaction times varied for the tests but on average were about 1200 a.u.. The time-steps were varied from 1 to 0.001 a.u., according to the propagation method used. We chose a laser amplitude of \( E_0 = 0.1 \) and an angular frequency \( \omega = 0.148 \).

These choices of parameters enabled us to perform all of the computational experiments on a desktop PC with Intel 3.4GHz Xeon(R) CPU in a practical amount of time. The code was written in FORTRAN and compiled using the Intel-Fortran compiler with -Ofast optimization. The Intel MKL libraries were used in scalar mode to...
IV. COMPUTATIONAL METHODS

A. Crank-Nicolson

The Crank-Nicolson (CN) method [11] is an implicit propagation/diffusion numerical method as the calculation of the solution at Nth time-step requires the solution of a set of linear algebraic equations. Employing the second-order accurate version of the CN approximation in time, yields,

\[ e^{-iH(x,t)\delta t} = e^{-iH(x,t)\delta t/2} e^{iH(x,t)\delta t/2} \approx \frac{1 - i\delta t/2H(x,t)}{1 + i\delta t/2H(x,t)}, \]  

(23)

\[ [1 + i\frac{\delta t}{2} H(x,t)]\psi(x,t+\delta t) = [1 - i\frac{\delta t}{2} H(x,t)]\psi(x,t). \]  

(24)

If we insert the finite-difference formula discussed above, we obtain a tridiagonal set of linear equations. These equations may be solved by a method which scales linearly with the number of unknowns. In higher dimensions, the coupling destroys the tridiagonal nature of the one-dimensional CN method but as we shall show later, it is still possible to derive second-order methods which scale reasonably well with matrix size. We also note, that there are a number of methods which can solve a tridiagonal system faster than \(O(n)\) in parallel. The reader is referred to Refs. [12][14] for more details.

B. Split Operator

Since the Hamiltonian can be split into a sum of time-independent and time-dependent parts, it is natural to consider propagation methods employing operator splitting. A second-order accurate split operator (SO) method for the time evolution of the wavefunction is [15][22]

\[ \psi(x,t+\delta t) = e^{-i\delta t V(x,t)/2} e^{-i\delta t H(x,t)/2} \psi(x,t). \]  

(25)

Higher-order approximations could be derived by the method of fractal decomposition [18]. The 4th-order split operator, for example, is the more complicated operator

\[ \hat{U}(t+\delta t; t) = \exp[i\delta t V(x,t+\delta t)] \exp[-i\delta t H_0(x)] \exp[i\delta t V(x,t+\delta t)], \]

(26)

where \(S = \frac{1}{4-\sqrt{4}}\). However, while the 4th-order splitting affords larger time-steps to be taken, the number of operations has increased five-fold as compared to the 2nd-order splitting. The trade-off between larger time-steps and larger number of operations did not lead to a performance advantage in our model problem.

A significant advantage of the SO approach is that it is an explicit time propagation method and does not require the solution of a set of linear algebraic equations to find the wavefunction at the next step. As is the case with CN, SO is more efficient in the length gauge, given that the matrix \(V(x,t)\) is a local, diagonal matrix in configuration space. The remaining challenge is how to treat the exponential operator involving \(H_0\). This is the computational bottleneck because a full diagonalization of the \(H_0\) can be quite expensive if the matrix is large. Even though our model enables the eigenvalues to be found quite efficiently, the eigenvectors are another matter and they are required to represent the exponential. The eigenvector matrix is not only dense but is, in principle, required for all the eigenvalues. A viable alternative, is to treat the exponentiation of the \(H_0\) with either Crank-Nicolson or Lanczos iteration, which is described in the next section.

A third approach, called the real space split operator method [16], has been used to bypass the use of the fast Fourier transform of the kinetic energy. Since the Hamiltonian in question is tridiagonal, it can be written as a sum of overlapping “even” and “odd” \(2 \times 2\) matrices.

\[
\begin{pmatrix}
  a_1 & b_1 \\
  b_1 & a_2 & b_2 \\
  b_2 & a_3 & b_3 & b_3 \\
  b_3 & a_4 & b_4 & b_4 & b_4 \\
  b_4 & a_5 & b_5 & b_5 & b_5 & a_5
\end{pmatrix}
= 
\begin{pmatrix}
  a_1 & b_1 \\
  b_1 & a_2/2 & b_2 \\
  a_2/2 & b_3 & a_3/2 & b_3 \\
  b_3 & a_4/2 & b_4 & b_4 \\
  a_4/2 & b_5 & b_5 & a_5
\end{pmatrix}
+ 
\begin{pmatrix}
  0 & 0 \\
  0 & a_2/2 & b_2 \\
  0 & a_2/2 & a_3/2 & b_3 \\
  0 & a_3/2 & b_4 & b_4 \\
  0 & a_4/2 & b_5 & b_5
\end{pmatrix}.
\]

2 Hatano and Suzuki [23] have made a typo error in their equation (63); the factor of the middle term’s exponentials remains \((1 - 4S_2)\), not \(S_2\).
If we now insert this into the exponential and employ a 2\textsuperscript{nd} order accurate splitting of the exponential we obtain,

$$\exp[-i\delta H_0] = \exp\left[\frac{i\delta}{2} H_0^{\text{odd}}\right] \exp[-i\delta H_0^{\text{even}}] \exp\left[\frac{i\delta}{2} H_0^{\text{odd}}\right],$$  \hspace{1cm} (27)

Since $H_0^{\text{even}}$ and $H_0^{\text{odd}}$ each consist of non-overlapping $2 \times 2$ matrices, it is easy to diagonalize them and to then propagate the solution using operations that only require the successive action of a set of $2 \times 2$ matrices on a vector. By employing this even-odd splitting approach we are neglecting commutators between the the discretized kinetic energy operator. This approach only becomes reasonable for small enough time-steps so as to provide an accurate representation of the physical process under consideration. A time comparison for the various splitting methods will be shown in section \( \text{V} \).

C. Lanczos Iteration

The Lanczos Iteration method was initially developed to find the smallest and largest eigenpairs of a large, sparse, $n \times n$ symmetric matrix \cite{24,26}. Lanczos-type solvers have also been used to deal with non-symmetric matrices (see, for example, Axelsson \cite{27} and references therein). It has been demonstrated that these eigenvalues converge in far fewer than $n$ steps. A significant advantage of the method is that the major computational step involves the multiplication of the matrix on a known vector plus a few scalar products. For a sparse matrix, this can be done in $O(M)$ multiplications, where $M$ is the number of non-zero matrix elements.

In effect, the Lanczos method may be viewed as reducing the large, $H_{n \times n}$ matrix to a smaller, $H_{m \times m}$ tridiagonal matrix, where ideally $m \ll n$ for the eigenpairs of interest. If the iteration continues until $m = n$, the eigenvalues and eigenvectors of $H^{(2)}$ would be identical to those of the $H_{n \times n}$. It should be noted that a naive implementation of the Lanczos iteration can lead to linear dependence which has a number of undesired side effects that have been discussed in the literature \cite{26,27}. Ways to circumvent the linear dependence are discussed in the literature and often require additional and expensive mathematical operations.

The transformation between the two representations is given by:

$$H^{(2)} = Q^T H Q,$$  \hspace{1cm} (28)

where $Q_{m \times n} = [|q_1\rangle \ |q_2\rangle \ldots \ |q_m\rangle]$ are the Lanczos vectors at the $m^{th}$ step of the process. One can view these vectors as linear combinations of the so-called Krylov subspace vectors.

$$K(H, q, m) = \text{span}\{|q_1\rangle, H|q_1\rangle, H^2|q_2\rangle, \ldots, H^{m-1}|q_1\rangle\} = \text{span}\{|q_1\rangle, |q_2\rangle, |q_3\rangle, \ldots, |q_m\rangle\}. $$ \hspace{1cm} (29)

where,

$$\beta_{k+1}|q_{k+1}\rangle = (H - \alpha_k I)|q_k\rangle - \beta_{k-1}|q_{k-1}\rangle.$$  \hspace{1cm} (30)

The vectors in Eq.(30) form an orthonormal set. The recursion relation may be started with any vector and continued until the desired eigenvalues are found to sufficient accuracy.

The application of the Lanczos method to time propagation is not directly related to the question of determining the eigenvalues \cite{28,29}. The process may be stated as follows; let the first Lanczos vector, $|q_1\rangle = |\psi(x,t)\rangle$. How can we determine a small set of additional vectors, $|q_2\rangle \ldots \ |q_m\rangle$, which effectively span the new subspace defined by $|\psi(x,t + \delta t)\rangle$ and provide a representation of the exponential function over the time-step? We assume the time-step is small enough that we can approximate the interaction of the electrons with the field by using the field’s value at the midpoint of the time-step. Then the time evolution operator, $\hat{U}$, may be approximated as

$$\hat{U}(t + \delta t|t) = \exp(-i\hat{H}(x,t + \frac{\delta t}{2} \delta t).$$ \hspace{1cm} (31)

The size of the the Krylov subspace matrix, $Q$, is determined by the sufficiently accurate approximation of $H$ by the tridiagonal matrix

$$H^{(2)} = \begin{bmatrix} \alpha_1 & \beta_2 & \ldots & \\ \beta_2 & \alpha_2 & \ldots & \\ & \ldots & \ldots & \ldots \\ & & \beta_m & \alpha_m \end{bmatrix}.$$ \hspace{1cm} (32)

The most computationally demanding step in the process involves the multiplication of $H$ onto the previously computed Lanczos vector. In our case of the 1-D propagation, this comes down to multiplying a tridiagonal matrix by a vector which is an $O(3m)$ process.

To compute the wavefunction at the next time using the Lanczos iterations we replace the time evolution operator of $H$ by its approximation in the Krylov subspace,

$$\hat{U}^L = \exp(-i \hat{H}^{(2)} \delta t).$$ \hspace{1cm} (33)

Therefore, $\hat{U}^L$ is restricted to the (hopefully) smaller dimensional Krylov subspace and is evaluated by direct diagonalization of the tridiagonal matrix produced by transforming $H$ to the Lanczos basis,

$$\hat{U}^L = \sum_i |\lambda_i\rangle \exp(-i \lambda_i \delta t) \langle \lambda_i|,$$  \hspace{1cm} (34)

where $|\lambda_i\rangle$ denotes the eigenvector of $H^{(2)}$ with eigenvalue $\lambda_i$. The propagated wave is then

$$|\Psi(x,t + \delta t)\rangle = \sum_i |\lambda_i\rangle \exp(-i \lambda_i \delta t) \langle \lambda_i|\Psi(x,t)\rangle.$$  \hspace{1cm} (35)
In matrix notation this is equivalent to
\[
\Psi(t + \delta t) = QA^T \exp(-i \delta t \text{ diag}[\lambda_1 \ldots \lambda_m]) A Q^T \Psi(t),
\]
where \( A = [ |\lambda_1 \rangle |\lambda_2 \rangle \ldots |\lambda_m \rangle ] \).

1. Algorithm

| Algorithm for Lanczos Propagation |
|-----------------------------------|
| **initialize:** \( |q_0 \rangle = 0; \quad \beta_1 = 0; \quad k = 1 \) |
| \( n_{\text{max}} \) = Lanczos maximum vector number/iteration |
| \( \epsilon \) = error threshold |
| \( N = \sqrt{\langle \Psi(x,t) | \Psi(x,t) \rangle} \) |
| \( |q_1 \rangle = |\Psi(x,t) \rangle / N \) |
| do while \( k < n_{\text{max}} \) |
| \( \alpha_k = \langle q_k | H |q_k \rangle \) |
| Calculate eigenvalues (\( \lambda_i \)) and eigenvectors (\( |\lambda_i \rangle \)) of tridiagonal matrix H\(^{(A)}\) (\( i \in \{1, \ldots, k\} \)) |
| \( |\Psi_k' \rangle = \sum_i \langle \lambda_i | \exp(-i \lambda_i \delta t) \langle \lambda_i | q_j \rangle |\Psi(x,t) \rangle \) |
| if \( (|| |\Psi_k \rangle - |\Psi_{k-1} \rangle ||_2 < \epsilon) \) |
| converged = True |
| break |
| \( |r_k \rangle = (H - \alpha_k I) |q_k \rangle - \beta_{k-1} |q_{k-1} \rangle \) |
| \( \beta_{k+1} = \sqrt{\langle r_k | r_k \rangle} \) |
| \( |q_{k+1} \rangle = |r_k \rangle / \beta_{k+1} \) |
| \( k = k + 1 \) |
| end 1st while loop |
| while .not. converged |
| \( \delta t = \delta t / 2 \) |
| recalculate \( |\Psi_k' \rangle \) and \( |\Psi_{k-1} \rangle \) |
| end 2nd while loop |
| \( \Psi(x, t + \delta t) = \sum_j \langle q_j | \Psi_k' \rangle \) |

**TABLE I.**

In our application of the Lanczos method, we adjusted the size of the time-step during the propagation to yield accurate results using the smallest number Krylov space vectors. For simplicity, this variable time-step approach is still referred to as Lanczos propagation in this paper. A brief description is as follows; we start the process by setting

\[
|q_1 \rangle = N |\Psi(x,t) \rangle ,
\]
\[
N = \sqrt{\langle \Psi(x,t) | \Psi(x,t) \rangle} .
\]

This is a normalized version of the wavefunction from the previous time-step. The Lanczos vectors are generated from the three-term recursion relationship,

\[
|r_k \rangle = \beta_{k+1} |q_{k+1} \rangle = \langle H - \alpha_k |q_k \rangle - \beta_{k-1} |q_{k-1} \rangle .
\]

Due to the orthonormality of the \( |q_k \rangle \)'s,

\[
\alpha_k = \langle q_k | H |q_k \rangle .
\]

Having found \( \alpha_k \), from Eq.\((40)\) we can find \( \beta_k \) from

\[
\beta_{k+1} = \sqrt{\langle r_k | r_k \rangle} ,
\]
\[
|q_{k+1} \rangle = |r_k \rangle / \beta_{k+1} .
\]

In setting the maximum number of Lanczos vectors to a modest value, for example, 20 vectors, the iteration process will or will not converge for the selected time-step. If it converges, we continue to the next step. If it does not converge, the time-step is halved and the convergence is again tested. This process is repeated until the propagated vector is sufficiently accurate to continue. Note that one does not need to compute any new Lanczos vectors for the reduced time-step. All that needs to be done is to re-evaluate the exponential on the vector, a numerically cheap operation. Pseudocode of the algorithm we implemented is given in Table I.

As with all applications of the Lanczos method, one needs to pay attention to the possible loss of orthonormality during the iteration process. This loss of linear independence can lead to disastrous results. We found that if the initial time-step is taken too small, the first and second Lanczos vectors are not sufficiently linearly independent to produce stable results. By re-orthogonalizing those two vectors, the rest of the process proceeds smoothly, without any need for additional Gram-Schmidt steps.

The algorithm as described here is robust and self-correcting. However, a slightly faster algorithm could be written for a fixed number of Lanczos vectors associated with a time scale, \( \delta t \), known in advance to converge. This would not only eliminate the need to check convergence at every step but would also reduce the need to tridiagonalize the Lanczos matrix multiple times. Such a scheme would necessarily need to be tested for convergence by an a priori adjustment of \( \delta t \); the same statement can be made about the operator splitting and Crank-Nicolson methods.

2. Split-Operator+Lanczos

It is possible to combine the splitting techniques of subsection IV B with the time-adjusting Lanczos scheme described above. The Hamiltonian in the length-gauge in our 1-D model problem lends itself to such a splitting.

By splitting the time evolution operator in the length-gauge into time independent and time-dependent exponentials, one produces, in second order, an expression consisting of a single, time independent, tridiagonal matrix sandwiched between two diagonal time-dependent exponentials. The time independent, tridiagonal matrix is treated via the Lanczos propagator and its diagonal scaling used to compute the required vector. We call this the Split-Operator+Lanczos method.

Even with this fact, the number of Lanczos vectors needed for convergence at a given time-step may change...
due to the diagonal, time-dependent scaling. The consequence is still a time-adjusting scheme. The time-step at each time iteration, for a fixed maximum number of Lanczos vectors, has to be adjusted to ensure convergence. As mentioned before, one could employ a fixed number of Lanczos vectors for all time-steps, if that could be determined prior to the propagation. Finally, higher order splittings could be used to allow larger time-steps, but the number of operations will also increase, which could result in overall longer run-times for the same accuracy.

D. Chebychev propagator

The final method we examined was the so-called Chebychev propagator [30, 31]. This approach has been used in the chemical physics community to treat a number of problems but, to our knowledge, has not seen widespread use in the atomic and molecular physics literature. The basic idea is to expand the short time evolution operator $\hat{U} = \exp(-i\hat{H}dt)$ in terms of a complex version of the Chebychev polynomials of the first kind. Since the Chebychev polynomials are defined on the interval $[-1, 1]$, this requires the Hamiltonian to have eigenvalues inside the unit circle. This can be accomplished if we can estimate the spectral range, $\Delta = E_{\text{max}} - E_{\text{min}}$ of $\hat{H}$, where $E_{\text{max}}$ and $E_{\text{min}}$ are respectively, the largest and smallest eigenvalues of $\hat{H}$. Then using,

$$\hat{H}_{\text{norm}} = 2\frac{\hat{H} - E_{\text{min}}I}{\Delta} - I,$$

the expansion coefficients can be shown [30] to be

$$a_k = (2 - \delta_{0k}) \exp\left(-i\frac{\Delta}{2} + E_{\text{min}}\right) dt |J_k\left(\frac{\Delta}{2} dt\right).$$

where $J_k$ are the Bessel functions of the first kind. If the spectrum of the Hamiltonian does not vary greatly in time, as is the case for the 1-D problem we discuss here, then a single set of coefficients $a_k$ can be obtained and truncated to a desired threshold limit for all time-steps. The propagation is performed in this case with the $H_{\text{norm}}$ and a normalized $\psi_0$, using the recursive relationship between Chebychev polynomials. Below is a quick outline for the Chebychev propagator which follows the more detailed explanation given in Goerz [32].

In Table II we present pseudocode for the propagation using Chebychev expansion. Here it is assumed that the coefficients of Eq. (43) are already calculated for the time-step.

In the next sections, we analyze the performance and results of the method comparisons.

TABLE II.

| \begin{align*} |v_k| &= -\frac{1}{2}(H|v_{k-1}) - \beta|v_{k-1}) + |v_{k-2} \rangle \\ |\omega_k| &= |\omega_{k-1} - a_k|v_k\rangle \end{align*} |

end for

return $|\psi(t\times dt)| = |\omega_n\rangle$

FIG. 1. Convergence of the population of the ground state over time using the Crank-Nicolson method, in the length gauge, for a smooth laser pulse with $E_0=0.1$, $\omega=0.14$, $T=1200$. There are 2000 points on either side of $x = 0$.

V. RESULTS

A. Performance

In this section we examine the numerical performance, accuracy and timing of the propagation methods mentioned in section IV. Low-order methods can often be quite efficient for a given time-step but might require very small time-steps to achieve high accuracy. Note also that performance and accuracy can depend on both the spatial and temporal discretization and the balance between the two. Consequently, it is important to examine both parameters before drawing any conclusions.

In the present context, the term accuracy is measured by the convergence of the populations of the bound and
continuum states as a function of time for some fixed spatial and time-step.

1. Excitation Results for a Spatial Grid $x = 0.1$

For the ground and low-lying bound states examined in section, it is sufficient to use a computational region that is 200 a.u. to either side of the origin and a spatial grid of 0.1 a.u. This spatial grid is sufficient to ensure that the populations of the ground and low lying excited states would not change at the end of the pulse if the grid was refined further. This grid results in a $4001 \times 4001$ tridiagonal matrix. As a baseline, we first examined the evolution of the ground state population, $|\langle \psi | v_0 \rangle|^2$, in time using the CN method for a smooth pulse in the length gauge.

In Fig. 1 the CN results are shown for three different time-step sizes. The time-steps in this graph increase top to bottom and are written next to each curve. Any line with $\delta t < 0.01$ will lie on top of the upper most curve (blue curve online). We are using a solid black line for $\delta t=1$, a solid thick (red online) for $\delta t=0.1$ and a dashed thin line (blue online) for $\delta t=0.01$. The figure indicates that for the CN method converged results for the probabilities require a $\delta t = 0.01$ or smaller. The upper panel of Fig. 2 displays a magnified portion of Fig. 1 while the lower panel displays two new plots using the Lanczos and Chebychev methods. The Lanczos calculations used a maximum of 20 vectors (i.e., $|v_k\rangle$ in Eq. 39 for $k \in \{1, \ldots , 20\}$), with an initial $\delta t = 1.0$. Two different convergence limits were used. The dash-dot line (green online) used a tolerance to $10^{-4}$, while the solid line (purple online) used a tolerance of $10^{-5}$. The adaptive time-step technique employed in the Lanczos method reduced the initial $\delta t = 1.0$ to $\delta t = 0.0625$ to attain or exceed convergence with this number of Lanczos steps. For completeness, we also show a plot of the result of applying the fully diagonalized time-dependent Hamiltonian Eq. 31, using a $\delta t = 0.1$ (black scattered dots) and the result of the Chebychev propagator (thin solid line, orange online) using 34 vectors with tolerance of $10^{-9}$ and time-step of $\delta t = 0.1$.

In Table III we have listed the average propagation times for each of the methods, as performed serially on a desktop PC with CPU of 3.4 GHz and with an Intel Fortran compiler with -Ofast optimization flag. The timings are for a spatial grid $\delta x = 0.1$ a.u., in a box of size 4001 grid points, centered at zero. All other parameters are as in Fig. 1. The total time of propagation is 1200 a.u. For Lanczos iteration a maximum iteration number of 20 was used and the error threshold was set to $10^{-5}$. Note, other settings can also effect the timing of Lanczos and Chebychev iterations, but again the purpose here is to achieve convergence in the probability of the populations of the ground and excited states.

| Method                  | Time-Step $\delta t$ | Avg. propagation time (sec.) |
|-------------------------|----------------------|-----------------------------|
| Crank-Nicolson          | 0.01                 | 29                          |
| Split-Operator$_{\text{ind}}$ | 0.5               | 32                          |
| Split-Operator$_{\text{fin}}$ | 1.0            | 100                         |
| Even-Odd-Split-Operator | 0.001               | 180                         |
| Lanczos                 | 1.0 - 0.0625         | 14                          |
| Split-Operator$_{\text{ind}}$ + Lanczos | 1.0 - 0.125  | 10                          |
| Split-Operator$_{\text{fin}}$ + Lanczos | 1.0 - 0.25  | 24                          |
| Chebychev               | 0.16                 | 8                           |

TABLE III. Average propagation times using different propagation methods. In all of the test cases the spatial grid was fixed at $\delta x = 0.1$, which results in a Hamiltonian matrix of size $N = 4001$ having 2000 points on either side of $x = 0$. $\delta t$ was chosen so the probability of the ground state converged to a consistent profile as $\delta t \to 0$.

We have also tested the influence of a square wave pulse Eq. (15) on the convergence of the probabilities of low-lying states in time. The square wave pulse shows an even greater dependence of the Crank-Nicolson on small time-steps. Figure 3 shows that a square wave pulse drives down the population of the ground state even more rapidly than a smooth pulse and it is necessary to choose...
\( \delta t = 0.001 \) in the CN method to achieve satisfactory convergence. On the other hand, the Lanczos method remains as accurate and as efficient as in the smooth pulse.

2. Reducing the Spatial Grid

Although it is not necessary to reduce the spatial grid to achieve converged results in this problem, we felt it was important to show data illustrating the performance of the methods under such conditions. As the grid is refined, the spectral radius of the discretized Hamiltonian can get quite large. The size of the spectral radius influences many explicit time propagation schemes. Of course, there may be ways to ameliorate these effects using preconditioning or other clever approaches, but those are considerations for another paper.

In Table IV we display what happens if \( \delta x \) is reduced. Here we apply the split-operator+Lanczos approach, using an adaptive time-step with a fixed maximum number of vectors. Notice that a smaller \( \delta x \) requires a larger number of Lanczos iterations and a smaller \( \delta t \) to achieve convergence. This is a direct consequence of the larger spectral radius of the discretized Hamiltonian. The numbers displayed for \( \delta t \) in column two are representative of the minimum time-step automatically chosen by the Lanczos propagation algorithm. At each time-step the method will automatically choose \( \delta t \) to converge and it is impossible to predict in advance what the size of that step would be. The Lanczos method is more efficient than CN at the larger spatial steps and comparable to it at the smaller steps.

Timings for Split-Operator + Lanczos with fixed maximum number of Lanczos Iterations and an adaptive time-step

| \( \delta x/N \) | \( \delta t \) | Iterations | computation time |
|----------------|-----------|------------|-----------------|
| 0.1/4001       | 0.0625    | 15         | 11 sec.         |
|                | 0.125     | 20         | 10 sec.         |
|                | 0.25      | 50         | 14 sec.         |
| 0.05/8001      | 0.03125   | 15         | 62 sec.         |
|                | 0.03125   | 20         | 63 sec.         |
|                | 0.125     | 50         | 85 sec.         |

TABLE IV. Column one displays the size of the spatial grid, \( \delta x \) and the matrix \( N \). The initial time-step is \( \delta t = 1.0 \) but is automatically reduced (column two) according to the convergence criterion (see Fig. 1) and the maximum number of Lanczos iterations (column three) allowed in the computations. The propagated solution is converged to a tolerance = \( 10^{-5} \). The computation times illustrate the sensitivity to the spectral range of the matrices.

In Table V we fix both the time-step and the maximum number of Lanczos iterations. Again, the Lanczos and CN methods are comparable at the larger spatial step but the spectral range of the Hamiltonian matrix for the smaller spatial step sizes slows down the Lanczos considerably if we do not employ an adaptive time-stepping scheme. The Chebychev propagator also suffers from dependence on the spectrum range of the Hamiltonian.

Timings for Split-Operator + Lanczos with a fixed number of Lanczos iterations and a fixed time-step

| \( \delta x/N \) | \( \delta t \) | Iterations | computation time |
|----------------|-----------|------------|-----------------|
| 0.1/4001       | 0.5       | 54         | 24 sec.         |
|                | 0.25      | 30         | 25 sec.         |
|                | 0.1       | 15         | 35.5 sec.       |
|                | 0.05      | 10         | 1.2 min.        |
| 0.05/8001      | 0.5       | 380        | 16 min.         |
|                | 0.25      | 94         | 2.8 min.        |
|                | 0.1       | 38         | 3.2 min.        |
|                | 0.05      | 20         | 5 min.          |
|                | 0.025     | 13         | 8.5 min.        |

TABLE V. First and second columns as in Table IV. The third column shows the number Lanczos iterations required to converge the results to the same profile as in Fig. 1. The last column displays the computation times.

One might wonder whether the slowing down of the Lanczos propagation with the decrease of spatial step size, \( \delta x \), is intrinsically due to the need to include terms associated with the large eigenvalues in the exponential sums. Recall that the time-evolution operator of the Lanczos propagator is:

\[
\hat{U}^L = \sum_k |\lambda_k\rangle \exp(-i\lambda_k \delta t)\langle \lambda_k|.
\]

The eigen-pairs \( \lambda_k \) and \( |\lambda_k\rangle \) change with each Lanczos iteration, \( k \). In contrast to what one might expect, the tests show that the convergence of this operator is actually more dependent on the accuracy of the lower-lying eigen-pairs rather than the high-lying ones. Indeed our tests show that when the wave-function convergence is achieved, for a given \( k \) number of iterations, the terms associated with higher eigenvalues in the exponential sum above have negligible effect on the convergence of \( \hat{U}^L \). This strongly suggests that other strategies employing a larger initial vector space, such as block Lanczos [33] or Davidson [34], might be profitably employed to increase efficiency.

B. Above Threshold Ionization Spectrum

As a final test of the propagation methods discussed in the previous sections, we reproduce some of the results of Javanainen et al. [3] for the above threshold ionization (ATI) spectrum. As a consequence of the spatial discretization inherent in the numerical method, the authors of Javanainen et al. [3] take the approach of finding probabilities of the photo-electron spectrum by averaging over the odd and even eigenstates of the atom.

Using this approach the photo-electron spectrum prob-
ability is defined as,

\[ P(E_{1/4}) = \frac{|\langle \phi_k | \psi \rangle|^2}{E_{k+1} - E_{k-1}} + \frac{|\langle \phi_{k+1} | \psi \rangle|^2}{E_{k+2} - E_{k}} \]  \hfill (45)

where

\[ E_{1/4} = \frac{1}{4}(E_{k-1} + E_k + E_{k+1} + E_{k+2}). \]  \hfill (46)

As can be seen in Fig. 3, convergence of the ionization probabilities can depend on the size of the computational region and the energy of electrons ionized. This is not too surprising since the true continuum states do not vanish at the boundaries of the box and the higher the energy, the larger the box size needed for convergence. Here a box of 800 a.u., or larger, on either side of the origin produces converged spectra.

If one defines the probabilities as in Eq.(46), the result depends critically on whether the wavefunction is in the length or velocity gauge. In the length gauge, the quiver motion of the electron is generally non-zero except at times \( t = n + 1/4 \) or \( n + 3/4 \) of the laser cycle. If the spectra are computed at other times, they show different features. In the velocity gauge, the quiver motion is always zero so the spectra are stationary.

It is legitimate to ask which gauge is “correct”, however, the widely adopted answer (which we use) is to perform the computation in either gauge and convert back to the length gauge before computing the probabilities. But, we always compute those probabilities at that point in the laser cycle where the quiver velocity is zero. In reality, the measurement of the probabilities should be made by propagating the wavepacket a few cycles after the pulse has been turned off. If that is done, the issue of the quiver velocity is irrelevant.

In one computational experiment we examined the probabilities when the quiver motion was non-zero. The quiver velocity adds kinetic energy to the electrons moving to the right and to the left of the origin. This results in a doubling of the peaks at times when quiver velocity is not exactly zero. Any method which employs a variable \( \delta t \) needs to ensure at the end of the propagation when the probabilities are measured that the quiver velocity is zero.

VI. EXTENSION TO HIGHER SPATIAL DIMENSIONS

Naturally one may ask how the conclusions of the current study carry over to higher dimensions. Let us take the case of two dimensions (x,y) in Cartesian coordinates,

\[ \left[ H_{XY} - \frac{i}{\partial t} \right] \Psi(X|Y; t) = 0 \]

\[ H_{XY} = H_X + H_Y + V_{XY} + U_X(t) + U_Y(t). \]  \hfill (47)

where, \( H_I \) is the Kinetic energy operator in the \( I^{th} \) dimension, \( V_{IJ} \) the interparticle interaction and \( U_I(t) \) the
interaction of coordinate \( I \) with the field. For the CN method, a straightforward generalization would require the solution of a much larger set of linear algebraic equations. However, operator splitting allows us to write the second order accurate, short-time evolution operator as,

\[
\exp(-iH_{XY}\delta t) \approx \exp(-i[H_X + U_X(t)]\delta t/2) \\
\exp(-i[H_Y + U_Y(t)]\delta t/2) \\
\exp(-iV_{XY}\delta t) \\
\exp(-i[H_X + U_X(t)]\delta t/2) \\
\exp(-i[H_Y + U_Y(t)]\delta t/2) . \tag{48}
\]

The CN approximation may then be applied to the first, second, fourth and fifth exponentials. The middle exponential is diagonal in space for a local operator. If one examines the most expensive part of applying the operator in Eq (48) to a vector, it involves the solution of a one dimensional set of linear equations that are parametric on the value of the other coordinate. The scaling is \( n_x n_y^2 \) (or \( n_y n_x^2 \)) for a general matrix.

For the tridiagonal case in this paper, there are further savings that reduce the scaling to \( 3n_x n_y \). For example, in the three-dimensional hydrogen atom, in the length gauge and for linearly polarized light, one may split the Hamiltonian into a radial part and a coupling term \([35]\). The coupling to the radiation field is only to nearest neighbor angular momenta and is therefore also tridiagonal. It thus becomes possible to reduce the problem to solving alternate sets of tridiagonal linear equations.

The situation in the velocity gauge is not quite as simple since the coupling depends on the derivative of the wavefunction with respect to the radial coordinate, but it is still possible, using splitting techniques, to reduce the problem to solving sets of tridiagonal linear systems.

For light with arbitrary polarization or for more complex problems, such as molecules exposed to laser fields, formulating the problem in a spherical coordinate system is possible, but intrinsically leads to coupling of large angular momenta due to the nature of the polarization of the light, the electron-nuclear interactions or both. Here reduction to tridiagonal form becomes impossible.

The Lanczos method only requires the application of the Hamiltonian to a vector. In performing this operation one may be able to use a variety of approaches to exploit the structure of the problem under study. Examining those operations, one can see that the scaling of the required matrix vector multiplies is the same as the CN method. In some problems, the application of the interaction potential \( V_{XY} \) can become time consuming. To cite one example, using a finite element discrete variable representation in space can improve the spectral accuracy over a finite difference method, but as an intermediate step it requires the solution of the Poisson equation. This can be done efficiently, but in general that is not the case. Further details will not be elaborated upon here.

The Lanczos or Chebychev methods offer the most flexibility in exploiting the structure of the matrix without the additional penalty of having recourse to operator splitting and behave well with the size of the time step. We are aware of an unpublished study \([10]\) that addresses the comparison of several propagation schemes for the TDSE of a hydrogen atom in a strong linearly polarized EM field. Most of the methods compared in the study \([10]\) are already in the literature \([35–45]\). Examining the available information, one sees strong overlaps in the methods. The spatial discretization schemes are either three point finite difference formula or finite element discrete variable representation. The time propagation approaches vary significantly from CN to Padé to split operator and Krylov space methods such as Lanczos/Arnoldi. It is difficult to discern how much of the differences between these methods is due to the accuracy of the spatial discretization and how much is due to the time propagation method. In addition, the question of the programming efficiency of the codes is hard to address. This last issue could in fact be a reason for the differences observed in the timing. One of the reasons we undertook the current study was to eliminate the question of the spatial discretization and focus on the time propagation. For better or worse, the current study of the methods was done by a single group.

**VII. CONCLUSIONS**

We presented a study of the soft-core, one-dimensional hydrogen atom in a strong electromagnetic field \([5–4]\). This model has been shown to be a reasonable approximation of the full hydrogen atom but does not include any effects due to angular momentum coupling. The solution was propagated in time using a number of methods; Crank-Nicholson \([11]\), various flavors of Split Operator \([15–22]\), the short iterative Lanczos \([24–26, 28]\), Chebychev propagator \([30]\), and a combination of Lanczos and split operator. The study examined the effects of the temporal and spatial step sizes on the efficiency of the propagation method as well as the question of using the length vs. velocity gauge. With proper care either gauge may be used in this problem although the velocity gauge may be more efficient for calculations using large distances \([3]\). Our conclusions demonstrate the efficiency of a method depends heavily on the spatial step size and how one propagates in time.

This may be counterintuitive to some readers’ beliefs that implicit techniques can be used with larger time-steps than explicit methods. However, section \([IV]\) outlines the drawbacks of each technique. Implicit methods tend to collapse with large \( N \), while explicit methods suffer from exponentiation issues of the operator \( H \). As seen in Table \([III]\), Crank-Nicolson excels in computation involving small \( \delta t \). The differences between convergence of CN and Lanczos were covered in Fig \([2]\]. Additional experiments proved the flexibility of Lanczos iteration in various sizes of spatial grid with Split-Operator propagation.
Table I indicates that other methods (block Lanczos, Davidson, etc.) may further improve computation time and accuracy given the convergence of Split-Operator+Lanczos method. The results also match the above-threshold ionization of Javanainen et al. [3]. We have also provided some guidance as to what to expect in higher spatial dimensions.

VIII. ACKNOWLEDGMENT

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IX. PERSONAL REMARKS

While it took a lot after brain scratching after 45 years, and with some assistance from Bill, we were able to reconstruct the history of when we first met. It was in Paris in 1973, at a satellite meeting prior to the VIII International Conference on Photonic, Electronic and Atomic Collisions (ICPEAC), which was held in Beograd, Yugoslavia. Bill did not attend the main conference, and even he does not recall the reason now. I do recall that there was a problem with the plumbing functioning at my hotel above the fourth floor which was a bit of an inconvenience for a few days. However, compared to what happened at the IX ICPEAC two years later in Paris, it was pretty minor. At the time Bill was a fellow in the chemistry department at Harvard and was able to attend some very high quality graduate students like Jimmie Doll, Rick Heller, Tom Rescigno, Steve Adelman, and Hashim Yamani to his group. Bill went on to faculty positions at the University of Colorado, the University of Pennsylvania and the University of Washington where he mentored a number of other very talented graduate students who have gone on to their own quite productive careers.

Bill and I remained friendly over the years but when I left the NSF and took a position at NIST as the General Editor of the Digital Library of Mathematical Functions (DLMF), a project dear to Bill’s heart and one where he had contributed three important chapters, that our friendship became much closer. Since 2014, when I arrived at NIST, Bill and I have discussed various aspects of the DLMF on a very regular basis. With a decision to expand the DLMF to include new material, these discussions became even more regular as Bill again became a contributor to the added material. We also found we had a lot more in common than just science and applied mathematics and often our conversations took extended detours into music and other topics of mutual interest. Bill retired from the University of Washington a few years ago and moved to Washington, DC, where he spent a few years, then very briefly to Boston and he now lives in Santa Fe, where BIS spent 20 years while working at Los Alamos. We talk regularly these days via various video-conferencing applications and are now pulling together the final touches on two of the chapters of the DLMF.

It has been a great pleasure to know Bill and his wife Katrina over these many years. I always come away from our interactions with either new found knowledge or just enjoyment from our discussions. I look forward to that continuing for years to come.

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