A new approach for determining the time step when propagating with the Lanczos algorithm

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1. Introduction

In chemical physics, and in many other fields, one often needs to solve

\[ i \frac{\partial}{\partial t} \psi(x, t) = H \psi(x, t) \]  

where \( H \) is a linear operator [1–6]. If \( H \) is a Hamiltonian operator this is the time-dependent Schrödinger equation (TDSE), in atomic units. Solving the TDSE enables one to calculate photodissociation cross section, rate constants, etc. [1,2,7–10]. Many algorithms have been suggested for solving the TDSE. In this communication we propose new ideas for using the short iterative Lanczos (SIL) approach. The SIL algorithm was proposed by Park and Light [6] and has been developed by other authors [11–18].

To solve Eq. (1), the first step is to convert it into a linear algebra problem by introducing a basis. In this fashion one obtains the equation,

\[ i \frac{\partial a(t)}{\partial t} = Ha(t) \]  

where

\[ \psi(x, t) = \sum_k a_k(t) \phi_k(x) \]  

and

\[ H_{k',k} = \langle \phi_{k'} | H | \phi_k \rangle. \]  

it being assumed that the basis functions are orthonormal. The SIL algorithm is one way of solving this equation, i.e., obtaining \( a(t) \) knowing \( a(t = 0) \).

2. Short iterative Lanczos

The Lanczos algorithm is widely used for calculating eigenvalues and eigenvectors [19–22]. Lanczos vectors \( q_j \), and elements, \( \alpha_j \) and \( \beta_j \), of a tridiagonal matrix are found recursively so that

\[ HQ_m = Q_{m+1} \tilde{T}_m \]  

where

\[ \tilde{T}_m = \begin{bmatrix} \alpha_1 & \beta_1 & 0 & \ldots & 0 \\ \beta_1 & \alpha_2 & \beta_2 & \ldots & 0 \\ 0 & \beta_2 & \alpha_3 & \beta_3 & \ldots \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ 0 & \ldots & \beta_{m-2} & \alpha_{m-1} & \beta_{m-1} \\ 0 & \ldots & 0 & \beta_{m-1} & \alpha_m \\ 0 & \ldots & 0 & 0 & \beta_m \end{bmatrix} \]  

and \( Q_m \) is a matrix whose \( m \) columns are the Lanczos vectors. If \( m \) is small enough, the Lanczos vectors are orthogonal and \( Q_m^T Q_{m+1} \) is a matrix having ones on the diagonal and zeros everywhere else. Therefore,
\( T_m = Q_m^T H Q_m \) \tag{7}

is an \( m \times m \) tridiagonal matrix \([19,23]\).

We wish to compute \( a(t) = e^{-itH}a(0) \). With the SL method, this is done by replacing \( a(t) \) with \( Q_m c(t) \) and premultiplying by \( Q_m^T \). If \( t \) is small enough one obtains,

\[
c(t) = Q_m^T a(t) \approx e^{-itT_m}c(0). \tag{8}
\]

This idea only works if \( m \) is small enough that numerical error does not ruin the orthogonality of \( Q_m \) \([24,25]\). Because \( m \) is small and \( T_m \) is tridiagonal Eq. (8) is easy to solve. If \( a(t) = 0 \) is the Lanczos starting vector then

\[
c(0) = c_0 = \begin{bmatrix} 1 \\ 0 \\ \vdots \\ 0 \end{bmatrix} \tag{9}
\]

The SL method is typically used by choosing a value of \( m \), propagating from \( t = 0 \) to \( t = \Delta t \), and then using \( a(\Delta t) \) as the starting vector and propagating again from \( t = \Delta t \) to \( t = 2\Delta t \), etc. Clearly, it is advantageous to make \( \Delta t \) as large as possible, hence \( \Delta t \) must be chosen so that the size of the error at each step is acceptably small. Park and Light suggest choosing \( \Delta t \) such that

\[
\left| c(\Delta t)_m \right|^2 \approx \frac{1}{(m-1)!} \prod_{k=1}^{m-1} \beta_k \leq \epsilon \tag{10}
\]

where \( \epsilon \) is a specified acceptable error and \( c(\Delta t)_m \) is the \( m \)-th component of the \( c(\Delta t) \) \([6]\). This method of choosing \( \Delta t \) is based on the idea that the propagation should be accurate if \( a(t) \) can be expanded in terms of the first \( m \) Lanczos vectors.

3. A New Estimate for \( \Delta t \)

Several authors have used the Taylor series expansion of the exponential to derive estimates for the error of the Krylov approximation to \( e^{iH}v \) where \( A \) is a matrix and \( v \) is a vector \([15,16]\). These error estimates are very conservative, i.e., the estimated error is considerably larger than the actual error. They depend on the spectral range of \( A \). If these error estimates are used to determine the size of the Krylov space necessary to obtain a good approximation for \( e^{iH}v \) one finds that a very large space is required. Often it is impractical to use large Krylov spaces. In this communication we focus on the problem of obtaining an error estimate for a Krylov approximation to \( e^{-\lambda M}v \). Park and Light \([6]\) addressed using a Taylor series approach.

Stewart and Leyk (SL) derive better error estimates for computing \( e^{iH}v \) where \( A \) is a matrix and \( v \) is a vector \([14]\). We begin with their analysis, but generalize it, so that we can obtain an error estimate for \( e^{-\lambda H}v \). According to Lemma 2 of SL,

\[
\left\| e^{-\lambda H}v - Q_m e^{-\lambda T_m} Q_m^T v \right\|_2 \leq 2 \|v\|_2 \|r\| \tag{11}
\]

where

\[
\|r\| = \min \left\{ \left\| e^{-\lambda} - P(\lambda) \right\| : \lambda \in [\lambda_{\min}, \lambda_{\max}] \right\}, \tag{12}
\]

with \( P \) a polynomial of degree \( m - 1 \), and \( T_m \) and \( Q_m \) are the tridiagonal matrix and the matrix of Lanczos vectors associated with \( A \), and \( \lambda_{\min} \) and \( \lambda_{\max} \) are minimum and maximum eigenvalues of \( A \). One chooses the polynomial to minimize and the value of \( \lambda \) to maximize. The SL approach is not based on a Taylor expansion but rather on a rigorous bound of the error of the Krylov subspace approximation. We need an estimate for

\[
\left\| e^{-i\Delta t H}v - Q_m e^{-i\Delta t T_m} Q_m^T v \right\|_2 \tag{13}
\]

where

\[
\|r\| = \min \left\{ \left\| e^{-i\lambda} - P(-i\lambda) \right\| : \lambda \in [\lambda_{\min}, \lambda_{\max}] \right\}, \tag{14}
\]

with \( \lambda_{\min} \) and \( \lambda_{\max} \) being now minimum and maximum eigenvalues of \( H \).

To bound \( \|r\| \) we choose a Chebyshev series for \( P(-i\lambda) \) and expand \( e^{-i\lambda H} \) in terms of Chebyshev polynomials. This is done following Ref. \([5]\). One begins by scaling \( H \).

\[
M = \frac{2H - I(\lambda_{\max} + \lambda_{\min})}{(\lambda_{\max} - \lambda_{\min})}. \tag{15}
\]

Now

\[
e^{-i(cM + d)} = e^{-i(cM + d)} \tag{16}
\]

where

\[
c = (\Delta t/2)(\lambda_{\max} - \lambda_{\min}); \quad d = (\Delta t/2)(\lambda_{\max} + \lambda_{\min}). \tag{17}
\]

Expanding \( e^{-i(cM + d)} \) in a Chebyshev series gives

\[
e^{-i(cM)} = \sum_{k=0}^{\infty} (2 - \delta_{k,0})(-i)^k f_k(c(t)) T_k(M) \tag{18}
\]

The bound on \( \|r\| \) is \( \sum_{k=0}^{\infty} |f_k| \). From Eq. (13) and the fact that \( v \) is normalized we deduce that the error is bounded by

\[
e_k = 2 \| f_m(c) \| + \| f_{m+1}(c) \| + \| f_{m+2}(c) \| + \cdots. \tag{19}
\]

In terms of Bessel functions this is

\[
e_k = 4 \left| J_m(c) \right| + \left| J_{m+1}(c) \right| + \left| J_{m+2}(c) \right| + \cdots. \tag{19}
\]

Defining \( \alpha = \frac{ ec}{2m} \left( \frac{2m}{\sqrt{2\pi m}} \right)^m = \frac{\alpha^m}{\sqrt{2\pi m}} \), we find that

\[
e_k < \frac{4\alpha^m}{\sqrt{2\pi m}} \left( 1 + \alpha + \alpha^2 + \alpha^3 + \cdots \right)
\]

where we have assumed that \( \alpha < 1 \). We equate the right side of Eq. (20) to a specified acceptable error, \( \epsilon \), solve for \( \alpha \) and from \( \alpha = \frac{ ec}{2m(\lambda_{\max} - \lambda_{\min})} \), determine \( \Delta t \). This method for determining \( \Delta t \) will only be valid if the value of \( \alpha \) obtained by solving Eq. (20) is less than unity, but that will always be the case if \( \epsilon < 1 \).

Eq. (20) can be solved using the Newton–Raphson method. Often a good starting point is obtained by dropping the \((1 - \alpha)\) in the denominator of Eq. (20), i.e.,

\[
\alpha \approx \left( \frac{\sqrt{\pi m}}{8} \right)^{\frac{n}{m}}. \tag{21}
\]

For test problems we have solved, the Newton–Raphson method converges in 5 or 6 iterations.
4. Calculations

To test the ideas we have propagated a Gaussian wavepacket in a 1d harmonic potential. The Hamiltonian (atomic units are used) is

$$H = \frac{p^2}{2m_0} + \frac{1}{2} m_0 \omega^2 x^2,$$

where \( m_0 = 1 \) and \( \omega = 2.7338 \times 10^{-4} \), corresponding to a wave-number of 60. The starting wavepacket is displaced from equilibrium by \( x_0 = 12 \):

$$\psi(x,0) = \frac{\alpha_0}{\sqrt{\pi}} e^{-\alpha_0^2(x-x_0)^2/2},$$

$$\alpha_0 = \sqrt{m_0 k^2 \hbar^2}^{1/4}; \quad \omega = (k/m_0)^{1/2}. $$

The exact solution is known,

$$\|\psi(x,t)\|^2 = \frac{\alpha_0}{\sqrt{\pi}} e^{-\alpha_0^2(x-x_0 \cos(\omega t))^2}. $$

[28].

The Hamiltonian matrix was constructed using the sinc discrete variable representation (DVR) of Ref. [29]. The size of the matrix is 80. We set \( m = 22 \). The DVR points are between \(-550\) and \(+550\) bohr. The spectral range is \( (\lambda_{max} - \lambda_{min}) = 0.0309 \) hartree. In Table 1 we compare values of \( |\psi_{exact}(x,200\Delta t) - \psi_{SIL}(x,200\Delta t)| \) at 7 different points and for 4 different choices of \( \epsilon \). In all cases the calculated errors are less than the specified acceptable error, \( \epsilon \). For \( \epsilon = 1.0 \times 10^{-4}, 1.0 \times 10^{-6}, 1.0 \times 10^{-8}, 1.0 \times 10^{-10} \), the values of \( \alpha \), found by solving Eq. (8) are 0.6581, 0.542, 0.4427, 0.3613 respectively. These are close to the zeroth-order estimates of Eq. (21): 0.69098, 0.560475, 0.4546187, 0.368755 respectively.

Is the new method better than previous approaches? A good time step criterion will minimize the number of matrix-vector products (i.e., the number of time steps) required to propagate to \( t = T \) without allowing error to creep into the solution. In Table 2, we compare results obtained choosing \( \Delta t \) according to Eq. (20) and with the Park and Light [6] and the Mohankumar and Auerbach (MA) [17] criteria, for the same test problem. Both the other methods are based on the Taylor expansion of the matrix exponential. The MA criterion ignores the importance of the error caused by projecting into a finite Krylov space. In all cases we set \( \epsilon = 10^{-12} \) and the wavepacket is propagated for about 1000 fs. In the Park and Light and the Mohankumar and Auerbach schemes, \( \Delta t \) is not constant and we report maximum and minimum values. The Park and Light scheme has larger time steps and therefore requires fewer time steps, however, this advantage is irrelevant because the Park and Light errors are much larger than \( \epsilon \). The Mohankumar and Auerbach scheme gives good errors. The calculated error is (slightly) larger than \( \epsilon \) at only two points. If the Eq. (20) criterion is used the error is smaller than \( \epsilon \) at all points, and the number of time steps is smaller than in the MA case. The superiority of the Chebyshev approach presented in this communication is obvious.

5. Conclusion

We have presented an explicit time step expression for the time evolution of wavepackets by Lanczos methods. It is derived by estimating the remainder of Chebyshev series representing the matrix exponential. If propagated with the new time step criterion the error in the exponential wavepacket never exceeds the specified error used to determine the time step. In addition the number of required matrix-vector products is moderate.

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References