ITVOLT: An Iterative Solver for Volterra Integral Equations with Application to the Time-Dependent Schrödinger Equation

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Abstract

We present a novel iterative method for solving Volterra integral equations of the second kind. Based on global Lagrange interpolation, the method is simple to implement and applicable to a wide variety of problems. Here, we present the method in detail and discuss several applications, emphasizing in particular its use on the time-dependent Schrödinger equation.

1 Introduction

Volterra integral equations of the second kind take the form

\[ f(t) = g(t) + \int_{t_0}^{t} K(t,t')f(t')dt', \quad t_0 \leq t \leq t_f \quad (1.1) \]

for two vector-valued functions \( f \) and \( g \) and a matrix integral kernel \( K \), each of which may be real or complex. In this general form, (1.1) describes a set of coupled, one-dimensional Volterra integral equations in the variable \( t \).

Studied first in one dimension by Liouville [14] and later Volterra [22], these equations arise in a variety of research disciplines, including systems theory, viscoelasticity, and probability (see Chapter 2 of Linz [13] for a survey of some of these applications). More generally, any first order differential equation can be converted to a Volterra integral equation by integrating. Given an inhomogeneity \( g \) and an integral kernel \( K \), the equations are solved for the unknown function \( f \).

Numerical methods for these problems have been developed over several decades and are the subject of numerous papers. Early efforts focused on applying a quadrature to the integral in the equation, beginning with the simple trapezoidal rule and generalizing to more complicated quadrature schemes. For an in-depth survey of these techniques, see Brunner and van der Houwen [4]. More recent approaches make use of increasingly complicated mathematical machinery, including interpolation [20], block methods [10], and homotopy theory [5].

The goal of this work is to develop a numerical method that is computationally simple yet capable of producing highly accurate solutions to (1.1) even when the interval \([t_0, t_f]\) is large. The method we present proceeds by applying a global Lagrange interpolation to the integrand and solving the resulting system of linear equations iteratively. This approach is similar to an iterative method proposed by Ndong et al. [15] for solving the time-dependent Schrödinger equation. While we will consider how our method applies to the Schrödinger equation in this paper, we present it in a more general way first, relevant for any problem that can be written as a Volterra integral equation.

The remainder of the paper is organized as follows. In sections two and three, we present the numerical details of the method and demonstrate its utility on a handful of toy problems. In the subsequent sections, we consider in depth the application of our method to the time-dependent Schrödinger equation, illustrating its ability to achieve highly accurate results on a number of physical problems.

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2 Iterative Volterra Propagator (ITVOLT)

In this section, we develop the methodology in detail and discuss both the numerics and some associated theoretical considerations. We follow this with several numerical examples.

2.1 Statement of the Method

We begin by choosing a set of points \( \{t_i\}_{i=1}^{n} \in [t_0, t_f] \) and expanding \( K(t, t')f(t') \) in the corresponding set of Lagrange polynomials \( l_i(t) = \prod_{j \neq i} \frac{t - t_j}{t_i - t_j} \):

\[
K(t, t')f(t') \approx \sum_{j=1}^{n} K(t, t_j)f(t_j)l_j(t'). \tag{2.1}
\]

Integrating the Lagrange polynomials over each sub-interval to obtain a set of weights \( w_{i,j} = \int_{t_0}^{t_i} l_j(t')dt' \), the Volterra integral equation (1.1) can then be rewritten as

\[
f(t_i) \approx g(t_i) + \sum_{j=1}^{n} K(t_i, t_j)w_{i,j}f(t_j), \tag{2.2}
\]

at any point \( t_i \). This replaces the integral in the equation with a quadrature at the points \( t_1, \ldots, t_n \), where the weights can be computed exactly using a sufficiently high-order Gauss quadrature. Note that we have a unique set of weights \( \{w_{i,j}\}_{j=1}^{n} \) for each quadrature point \( t_i \), where \( w_{i,j} \) corresponds to integrating the \( j \)th Lagrange polynomial over \( [t_0, t_i] \). By choosing a global set of points - i.e., that span the entire region \( [t_0, t_f] \) - and computing integration weights for all of the sub-intervals, we are able to achieve arbitrary accuracy limited solely by the number of points used. Moreover, the resulting quadrature is semi-global, since evaluating the integral on \( [t_0, t_i] \) requires using all of the points, including those outside the interval.

At this point, (2.2) is a set of linear equations that can be solved using standard numerical tools. For the multi-dimensional problems we are ultimately interested in solving, however, this would be computationally expensive. Instead, we suggest an iterative approach, where at the \((k+1)\)st step of the process we use \( f^{(k)}(t) \), the \( k \)th iterate, on the right hand side of (2.2) to obtain

\[
f^{(k+1)}(t_i) = g(t_i) + \sum_{j=1}^{n} K(t_i, t_j)w_{i,j}f^{(k)}(t_j). \tag{2.3}
\]

For an initial approximation, we can take \( f^{(0)}(t) = g(t) \).

This Jacobi type iteration uses only the values of \( f^{(k)} \) to evaluate \( f^{(k+1)} \) at any \( t_i \). A more accurate, but also more computationally demanding, process is a Gauss-Seidel type iteration, where at each step the currently available values of the unknowns \( f^{(k+1)}(t_1), \ldots, f^{(k+1)}(t_{i-1}) \) are used to compute the next value according to

\[
(I - w_{i,i}K(t_i, t_i))f^{(k+1)}(t_i) = g(t_i) + \sum_{j=1}^{i-1} K(t_i, t_j)w_{i,j}f^{(k+1)}(t_j) + \sum_{j=i+1}^{n} K(t_i, t_j)w_{i,j}f^{(k)}(t_j). \tag{2.4}
\]

These iterative expressions are the basis for our method, which proceeds as follows:

1. Choose a set of quadrature points \( \{t_i\}_{i=1}^{n} \in [t_0, t_f] \) and compute the corresponding set of weights.
2. Evaluate \( g \) at the quadrature points, setting \( f^{(0)}(t) = g(t) \).
3. Given \( f^{(k)}(t) \), apply either the Jacobi (2.3) or Gauss-Seidel (2.4) iterations.
4. Continue until \( \max_i ||f^{(k+1)}(t_i) - f^{(k)}(t_i)||_2 \) falls below a given tolerance or a maximum number of iterations is reached.

Throughout, we refer to our method as ITVOLT, short for Iterative Volterra Propagator. When relevant, we distinguish between the two versions of the iteration as ITVOLT (Jacobi) and ITVOLT (Gauss-Seidel).
2.2 Numerical Details

The preceding outline emphasizes the simplicity of our method. Nevertheless, there are still a number of subtle numerical details that need to be elucidated.

First, we need to decide on a set of quadrature points. While in principle any set of points would work, the stability and accuracy of the method depend critically on how they are chosen. Equally spaced points are attractive for their simplicity, but they are subject to Runge’s phenomenon [18], which can lead to wild oscillations away from the interpolating points. Although this can be somewhat counteracted by enforcing the condition that all weights are positive [9] or by using barycentric formulas for the interpolation [2], the possibility for increased instability as we push to higher accuracy is counterproductive. Of the many types of non-equally spaced points, we choose Gauss-Lobatto points so that the endpoints $t_0$ and $t_f$ are included in the quadrature. This enables us to solve the integral equation over multiple intervals, where we require the solution from the previous interval to initialize the solution at the next one.

Once the quadrature points are chosen, we must next decide how many to use in the interval $[t_0, t_f]$. While increasing the number of points improves the accuracy of the quadrature, it also increases the computational costs and may impact the number of iterations required for convergence.

Lastly, we need to choose between the Jacobi and Gauss-Seidel versions of ITVolt. The Jacobi iteration is computationally simpler and parallelizable, as each $f^{(k+1)}(t_i)$ can be computed independently given $f^{(k)}$, but it typically requires more iterations than Gauss-Seidel to reach the same level of convergence. The drawback for the Gauss-Seidel iteration is the need to solve a linear system of equations at each quadrature point at every iteration. That is, if $f$ is a vector-valued function in $\mathbb{R}^d$ or $\mathbb{C}^d$, (2.4) is a $d \times d$ system to be solved, which may be expensive if $K(t,t')$ is dense.

In the subsequent numerical examples, we will explore in detail how these parameters/choices affect the results of the method. At the end of the paper, we summarize our recommendations for researchers interested in using it on a problem of their own.

2.3 Convergence Considerations

Before moving to the numerical tests, we explore one last fundamental question: under what conditions should we expect ITVolt to converge? To do this, consider

$$e^{(k)} = \begin{bmatrix} f^{(k)}(t_1) - f^{(k-1)}(t_1) \\ \vdots \\ f^{(k)}(t_n) - f^{(k-1)}(t_n) \end{bmatrix}, \quad k = 1, 2, \ldots \tag{2.5}$$

which records the difference in successive iterates at all of the quadrature points. Rearranging (2.3), we can rephrase the Jacobi iteration in terms of $e^{(k)}$ as $e^{(k+1)} = A e^{(k)}$ for

$$A = \begin{bmatrix} w_{i,j} K(t_i, t_j) \end{bmatrix}_{1 \leq i,j \leq n}. \tag{2.6}$$

Similarly, if we decompose $A$ as $A = A_L + A_U$ for $A_L$ lower triangular and $A_U$ strictly upper triangular, the Gauss-Seidel iteration (2.4) can be rephrased as

$$e^{(k+1)} = (I - A_L)^{-1} A_U e^{(k)}, \tag{2.7}$$

assuming $I - A_L$ is invertible. Observing that $\|e^{(k)}\|_2 \to 0$ if and only if $\|f^{(k)}(t_i) - f^{(k-1)}(t_i)\|_2 \to 0$ for all $t_i$, we obtain the following.

**Theorem 1.** For any kernel $K$ and any quadrature points $\{t_i\}$ and weights $\{w_{i,j}\}$ on $[t_0, t_f]$, let $A = A_L + A_U$ as in (2.6), where $A_U$ is strictly upper triangular. Then the Jacobi version of ITVolt (2.3) converges if and only if $\rho(A)$, the spectral radius of $A$, satisfies $\rho(A) < 1$. Similarly, the Gauss-Seidel version (2.4) converges if and only if $I - A_L$ is invertible and $\rho((I - A_L)^{-1} A_U) < 1$.

**Proof.** This follows directly from a standard result in linear algebra, see for example section 6.10 of [6]. \qed

Of course, this result is only true in exact arithmetic. Nevertheless, the spectral radius $\rho$ is still a good indicator of convergence in practice, and while computing it is not particularly simple, we can easily obtain an upper bound by computing any norm of the matrix.
3 Simple Numerical Tests

In this section, we apply ITVOLT to two toy problems: a two-channel Volterra integral equation and a one-dimensional ordinary differential equation (ODE). Since both of these problems have simple analytic solutions, they serve as a first exploration of how the method is applied in practice as well as how it performs against other techniques. In the next section, we consider how this generalizes to more complicated problems, in particular the time-dependent Schrödinger equation.

In these and all subsequent examples, results were obtained on a Linux machine with sixteen 3.60 GHz processors. Each problem was implemented in Fortran 90 and compiled with gfortran version 8.5.0.

3.1 Two-Channel Problem

As a first test, we solve a simple two-channel example taken from Wang and Wang [23]. Here, our Volterra integral equation (1.1) has kernel

\[
K(t, t') = \begin{pmatrix}
\sin((t - t') - 1) & 1 - t' \cos(t) \\
1 & t - t'
\end{pmatrix}
\] (3.1)

and inhomogeneous term \(g(t) = (g_1(t), g_2(t))^T\) for

\[
g_1(t) = (-\cos^2(t) - \frac{1}{2} \sin(t - 1)) t + 2 \cos(t) + \sin(t) \cos(t) - \frac{1}{4} \cos(1 + t) + \frac{1}{4} \cos(1 - t) - 1
\] (3.2)

and \(g_2(t) = \sin(t) - t\).

For this problem, (1.1) is a system of two coupled Volterra integral equations; at each time \(t\), \(f(t) = (f_1(t), f_2(t))^T\) is a vector in \(\mathbb{R}^2\) with analytic solution \(f_1(t) = \cos(t)\) and \(f_2(t) = \sin(t)\). We are interested in solving this two dimensional problem over the interval \([0, 1]\).

If \(f_{1}^{(k)}(t)\) and \(f_{2}^{(k)}(t)\) are the entries of the converged iterate \(f^{(k)}\) and \(t_1, \ldots, t_n\) are the quadrature points used in \([0, 1]\), we measure accuracy by computing two values:

\[
\varepsilon_{1\text{sol}}^{1} = \max_t \left| f_{1}^{(k)}(t_i) - \cos(t_i) \right|
\]

\[
\varepsilon_{2\text{sol}}^{2} = \max_t \left| f_{2}^{(k)}(t_i) - \sin(t_i) \right|
\] (3.3)

These quantities give the largest error in the solution at any quadrature point for each component. We also track \(k\) - the number of iterations needed to reach convergence, where our convergence tolerance is set to \(10^{-10}\).

Results for both versions of ITVOLT are summarized in Table 1. As is clear from the data, both iterative methods converge and are capable of achieving highly accurate numerical solutions, with results improving as the number of quadrature points is increased. In fact, the only difference between the Jacobi and Gauss-Seidel versions is the number of iterations required for convergence, with Gauss-Seidel requiring significantly fewer. This suggests that Gauss-Seidel may be more efficient than Jacobi despite the increased computational complexity.

Wang and Wang [23] propose a block method for solving this problem that is similarly rooted in Lagrange interpolation. For a direct comparison to their work, we can replace the Gauss-Lobatto points with a Newton-Cotes quadrature, which allows us to compute the solution at equally spaced points. Table 2 shows the absolute solution error of the Gauss-Seidel version of our method against the solution error of Wang and Wang at these points (taken from Table 2 of their paper).

As mentioned earlier, we do not recommend using an equally spaced quadrature in our method since they tend to become unstable (and slow convergence) as the number of points grows; this is confirmed by the fact that the eleven point Newton-Cotes quadrature used to generate the results in Table 2 required 25 iterations to converge, nearly four times the number of points required for any of the cases in Table 1. Nevertheless, ITVOLT still outperforms the numerical approach of Wang and Wang by several orders of magnitude. Note also that Wang and Wang’s method is significantly more computationally demanding, requiring for this problem that \([0, 1]\) be divided into 1200 subintervals to achieve the results listed.
Two Channel Problem: Solution Error

| $n$ | $\varepsilon^1_{\text{sol}}$ | $\varepsilon^2_{\text{sol}}$ | $k$ | $\varepsilon^1_{\text{sol}}$ | $\varepsilon^2_{\text{sol}}$ | $k$ |
|-----|-------------------------------|-------------------------------|-----|-------------------------------|-------------------------------|-----|
| 3   | $3.48 \times 10^{-3}$         | $5.23 \times 10^{-3}$         | 24  | $3.48 \times 10^{-3}$         | $5.23 \times 10^{-3}$         | 7   |
| 4   | $4.11 \times 10^{-4}$         | $2.71 \times 10^{-4}$         | 19  | $4.11 \times 10^{-4}$         | $2.71 \times 10^{-4}$         | 7   |
| 5   | $9.89 \times 10^{-6}$         | $9.76 \times 10^{-6}$         | 18  | $9.89 \times 10^{-6}$         | $9.76 \times 10^{-6}$         | 7   |
| 6   | $2.50 \times 10^{-6}$         | $6.31 \times 10^{-7}$         | 16  | $2.50 \times 10^{-6}$         | $6.31 \times 10^{-7}$         | 7   |
| 7   | $3.95 \times 10^{-8}$         | $1.45 \times 10^{-8}$         | 15  | $3.95 \times 10^{-8}$         | $1.45 \times 10^{-8}$         | 7   |
| 8   | $8.88 \times 10^{-9}$         | $8.20 \times 10^{-10}$        | 15  | $8.88 \times 10^{-9}$         | $8.20 \times 10^{-10}$        | 7   |
| 9   | $1.02 \times 10^{-10}$        | $1.60 \times 10^{-11}$        | 15  | $1.02 \times 10^{-10}$        | $1.60 \times 10^{-11}$        | 7   |

Table 1: Solution error for ITVOLT applied to the two-channel problem \((3.1)\) on \([0, 1]\). Here, \(n\) is the number of quadrature points used, \(\varepsilon^1_{\text{sol}}\) and \(\varepsilon^2_{\text{sol}}\) measure solution error \((3.3)\), and \(k\) is the number of iterations needed to reach convergence under a tolerance of \(10^{-10}\).

Two Channel Problem: Error Comparison

| $x$  | Error in $f_1$ | Error in $f_2$ | Error in $f_1$ | Error in $f_2$ |
|------|----------------|----------------|----------------|----------------|
| 0    | 0              | 0              | 0              | 0              |
| 0.1  | $1.00 \times 10^{-8}$ | $1.00 \times 10^{-8}$ | $1.71 \times 10^{-11}$ | $1.52 \times 10^{-11}$ |
| 0.2  | $1.35 \times 10^{-7}$ | $2.00 \times 10^{-7}$ | $1.73 \times 10^{-11}$ | $1.42 \times 10^{-11}$ |
| 0.3  | $5.63 \times 10^{-7}$ | $9.30 \times 10^{-7}$ | $8.56 \times 10^{-12}$ | $8.37 \times 10^{-12}$ |
| 0.4  | $1.42 \times 10^{-6}$ | $2.27 \times 10^{-6}$ | $1.29 \times 10^{-11}$ | $8.25 \times 10^{-12}$ |
| 0.5  | $2.63 \times 10^{-6}$ | $5.88 \times 10^{-6}$ | $5.49 \times 10^{-12}$ | $6.64 \times 10^{-13}$ |
| 0.6  | $3.84 \times 10^{-6}$ | $1.07 \times 10^{-5}$ | $1.30 \times 10^{-11}$ | $6.59 \times 10^{-12}$ |
| 0.7  | $4.42 \times 10^{-6}$ | $1.68 \times 10^{-5}$ | $9.30 \times 10^{-12}$ | $2.23 \times 10^{-12}$ |
| 0.8  | $3.47 \times 10^{-6}$ | $2.31 \times 10^{-5}$ | $9.67 \times 10^{-12}$ | $1.70 \times 10^{-12}$ |
| 0.9  | $1.02 \times 10^{-7}$ | $2.75 \times 10^{-5}$ | $8.10 \times 10^{-12}$ | $5.94 \times 10^{-13}$ |
| 1    | $6.20 \times 10^{-6}$ | $2.64 \times 10^{-5}$ | $1.05 \times 10^{-11}$ | $7.69 \times 10^{-13}$ |

Table 2: Absolute error comparison between ITVOLT (with an 11 point Newton-Cotes quadrature) and the method of Wang and Wang. Tolerance was once again \(10^{-10}\) and convergence was reached after 25 iterations.
3.2 One-Dimensional ODE

To explore next how ITVOLT applies to first order differential equations, consider the following one-dimensional example:

\[
\begin{aligned}
\left[ i \frac{\partial}{\partial t} - t \right] \psi(t) &= 0, \quad \psi(0) = 1. \\
\end{aligned}
\]  

(3.4)

The solution to this problem is \( \psi(t) = e^{-it^2/2} \). While analytically simple, \( \psi \) oscillates wildly at large times (see Figure 1), making it relatively difficult to handle numerically.

By comparing the results of our method with others on (3.4), we demonstrate the value of approaching problems like this via their corresponding Volterra integral equation. Moreover, we justify our recommendation to build a quadrature from Gauss-Lobatto points and Lagrange interpolation.

In what follows, we are interested in solving (3.4) over \([0, 25] \) on intervals of the form \([\tau_j, \tau_{j+1}]\), beginning with \( \tau_1 = 0 \). We immediately see here the value of using Gauss-Lobatto points: by ensuring on each interval that \( \tau_j \) and \( \tau_{j+1} \) are included as quadrature points, we can pass the solution computed at \( \tau_{j+1} \) as an initial condition to solve the equation on \([\tau_{j+1}, \tau_{j+2}]\).

To formulate ITVOLT on the interval \([\tau_j, \tau_{j+1}]\), we first rewrite (3.4) as

\[
\begin{aligned}
i \frac{\partial}{\partial t} \psi(t) &= t_\alpha \psi(t) + (t - t_\alpha)\psi(t) \\
\end{aligned}
\]

(3.5)

for some fixed \( t_\alpha \). Differentiating \( e^{it_\alpha t}\psi(t) \) and applying (3.5), we obtain

\[
\begin{aligned}
i \frac{\partial}{\partial t} \left[ e^{it_\alpha t} \psi(t) \right] &= (t - t_\alpha)e^{it_\alpha t}\psi(t) \\
\end{aligned}
\]

(3.6)

which can be integrated to yield

\[
\psi(t) = e^{-it_\alpha(t-\tau_j)}\psi(\tau_j) - i \int_{\tau_j}^{t} e^{-it_\alpha(t-t')} (t' - t_\alpha) \psi(t') dt', \quad \tau_j \leq t \leq \tau_{j+1}.
\]

(3.7)

This is the Volterra integral equation to be iterated. Note that for this problem (2.4) is a scalar equation, meaning that ITVOLT (Gauss-Seidel) does not require solving systems of equations.

We will consider two choices of \( t_\alpha \) in the tests that follow: \( t_\alpha = 0 \) and \( t_\alpha = \frac{\tau_j + \tau_{j+1}}{2} \). In the first of these cases, our method collapses to a Picard iteration:

\[
\psi(t) = \psi(\tau_j) - i \int_{\tau_j}^{t} t' \psi(t') dt'.
\]

(3.8)

While the decision to add and subtract a constant from the differential equation before integrating may feel arbitrary, we will see later that this can increase accuracy.
To gain a reference frame for how ITVOLT performs, we will compare the results against two alternative methods for solving (3.4). Each of these is again solved over \([0, 25]\) in successive intervals and uses the same set of quadrature points. Below, we describe briefly how each is performed on \([\tau_j, \tau_{j+1}]\).

1. **Iteration with Power Expansion:** First, we test against a similar iterative method that swaps the Lagrange interpolation for a power expansion. In this case, we interpolate the integrand of (3.7) in monomial powers by solving a system of equations at the Gauss-Lobatto points \(t_1, \ldots, t_n\) of the form

\[
\sum_{k=0}^{n-1} a_k(t_j) t_j^k = e^{-it(t_1-t)} (t_j - t_a) \psi(t_j), \quad 1 \leq j \leq n. \tag{3.9}
\]

Once the coefficients are found, the integral of the interpolating polynomial can then be done analytically. Thus, if \(a_0(t_1), \ldots, a_{n-1}(t_1)\) are found to interpolate \(e^{-it(t_1-t')} (t' - t_a) \psi^{(k)}(t')\), we obtain an iteration of the form

\[
\psi^{(k+1)}(t_j) = e^{-it(t_1-t_j)} \psi^{(k)}(t_j) - i \sum_{k=0}^{n-1} \frac{a_k(t_j)}{k+1} [t_j^{k+1} - t_j^k]. \tag{3.10}
\]

In this approach, new coefficients for the power expansion must be solved for each time \(\psi^{(k+1)}(t_j)\) is evaluated at a different quadrature point (corresponding to \(t_j\) in (3.9) and (3.10)). Note also that, unlike ITVOLT, this approach can only employ a Jacobi type iteration.

2. **Discrete Variable Representation (DVR):** To compare with a non-iterative method, we examine a discrete variable representation (DVR) employing a set of Lagrange polynomials as a basis. DVR methods combine a Gauss quadrature and a corresponding set of orthogonal polynomials to convert a differential equation into a linear system of equations. For more information about how these methods are derived and the many problems they have been applied to, see Light and Carrington [12]. To apply a DVR here, we rewrite (3.6) as

\[
i \frac{\partial}{\partial t} \phi(t) = (t - t_a) \phi(t) \tag{3.11}
\]

for \(\phi(t) = e^{it\sigma} \psi(t)\). On the interval \([\tau_j, \tau_{j+1}]\), we then set \(\phi(t) = \chi(t) + \phi(t_j)\) with \(\chi(t_j) = 0\) and expand \(\chi\) in Lagrange polynomials as

\[
\chi(t) = \sum_{m=2}^{n} c_m \phi_m(t) = \sum_{m=2}^{n} c_m \frac{l_m(t)}{\sqrt{w_m}}. \tag{3.12}
\]

In this expansion, \(l_m(t) = \prod_{k \neq m} \frac{t - t_k}{t_m - t_k}\) is the Lagrange polynomial corresponding to the Gauss-Lobatto points \(t_1, \ldots, t_n\) and weights \(w_1, \ldots, w_n\) on \([\tau_j, \tau_{j+1}]\) (i.e. the points and last set of weights from the quadrature in our method). Note that \(l_1(t)\) is not used in the expansion to enforce the initial condition \(\chi(t_j) = 0\). Inserting this into (3.11) yields

\[
\sum_{m=2}^{n} c_m \left[ i \frac{\partial}{\partial t} (t - t_a) \right] \phi_m(t) = (t - t_a) \phi(t_j). \tag{3.13}
\]

Integrating both sides of this expression against \(\phi_k\) on \([\tau_j, \tau_{j+1}]\), we obtain a linear equation in the coefficients \(c_2, \ldots, c_n\)

\[
\sum_{m=2}^{n} c_m \left[ i \int_{\tau_j}^{\tau_{j+1}} \phi_k(t') \frac{\partial \phi_m(t')}{\partial t'} dt' - (t_m - t_a) \delta_{km} \right] = \sqrt{w_k} (t_k - t_a) \phi(t_j), \tag{3.14}
\]

where the remaining integral can be evaluated using the quadrature. Constructing (3.14) for \(2 \leq k \leq n\), we obtain an \((n-1) \times (n-1)\) system that can be solved for the coefficients \(c_2, \ldots, c_n\). Once these are computed, we have at any quadrature point \(t_2, \ldots, t_n\)

\[
\psi(t_l) = e^{-it\sigma} \psi(t_l) = e^{-it\sigma} \left( e^{it\sigma} \psi(t_j) + \frac{c_l}{\sqrt{w_l}} \right). \tag{3.15}
\]
The main drawback to a DVR approach is the need to solve an equation (appears to respond best to increasing \( n \)), which scales poorly as the problem grows in dimension. In particular, an ODE in two variables expanded in interval \( [0, \tau] \) to reach convergence. The results are summarized in Tables 3 and 4.

Comparing the two tables, we first observe - as mentioned earlier - that all of the methods improve when \( t_\alpha = 0.1 \). Even when decent results are obtained, the tolerance for convergence is set to \( 10^{-10} \), with a maximum of 5\( n \) iterations allowed. Here, INF means the method diverged. The power expansion is omitted from this table since it diverged for each case.

To measure the accuracy of each of these methods, we compute the worst-case absolute error at any quadrature point used in \([0, 25]\). That is, if \( \psi \) is the converged numerical result, we track

\[
\varepsilon_{\text{sol}} = \max_j \max_{t_i \in [\tau_j, \tau_{j+1}]} \left| \psi(t_i) - e^{-it_i^2/2} \right|.
\]

In addition for the iterative methods, we track \( k_{\text{max}} \) - the maximum number of iterations needed at any interval \([\tau_j, \tau_{j+1}]\) to reach convergence. The results are summarized in Tables 3 and 4.

| \( \Delta \tau \) | \( n \) | \( \varepsilon_{\text{sol}} \) | \( k_{\text{max}} \) | \( \varepsilon_{\text{sol}} \) | \( k_{\text{max}} \) | \( \varepsilon_{\text{sol}} \) |
|---|---|---|---|---|---|---|
| 0.1 | 6 | \( 9.66 \times 10^{-5} \) | 26 | \( 9.66 \times 10^{-5} \) | 11 | \( 5.33 \times 10^{-2} \) |
| | 12 | \( 1.03 \times 10^{-10} \) | 20 | \( 4.87 \times 10^{-11} \) | 8 | \( 3.64 \times 10^{-9} \) |
| | 18 | \( 1.02 \times 10^{-10} \) | 20 | \( 3.34 \times 10^{-11} \) | 7 | \( 2.09 \times 10^{-13} \) |
| 1.0 | 15 | INF | 75 | 0.32 | 75 | 0.98 |
| | 30 | \( 1.23 \times 10^{-7} \) | 150 | \( 6.28 \times 10^{-10} \) | 22 | \( 2.84 \times 10^{-8} \) |
| | 45 | \( 7.42 \times 10^{-7} \) | 225 | \( 2.21 \times 10^{-11} \) | 17 | \( 8.23 \times 10^{-14} \) |

Table 3: Solution error for various methods of solving the one-dimensional model problem (3.4) with \( t_\alpha = 0 \). In each case, the problem is solved over \([0, 25]\) in intervals \([\tau_j, \tau_{j+1}]\) of size \( \Delta \tau \). On each interval, \( n \) quadrature points are used. The variable \( \varepsilon_{\text{sol}} \) records the worst-case error at any quadrature point (3.16) and \( k_{\text{max}} \) records the maximum number of iterations needed at any step, if applicable. For both iterative methods, the tolerance for convergence is set to \( 10^{-10} \) with a maximum of \( 5n \) iterations allowed. Here, INF means the method diverged. The power expansion is omitted from this table since it diverged for each case.

The DVR is the only method capable of rivaling the accuracy of ITVOLT (Gauss-Seidel). Moreover, it appears to respond best to increasing \( n \), consistently improving until the error reaches \( 10^{-14} \) in magnitude. The main drawback to a DVR approach is the need to solve an \((n-1) \times (n-1)\) system of equations on each interval, which scales poorly as the problem grows in dimension. In particular, an ODE in two variables expanded in \( n \) and \( m \) basis functions in the two variables respectively would require solving a system of size \( O(nm) \).

Together, these results make a compelling case for our method, which is able to avoid both the instability of the power expansion and the computational costs of the DVR system solve.
4 Application to the Time-Dependent Schrödinger Equation

We turn now to a particular physical application of interest. In atomic units, the time-dependent Schrödinger equation (TDSE) is

\[
\frac{i}{\hbar} \frac{\partial}{\partial t} \psi(t) = H(t) \psi(t), \tag{4.1}
\]

for a Hamiltonian \(H(t)\) and corresponding wave function \(\psi\). This first order differential equation governs the evolution of any quantum mechanical system. For more background information about the importance of the TDSE as well as its derivation, see the standard reference [8].

The formal solution to (4.1) satisfies \(\psi(t) = U(t, t_0)\psi(t_0)\) for \(U\) a time evolution operator given by the time-ordered Dyson series

\[
U(t, t_0) = 1 + \sum_{n=1}^{\infty} (-i)^n \int_{t_0}^{t} dt_1' \int_{t_0}^{t_1'} dt_2' \cdots \int_{t_0}^{t_{n-1}} dt_n' H(t_1')H(t_2') \cdots H(t_n') \tag{4.2}
\]

with \(t > t_1' > \cdots t_n' > t_0\). If \(H\) commutes with itself at different times, this series simplifies to

\[
U(t, t_0) = e^{-i \int_{t_0}^{t} H(t')dt'}. \tag{4.3}
\]

Moreover, when \(H\) is time-independent and therefore trivially commutes with itself, \(U(t, t_0) = e^{-iH(t-t_0)}\). Though perhaps less obvious in the general case, \(U(t, t_0)\) is always a unitary operator.

Typically, there is no analytic way to evaluate (4.2), leaving numerical methods as the only option for constructing solutions to (4.1). Researchers who use these solutions for modeling or simulations often require highly accurate results on incredibly large problems. See for example work in molecular dynamics [17].

A common starting point is a short-time approximation, where (4.1) is solved over an interval \([t_0, t_f]\) by assuming that \(H\) is time-independent and equal to \(\tilde{H} = H(\frac{t_0 + t_f}{2})\), implying \(\psi(t) = e^{-i\tilde{H}(t-t_0)}\psi(t_0)\). The computational challenge of this approach is twofold. First, the assumption that \(H\) is time-independent on \([t_0, t_f]\) is likely only realistic if the interval is sufficiently small, meaning the method may be expensive despite being explicit. Second, the matrix exponential involving \(\tilde{H}\) must be evaluated, which is a complicated numerical question of its own. For an in-depth summary of common approaches for handling matrix exponentials, specifically in the context of the TDSE, see Gharibnejad et al. [7]. Common numerical methods of higher accuracy include fourth-order Runge-Kutta (RK4) [21], which avoids matrix exponentials altogether, and the Magnus expansion [3].

In the remainder of this section, we outline how ITVOLT can be applied to the TDSE and compare it

| \(\Delta t\) | \(n\) | \(\varepsilon_{sol}\) | \(k_{\text{max}}\) | \(\varepsilon_{sol}\) | \(k_{\text{max}}\) | \(\varepsilon_{sol}\) | \(k_{\text{max}}\) | \(\varepsilon_{sol}\) |
|---|---|---|---|---|---|---|---|---|
| 0.1 | 3 | 7.81 \times 10^{-7} | 2 | 7.81 \times 10^{-7} | 2 | 7.81 \times 10^{-7} | 2 | 7.81 \times 10^{-7} |
| | 6 | 8.96 \times 10^{-13} | 4 | 8.94 \times 10^{-13} | 3 | 1.89 \times 10^{-8} | 30 | 2.48 \times 10^{-8} |
| | 12 | 8.79 \times 10^{-13} | 4 | 8.79 \times 10^{-13} | 3 | INF | 60 | 1.12 \times 10^{-12} |
| 1.0 | 5 | 4.66 \times 10^{-5} | 8 | 4.66 \times 10^{-5} | 5 | 4.66 \times 10^{-5} | 8 | 4.66 \times 10^{-5} |
| | 15 | 1.48 \times 10^{-12} | 7 | 2.82 \times 10^{-14} | 5 | INF | 75 | 5.87 \times 10^{-14} |
| | 30 | 1.45 \times 10^{-12} | 7 | 5.60 \times 10^{-14} | 4 | INF | 150 | 8.22 \times 10^{-14} |
to a recent method proposed by Ndong et al. [15] and studied further by Schaefer et al. [19]. In their paper, Ndong et al. define an iterative method that, like ours, is based on a Volterra integral equation representation of the TDSE. Both approaches are similar in spirit, but differ substantially in the numerical details. To easily compare the two approaches, we employ much of the same notation as in [15].

We start by decomposing \( H \) as \( H(t) = H_0 + W(t) \) for a time-independent piece \( H_0 \) and a time-dependent piece \( W(t) \). On the interval \([\tau_j, \tau_{j+1}]\), this allows us to rewrite (4.1) as

\[
\frac{\partial}{\partial t} \psi(t) = \left[ H_0 + W \left( \frac{\tau_j + \tau_{j+1}}{2} \right) \right] \psi(t) + \left[ W(t) - W \left( \frac{\tau_j + \tau_{j+1}}{2} \right) \right] \psi(t). \tag{4.4}
\]

Here, we have taken a cue from our model problem by adding and subtracting the value of the time-dependent piece of the Hamiltonian at the midpoint of the interval. Setting \( H_j = H_0 + W \left( \frac{\tau_j + \tau_{j+1}}{2} \right) \) and \( V_j(t) = W(t) - W \left( \frac{\tau_j + \tau_{j+1}}{2} \right) \) and integrating, we obtain the Volterra integral equation

\[
\psi(t) = e^{-iH_j(t-\tau_j)} \psi(\tau_j) - i \int_{\tau_j}^{t} e^{-iH_j(t-t')} V_j(t') \psi(t') dt'. \tag{4.5}
\]

Choosing quadrature points \( t_1, \ldots, t_n \) in \([\tau_j, \tau_{j+1}]\), we then have Jacobi iteration

\[
\psi^{(k+1)}(t_p) = e^{-iH_j(t_p-\tau_j)} \psi^{(k)}(\tau_j) - i \sum_{l=1}^{n} e^{-iH_j(t_{p-l})} V_j(t_l) w_{p,l} \psi^{(k)}(t_l), \tag{4.6}
\]

and Gauss-Seidel iteration

\[
(I + w_{p,p} V_j(t_p)) \psi^{(k+1)}(t_p) = e^{-iH_j(t_p-\tau_j)} \psi^{(k+1)}(\tau_j) - i \sum_{l=1}^{p-1} e^{-iH_j(t_{p-l})} V_j(t_l) w_{p,l} \psi^{(k+1)}(t_l)
- i \sum_{l=p+1}^{n} e^{-iH_j(t_{p-l})} V_j(t_l) w_{p,l} \psi^{(k)}(t_l). \tag{4.7}
\]

As usual, (4.7) is a system of equations with size equal to the number of states in the representation of \( \psi \).

In (4.4) above, we added and subtracted the value of \( W \) at the midpoint of \([\tau_j, \tau_{j+1}]\) in the hopes that it would better incorporate the time dependence of \( H \) into (4.1). By doing this, the inhomogeneous term in (4.5) becomes a short-time approximation to \( \psi(t) \). This is likely a better starting point to our iteration than \( \psi(\tau_j) \), lending support to the idea that, as in the model problem, making this addition and subtraction should improve convergence.

Unlike the model problem, however, this comes at the cost of evaluating a matrix exponential, which now appears repeatedly in both versions of the iteration. In principle, any of the many established approaches for handling matrix exponentials work with our method, and we do not specify a particular choice. In the following examples, we will discuss how the method performs with a few of the more common approaches - specifically full diagonalization, Lanczos iteration, and Chebyshev propagation.

Note that the Volterra integral equation (4.5) is the same expression iterated by Ndong et al. [15] in their method. The key difference between their work and ours is the way in which the integral is handled. Where we apply a simple Gauss-Lobatto quadrature, Ndong et al. instead expand \( V_j(t) \psi(t) \) in Chebyshev polynomials, convert this expansion to a power series, and integrate the product of the matrix exponential and the power series analytically. While both Ndong et al. and subsequently Schaefer et al. [19] demonstrate that their approach is capable of high accuracy, we believe ITVOLT can do the same while being computationally simpler.

## 5 Physical Examples

In this section, we use ITVOLT to solve the TDSE for two quantum mechanical systems: a two-level atom exposed to a laser and a linearly driven harmonic oscillator. In both cases, solutions are known analytically, allowing for a precise quantification of accuracy. These problems are also handled by Ndong et al. [15], and we comment throughout where the setup of the problems differs and how the results compare. Note that, as in the previous section, all equations are presented in atomic units.
5.1 Two-Level Atom

We start with the simpler two-level problem. In the rotating wave approximation, the Hamiltonian for a two-level atom driven resonantly by a laser field $E(t)$ with transition dipole strength 1 a.u. is

$$H(t) = \begin{pmatrix} 0 & E(t) \\ E(t) & 0 \end{pmatrix}. \quad (5.1)$$

If we decompose this Hamiltonian as $H(t) = H_0 + W(t)$ as done above, $H_0 = 0$. As mentioned earlier, running either version of our method requires computing matrix exponentials involving $H_j = H(\frac{\tau_j + \tau_{j+1}}{2})$. For this problem, since any anti-diagonal matrix can be easily diagonalized, these exponentials can be done analytically. In particular,

$$e^{-iH_j(t-t')} = \begin{pmatrix} \cos(\theta) & -i\sin(\theta) \\ -i\sin(\theta) & \cos(\theta) \end{pmatrix}, \quad \theta = E\left(\frac{\tau_j + \tau_{j+1}}{2}\right)(t-t'). \quad (5.2)$$
For our tests, we solve the TDSE on $[0, T]$ with a laser pulse of the form

$$E(t) = \frac{1}{2} E_0 \sin^2 \left( \frac{\pi t}{T} \right),$$

for $E_0$ a field amplitude. In this setting, the analytic solution is $\psi(t) = (c_g(t), c_e(t))^T$ for ground and excited states

$$c_g(t) = \cos \left[ \frac{1}{4} E_0 \left( t - \frac{T}{2\pi} \sin \left( \frac{2\pi t}{T} \right) \right) \right],$$

$$c_e(t) = -i \sin \left[ \frac{1}{4} E_0 \left( t - \frac{T}{2\pi} \sin \left( \frac{2\pi t}{T} \right) \right) \right].$$

Figure 2 shows $E(t)$ and the corresponding ground and excited state population probabilities for different choices of $E_0$. As anticipated, increasing the amplitude also increases the number of transitions between the two states; for this reason, we expect the problem to be more difficult to solve numerically as the pulse amplitude grows.

Similar to the earlier examples, we measure accuracy by tracking the worst-case error in the population probabilities of each state at any quadrature point used in $[0, T]$. That is, if $\psi^{(k)}(t) = (c_1^{(k)}(t), c_2^{(k)}(t))^T$ is the converged result, we compute

$$\epsilon_1^{(k)} = \max_j \left[ \max_{t \in [\tau_j, \tau_{j+1}]} \left| c_1^{(k)}(t) - |c_g(t)|^2 \right|^2 \right],$$

$$\epsilon_2^{(k)} = \max_j \left[ \max_{t \in [\tau_j, \tau_{j+1}]} \left| c_2^{(k)}(t) - |c_e(t)|^2 \right|^2 \right].$$

We once again also track the maximum number of iterations required to reach convergence at any step in the propagation, $k_{\text{max}}$. Lastly, we include system run times to quantify efficiency.

Table 5 shows this data for both versions of ITVOLT with a variety of choices of $\Delta \tau$, the length of the intervals $[\tau_j, \tau_{j+1}]$, and $n$, the number of quadrature points used. We see here many of the same patterns illustrated by the previous examples. As in the model two-channel problem, adding quadrature points consistently improves the results and can even reduce the number of iterations required to reach convergence. Similar to the model ODE, the Gauss-Seidel version shows additional benefits over Jacobi when the step size is large.

In most cases, the Gauss-Seidel version requires significantly fewer iterations to converge than Jacobi. With the system times included, we can see that this does offer efficiency improvements, though not in all cases. It is worth noting that the implementation of the Jacobi iteration used was not parallelized; consequently, the run times listed can likely be improved, possibly eliminating any efficiency benefits of Gauss-Seidel.

We cannot make a direct comparison between Table 5 and the results of Ndong et al. [15] since their paper does not list the pulse amplitude used. Nevertheless, the rest of the setup is followed exactly, including the use of a pulse that ensures $c_g(T) = 0$ and $c_e(T) = 1$. One comparison we can make, however, is to a short-time approximation. As shown in Table 6, this approach, even with the exponentials done exactly via diagonalization, requires an incredibly small time step to achieve high accuracy.

### 5.2 Driven Harmonic Oscillator

Our final test is the most computationally demanding so far: the TDSE for a driven harmonic oscillator with frequency $\omega = 1$ a.u. The Hamiltonian for this system is

$$H(x, t) = -\frac{1}{2} \frac{\partial^2}{\partial x^2} + \frac{1}{2} x^2 + xE(t),$$

for a field

$$E(t) = E_0 \sin^2 \left( \frac{\pi t}{T} \right) \cos(\omega_0 t).$$

---

12
Two Level Atom: Solution Error

| $\Delta \tau$ | $n$ | $\varepsilon_{sol}^1$ | $\varepsilon_{sol}^2$ | $k_{max}$ | System time | $\varepsilon_{sol}^1$ | $\varepsilon_{sol}^2$ | $k_{max}$ | System time |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| 10 | 3 | $1.78 \times 10^{-6}$ | $1.80 \times 10^{-6}$ | 3 | 0.012 s | $1.78 \times 10^{-6}$ | $1.80 \times 10^{-6}$ | 3 | 0.023 s |
| | 6 | $3.40 \times 10^{-13}$ | $3.38 \times 10^{-13}$ | 4 | 0.015 s | $3.38 \times 10^{-13}$ | $3.42 \times 10^{-13}$ | 3 | 0.017 s |
| 20 | 4 | $2.53 \times 10^{-8}$ | $2.56 \times 10^{-8}$ | 4 | 0.007 s | $2.53 \times 10^{-8}$ | $2.56 \times 10^{-8}$ | 3 | 0.015 s |
| | 8 | $3.31 \times 10^{-13}$ | $3.28 \times 10^{-13}$ | 4 | 0.027 s | $3.31 \times 10^{-13}$ | $3.25 \times 10^{-13}$ | 4 | 0.018 s |
| 50 | 5 | $1.27 \times 10^{-6}$ | $1.33 \times 10^{-6}$ | 6 | 0.007 s | $1.27 \times 10^{-6}$ | $1.33 \times 10^{-6}$ | 4 | 0.014 s |
| | 10 | $6.85 \times 10^{-13}$ | $8.20 \times 10^{-13}$ | 6 | 0.025 s | $3.33 \times 10^{-13}$ | $3.33 \times 10^{-13}$ | 4 | 0.013 s |
| 100 | 6 | $1.47 \times 10^{-6}$ | $1.49 \times 10^{-6}$ | 8 | 0.005 s | $1.47 \times 10^{-6}$ | $1.49 \times 10^{-6}$ | 5 | 0.005 s |
| | 12 | $3.27 \times 10^{-12}$ | $3.45 \times 10^{-12}$ | 8 | 0.013 s | $4.76 \times 10^{-13}$ | $5.69 \times 10^{-13}$ | 5 | 0.013 s |
| 300 | 9 | $3.54 \times 10^{-4}$ | $3.60 \times 10^{-4}$ | 18 | 0.006 s | $3.54 \times 10^{-4}$ | $3.60 \times 10^{-4}$ | 9 | 0.011 s |
| | 18 | $7.37 \times 10^{-11}$ | $7.54 \times 10^{-11}$ | 15 | 0.023 s | $7.31 \times 10^{-11}$ | $7.86 \times 10^{-11}$ | 8 | 0.016 s |
| 900 | 20 | INF | INF | 60 | 0.016 s | $2.11 \times 10^{-2}$ | $3.04 \times 10^{-2}$ | 38 | 0.028 s |
| | 40 | $1.78 \times 10^{-9}$ | $1.87 \times 10^{-9}$ | 51 | 0.058 s | $1.81 \times 10^{-9}$ | $1.85 \times 10^{-9}$ | 18 | 0.043 s |

Table 5: Population probability error (5.5) for the Gauss-Seidel and Jacobi versions of ITVOLT applied to the two-level atom with a field amplitude $E_0 = 2\pi/9$. Here, $n$ quadrature points were used over each time step $[\tau_j, \tau_{j+1}]$ of size $\Delta \tau$ for a total time of $T = 9000$ a.u. The variable $k_{max}$ is the maximum number of iterations needed at any time step to reach convergence, where the tolerance was set to $10^{-10}$. 3n was the maximum number of iterations allowed if this tolerance was not met.

Two Level Atom: Short Time Approximation

| $\Delta \tau$ | $\varepsilon_{sol}^1$ | $\varepsilon_{sol}^2$ | System time |
| --- | --- | --- | --- |
| 0.01 | $3.16 \times 10^{-8}$ | $3.16 \times 10^{-8}$ | 1.858 s |
| 0.1 | $5.07 \times 10^{-8}$ | $5.07 \times 10^{-8}$ | 0.193 s |
| 1.0 | $5.07 \times 10^{-6}$ | $5.07 \times 10^{-6}$ | 0.030 s |
| 10 | $5.07 \times 10^{-4}$ | $5.07 \times 10^{-4}$ | 0.010 s |
| 100 | $5.06 \times 10^{-2}$ | $5.06 \times 10^{-2}$ | 0.004 s |

Table 6: Solution error for the two-level atom via a short-time approximation. As in the previous examples, $T = 9000$ and $E_0 = 2\pi/9$. Note that no quadrature points are used here, so the errors are only computed at the right endpoint of each interval $[\tau_j, \tau_{j+1}]$ of size $\Delta \tau$. 


As in the previous example, $E_0$ is the amplitude of the pulse and $T$ is the final propagation time, set here to be 100 a.u. $\omega_0$ is the driving frequency. We’ll consider two choices for this frequency: $\omega_0 = 0$ and $\omega_0 = 1$ a.u. In the latter case, $\omega_0$ is equal to the spacing between energy levels of the unforced oscillator, leading to an increase in transitions during the interaction. Figure 3 shows $E(t)$ for both choices of $\omega_0$ with $E_0 = 1$.

One of the challenges of this problem is the inclusion of a spatial variable; at each time $t$, we need to decide how to represent the Hamiltonian (5.6) in space. Ndong et al. [15] choose to do this by representing $H$ on a Fourier grid, replacing the spatial derivative with a finite difference approximation over the grid. While our method could handle the matrix representation of $H$ obtained this way, we choose instead to expand the wave function $\psi(x,t)$ in the eigenfunctions of the unforced harmonic oscillator. That is,

$$\psi(x,t) = \sum_{n=0}^{m} c_n(t) \psi_n(x),$$

(5.8)

where

$$\psi_n(x) = \frac{1}{\pi^{1/4}\sqrt{2^n n!}} e^{-\frac{1}{2} x^2} H_n(x), \quad n = 0, 1, 2, \cdots$$

(5.9)

for $H_n(x)$ the $n^{th}$ Hermite polynomial. Each of these eigenfunctions satisfies the time-independent Schrödinger equation with $E_n = \frac{2n+1}{2}$. If $c(t) = (c_0(t) \cdots c_m(t))^T$ this allows us to convert the TDSE to a set of coupled differential equations in time of the form

$$\frac{\partial}{\partial t} c(t) = H(t)c(t),$$

(5.10)

for the matrix

$$H(t) = \begin{bmatrix}
E_0 & \frac{1}{\sqrt{2}} E(t) \\
\frac{1}{\sqrt{2}} E(t) & E_1 & E(t) \\
E(t) & E_2 & \sqrt{2} E(t) \\
& \ddots & \ddots & \ddots \\
& & \sqrt{\frac{m-1}{2}} E(t) & E_{m-1} & \sqrt{\frac{m}{2}} E(t) \\
& & & \sqrt{\frac{m}{2}} E(t) & E_m
\end{bmatrix}.$$  

(5.11)

The tridiagonal structure of $H$ comes from the fact that $x$ acts on the eigenfunctions $\psi_n(x)$ as a ladder operator.
Figure 4: First four population probabilities (5.15) for the driven harmonic oscillator with $E_0 = 1$.

If $\psi(0, x) = \psi_n(x)$, the solution to the TDSE is known analytically:

$$\Psi_n(x, t) = N_n e^{ip_0(t)x} e^{-i \Delta E_n t} e^{-\frac{1}{2} \alpha^2 (x-x_0(t))^2} H_n(\alpha(x-x_0(t))).$$  (5.12)

In this solution, $x_0(t)$ and $p_0(t) = x'_0(t)$ are the position and momentum of a classical forced harmonic oscillator satisfying

$$\frac{\partial^2}{\partial t^2} x_0(t) + x_0(t) = -E(t),$$  (5.13)

with initial conditions $x_0(0) = 0 = x'_0(0)$. Similarly, $\delta(t)$ is the classical Lagrangian for an unforced harmonic oscillator with the unforced position replaced by $x_0(t)$. Lastly, $N_n$ and $\alpha$ are normalization constants. Details for how this wave function is derived can be found in [11].

For our tests, we assume $\psi(x, 0) = \psi_0(x)$, in which case $\Psi_0$ simplifies and the probability that the wave function is in the ground state of the unforced harmonic oscillator as a function of time can be computed exactly as

$$P_0(t) = \left| \int_{-\infty}^{\infty} \Psi_0(x, t) \psi_0(x) dx \right|^2 = \left| e^{\frac{i}{2} (x_0(t) + ip_0(t))^2 - \frac{1}{2} x_0(t)^2} \right|^2. \quad (5.14)$$

More generally, the probability that the system is in the $n$-th state of the unforced oscillator is

$$P_n(t) = \left\| \Psi_n(x, t) \psi_0(x) dx \right\|^2 = \left| e^{\frac{i}{2} (x_0(t) + ip_0(t))^2 - \frac{1}{2} x_0(t)^2} \right|^2. \quad (5.15)$$

To evaluate these at any time $t$ we need only compute $x_0(t)$ and $p_0(t)$, which can be done analytically using the Green’s function for the classical forced harmonic oscillator equation. Figure 4 shows the first four transition probabilities with $E_0 = 1$ and $\omega_0 = 0$ or $\omega_0 = 1$. From these, we immediately see how much more challenging the $\omega_0 = 1$ problem is: where a pulse amplitude of $E_0 = 1$ fails to deplete the ground state when $\omega_0 = 0$, it quickly excites the entire population beyond the first four states when $\omega_0 = 1$. For this reason, we will only consider the $\omega_0 = 1$ case in our subsequent numerical tests.

As in the previous example, we solve the TDSE on $[0, T]$ in equally spaced subintervals of the form $[\tau_j, \tau_{j+1}]$. To measure accuracy, we compute the worst-case error in the ground state population probability at the endpoints of each interval, which can be computed easily from the converged coefficients $c(t)$ as

$$\varepsilon_{\text{sol}} = \max_j \left| \left| c_0(\tau_j) \right|^2 - P_0(\tau_j) \right|. \quad (5.16)$$

Since this is a many state problem, we also compute the worst-case deviation from unity at any point in the propagation, i.e.

$$\varepsilon_{\text{norm}} = \max_j \left( 1 - \left| \left| c(\tau_j) \right|^2 \right| \right). \quad (5.17)$$
Above, we use a LAPACK routine [1] to diagonalize matrices. In this approach, all matrix exponentials were handled via full diagonalization. In this section, we consider replacing the diagonalization with one of the following. For a more detailed description of these methods as well as a comparison with other techniques, see [7].

1. Lanczos Iteration: For an iterative approach, we can apply \( e^{-i \mathbf{H}_j (t-t')} \) to a vector \( \mathbf{v} \) by approximating it in a Krylov subspace. To do this, we start by building an orthonormal basis \( \mathbf{q}_1, \mathbf{q}_2, \ldots, \mathbf{q}_m \) such that
\[
\text{span} \{ \mathbf{q}_1, \mathbf{q}_2, \ldots, \mathbf{q}_m \} = \text{span} \{ \mathbf{v}, \mathbf{H}_j \mathbf{v}, \mathbf{H}_j^2 \mathbf{v}, \ldots, \mathbf{H}_j^{m-1} \mathbf{v} \}.
\]
Since \( \mathbf{H}_j \) is symmetric, we can find the vectors \( \mathbf{q}_k \) via a simple three-term recurrence relation, beginning with \( \mathbf{q}_1 = \mathbf{v}/||\mathbf{v}||_2 \). Forming the matrix \( \mathbf{Q} \) whose columns are \( \mathbf{q}_1, \ldots, \mathbf{q}_m \), we then have
\[
e^{-i \mathbf{H}_j (t-t')} \mathbf{v} \approx \mathbf{Q} e^{-i \mathbf{H}_j^{(m)} (t-t')} \mathbf{Q}^T \mathbf{v},
\]
for \( \mathbf{H}^{(m)} \) an \( m \times m \) tridiagonal matrix. Since \( m \) is much smaller than the size of \( \mathbf{H}_j \), the inner exponential can be done via diagonalization. The parameters for the method are a maximum number of iterations and a convergence criteria. At each step, the difference between the approximation using \( k \) vectors and \( k-1 \) vectors is measured; if this error does not fall below the convergence criteria another vector is added, stopping once the maximum number of iterations is reached.

2. Chebyshev Propagation: Alternatively, we can expand the exponential \( e^{-i \mathbf{H}_j (t-t')} \) in Chebyshev polynomials as
\[
\sum_n (2 - \delta_{mn}) e^{-i (\mathbf{\Phi} + \lambda_{\text{min}}(\mathbf{H}_j))(t-t')} J_n \left( \frac{\Delta}{2} (t-s) \right) T_n (-i \mathbf{H}_j^{\text{norm}}).
\]

Table 7: Solution \((5.16)\) and norm \((5.17)\) errors for ITVOLT applied to the driven harmonic oscillator. In each case, \( E_0 = 1, \omega_0 = 1, T = 100 \) a.u., and 200 states were used in the expansion. As in the previous examples, a quadrature points were used on each interval of size \( \Delta \tau \), iterating with a convergence criteria of \( 10^{-10} \). Here, all matrix exponentials were handled with full diagonalization.

| \( \Delta \tau \) | \( n \) | \( \varepsilon_{\text{sol}} \) | \( \varepsilon_{\text{norm}} \) | \( k_{\text{max}} \) | \( \varepsilon_{\text{sol}} \) | \( \varepsilon_{\text{norm}} \) | \( k_{\text{max}} \) |
|----------------|------|-----------------|-----------------|-------------|-----------------|-----------------|-------------|
| \( 10^{-2} \) | 3    | \( 5.82 \times 10^{-12} \) | \( 1.40 \times 10^{-8} \) | 2 | \( 5.82 \times 10^{-12} \) | \( 1.40 \times 10^{-8} \) | 2 |
|       | 6    | \( 2.71 \times 10^{-13} \) | \( 7.85 \times 10^{-13} \) | 3 | \( 2.74 \times 10^{-13} \) | \( 7.69 \times 10^{-13} \) | 3 |
| \( 10^{-1} \) | 5    | \( 9.60 \times 10^{-15} \) | \( 1.38 \times 10^{-9} \) | 5 | \( 8.05 \times 10^{-15} \) | \( 1.38 \times 10^{-9} \) | 4 |
|       | 10   | \( 7.55 \times 10^{-15} \) | \( 8.33 \times 10^{-14} \) | 5 | \( 7.99 \times 10^{-15} \) | \( 6.59 \times 10^{-14} \) | 4 |
| 1.0   | 10   | \( 1.19 \times 10^{-13} \) | \( 4.77 \times 10^{-6} \) | 22 | \( 6.62 \times 10^{-15} \) | \( 4.77 \times 10^{-6} \) | 10 |
|       | 20   | \( 1.19 \times 10^{-13} \) | \( 3.30 \times 10^{-12} \) | 20 | \( 1.55 \times 10^{-15} \) | \( 3.37 \times 10^{-12} \) | 8 |

Note that these maximum errors are taken over the endpoints of the intervals; we do not include the quadrature points as in the previous examples to allow for a later comparison to RK4.

While this is a highly accurate method, it is computationally demanding and therefore impractical, even for a tridiagonal matrix.

To explore how these results change if a different method for evaluating matrix exponentials is used, we consider replacing the diagonalization with one of the following. For a more detailed description of these methods as well as a comparison with other techniques, see [7].
Table 8: Solution (5.16) and norm (5.17) errors for ITVOLT (Gauss-Seidel) with different methods for computing matrix exponentials. For each $\Delta \tau$, $n$ is the number of quadrature points used. Lanczos is run with a convergence criteria of $10^{-12}$ and a maximum of 25 iterations. The Chebyshev propagator is done with a maximum of 1000 terms, truncating the expansion once the coefficients fall below $10^{-15}$ in modulus. In all cases, $E_0 = 1$, $\omega_0 = 1$, $T = 100$ a.u., 200 states are used in the expansion, and the iteration was run with a convergence criteria of $10^{-10}$ for a maximum of 5$n$ iterations.

| $\Delta \tau$ ($n$) | $\varepsilon_{\text{sol}}$ | $\varepsilon_{\text{norm}}$ | System time | $\varepsilon_{\text{sol}}$ | $\varepsilon_{\text{norm}}$ | System time | $\varepsilon_{\text{sol}}$ | $\varepsilon_{\text{norm}}$ | System time |
|---------------------|-----------------|-------------------|--------------|-----------------|-------------------|--------------|-----------------|-------------------|--------------|
| $10^{-2}$ (6)       | $4.14 \times 10^{-13}$ | $1.79 \times 10^{-12}$ | 56.63 s      | $3.22 \times 10^{-13}$ | $1.28 \times 10^{-13}$ | 34.60 s      | $2.74 \times 10^{-13}$ | $7.69 \times 10^{-13}$ | 197.17 s     |
| $10^{-1}$ (10)      | $4.95 \times 10^{-14}$ | $1.86 \times 10^{-13}$ | 79.17 s      | $1.07 \times 10^{-14}$ | $1.05 \times 10^{-14}$ | 23.89 s      | $7.99 \times 10^{-15}$ | $6.59 \times 10^{-14}$ | 54.29 s      |
| 1.0 (20)            | $765.15$         | $22013.73$        | 206.49 s     | $7.49 \times 10^{-15}$ | $3.38 \times 10^{-12}$ | 54.94 s      | $1.55 \times 10^{-15}$ | $3.37 \times 10^{-12}$ | 33.88 s      |

Table 9: Solution (5.16) and norm (5.17) error for RK4 applied to the driven harmonic oscillator. In each case, $E_0 = 1$, $\omega_0 = 1$, $T = 100$ a.u. and 200 functions are used in the expansion.

| $\Delta \tau$ | $\varepsilon_{\text{sol}}$ | $\varepsilon_{\text{norm}}$ | System time |
|--------------|-----------------|-------------------|--------------|
| $10^{-6}$    | $3.77 \times 10^{-10}$ | $6.35 \times 10^{-13}$ | 1988.70 s     |
| $10^{-5}$    | $3.73 \times 10^{-11}$ | $1.27 \times 10^{-12}$ | 199.24 s      |
| $10^{-4}$    | $2.28 \times 10^{-12}$ | $1.23 \times 10^{-7}$  | 20.39 s       |
| $10^{-3}$    | $5.45 \times 10^{-13}$ | $1.22 \times 10^{-2}$  | 2.06 s        |
| $10^{-2}$    | $3.27 \times 10^{-10}$ | $1.00$             | 0.22 s        |

In this expression, $\Delta = \lambda_{\text{max}}(H_j) - \lambda_{\text{min}}(H_j)$ for $\lambda_{\text{max}}(H_j)$ and $\lambda_{\text{min}}(H_j)$ the largest and smallest eigenvalues of $H_j$ respectively and $J_n$ is the $n^{th}$ Bessel function of the first kind. Note that to apply this expansion to a vector, we can exploit the recurrence relationship for the Chebyshev polynomials to save on matrix/vector multiplications. As with Lanczos, this method takes two parameters: a coefficient threshold and a maximum number of terms. As the expansion coefficients are computed we check their magnitude, truncating the expansion once the last coefficient falls below the threshold in magnitude or the maximum number of terms is reached.

Table 8 repeats a few of the cases from Table 7 for ITVOLT (Gauss-Seidel) with these methods. As is clear from the results, our method with both Lanczos and Chebyshev is capable of reproducing the high accuracy of full diagonalization, although Lanczos is limited to smaller step sizes. Both also show efficiency improvements in certain cases, in particular Chebyshev which achieves highly accurate results in the shortest amount of time. These results indicate that ITVOLT is viable for larger problems, as more efficient methods for handling the matrix exponentials can be employed without sacrificing accuracy.

As a final assessment of our results on this problem, we compare with RK4. Table 9 shows the solution and norm errors for RK4 with different step sizes. As is clear from the data, while able to fairly accurately compute the ground state probability, it struggles to preserve unit norm. Neither ITVOLT nor RK4 is explicitly unitary, so this indicates that our method is better able to approximate the probabilities for all states in the expansion. To get a feel for how the error evolves over the propagation, Figure 5 plots the solution and norm error for both versions of ITVOLT and RK4. Note that the results shown for RK4 correspond to a step size of $10^{-5}$. While RK4 is the least accurate in both plots, it is particularly bad for the norm error, consistently running at least one order of magnitude above the others. The norm error also
Figure 5: Evolution of the solution (5.16) and norm (5.17) errors for RK4 and both versions of ITVOLT applied to the harmonic oscillator with $\omega_0 = 1$ and $E_0 = 1$. RK4 was done with a step size of $10^{-5}$ while the Jacobi and Gauss-Seidel iterations were done with $\Delta \tau = 10^{-1}$, $n = 10$, and a convergence criteria of $10^{-10}$. For the latter two, matrix exponentials were done via the Chebyshev propagator.

exhibits some differentiation between the Jacobi and Gauss-Seidel versions of ITVOLT. This echoes something we have seen throughout all of our examples: on easy problems the two variants produce nearly identical results, while on more difficult problems Gauss-Seidel outperforms Jacobi.

6 Conclusions and Future Work

In this paper, we have constructed a simple iterative method for solving Volterra integral equations of the second kind. Based on Gauss-Lobatto quadrature, our method is both flexible and simple to implement. Moreover, as shown in the previous sections, it is capable of accurately and efficiently solving a variety of problems, outperforming a number of established techniques.

Based on the data presented, we make the following observations, which we hope guide researchers interested in applying ITVOLT to their own problems.

1. If solving a problem over multiple intervals (as done for both TDSE examples and the model ODE) ITVOLT performs best for a medium step size, where the integral contributes significantly to the iteration but does not require too many quadrature points for accurate results.

2. Up to a certain point, increasing the number of quadrature points used improves the results and can even cut down on the number of iterations required for convergence.

3. Unless the Jacobi iteration is implemented in an explicitly parallel way, the Gauss-Seidel version is likely to be both more accurate and roughly as efficient if not more so.

Current efforts are focused on implementing ITVOLT on even larger problems, in particular the TDSE for the three dimensional hydrogen atom. In future work, we plan to quantify how our approach compares to high order methods designed specifically to handle the TDSE, in particular the Magnus expansion.
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