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Integrators for Quantum Dynamics: A Numerical Analyst's Brief Review

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This note discusses and compares – in theoretical respects – various old and new approaches to numerical time integration for quantum dynamics: implicit vs. exponential midpoint rule; splitting, Chebyshev and Lanczos approximations to the exponential; Magnus integrators; integrators for almost-adiabatic quantum dynamics.

1 Introduction

This paper gives a concise review of numerical integrators for time-dependent Schrödinger equations

$$i\dot{\psi}(t) = H(t)\psi(t)\ , \qquad \psi(0) = \psi_0\ . \tag{1}$$

Numerical difficulties in the solution of such problems are due both to discretizing or modeling in space (which is not considered here) and to discretization in time, on which the focus is put in the present article. The computational Hamiltonian $H(t)$ is a space discretization or other finite-dimensional model of $H(t) = T + V(t)$ with a kinetic part such as $T = -(2m)^{-1}\Delta_x$, and with a potential $V(t)$ acting as a multiplication operator. Numerical problems are caused by the unbounded nature of the Hamiltonian and the resulting highly oscillatory behaviour of the wave function.

Several new and promising numerical methods have been devised in the last few years, and an improved understanding of well-established methods could be gained. In this review I will put a stronger emphasis on theoretical error bounds than is usual in computational physics articles. This is not done out of mathematical vanity, but because theoretical insight – together with numerical experiments – is important in identifying and comparing merits and flaws of different methods, and in guiding the way to improved methods. It is also useful to question the uncritical use of such universally accepted concepts as "second-order scheme", which may be misleading for the problem at hand.

We start from the classical implicit midpoint rule and compare it with the exponential midpoint rule. The latter method requires computing the exponential of the Hamiltonian applied to a vector, for which we discuss three computational approaches: Splitting, Chebyshev and Lanczos approximations. As a way to enhance the accuracy of the exponential midpoint rule, Magnus integrators are then discussed. In a final section, we turn to novel integrators that are devised for treating almost-adiabatic quantum dynamics.

2 The Implicit Midpoint Rule

Contrary to most of the classical numerical integrators, such as explicit or implicit Runge-Kutta or multistep methods, the *implicit midpoint rule*

$$i\,\frac{\psi_{n+1}-\psi_n}{\Delta t} = H(t_{n+1/2})\,\tfrac{1}{2}(\psi_{n+1}+\psi_n) \tag{2}$$

(with $t_{n+1/2} = \frac{1}{2}(t_{n+1}+t_n)$ and $t_n = n\Delta t$) has a *unitary propagator*:

$$\psi_{n+1} = r\big(-i\Delta t\,H(t_{n+1/2})\big)\psi_n \qquad \text{with} \quad r(z) = \frac{1+\frac{1}{2}z}{1-\frac{1}{2}z}\,. \tag{3}$$

This is an important qualitative feature which the numerical method shares with the exact solution operator. It implies that the method preserves the L^2 norm (or the Euclidean norm in the spatially discretized situation):

$$\|\psi_{n+1}\| = \|\psi_n\|\,,$$

and hence is stable for arbitrary time steps Δt.

A further useful property is the *time reversibility* of the numerical scheme: exchanging $n \leftrightarrow n+1$ and $\Delta t \leftrightarrow -\Delta t$ gives the same numerical method again. In terms of the propagator function $r(z)$, this is reflected by the property

$$r(-z) = r(z)^{-1},$$

which $r(z)$ shares with the exponential e^z.

But what can be said about the accuracy of the implicit midpoint rule? In the classical ODE setting of *bounded* and smooth $H(t)$, it is a well-known fact that the implicit midpoint rule is a *second-order* method, that is, the error satisfies

$$\|\psi_n - \psi(t_n)\| = \mathcal{O}(\Delta t^2) \tag{4}$$

uniformly for $n\Delta t$ in a bounded time interval $[t_0, t_{\rm end}]$. Such statements on the order of a method should, however, be taken with caution in the present context: in our situation of a (spatially discretized) Schrödinger equation (1), the norm of $H(t)$ can be arbitrarily large or even unbounded, and hence the classical numerical ODE theory does not apply. Nevertheless, for the particular case of the implicit midpoint rule it can be shown that the constant hidden in the $\mathcal{O}(\Delta t^2)$ error bound is in fact independent of bounds of $H(t)$. It does depend, however, on bounds of $\dot H$ and $\ddot H$, and on the maximum of the norm of the third time derivative of the solution ψ on the time interval under consideration. This latter dependence on solution derivatives is an unpleasant feature: unless we start from spatially very smooth initial data, the wave function $\psi(t)$ is highly oscillatory in time, and hence higher time derivatives can become large. Good accuracy can then be expected only for very small time steps, and this is indeed what happens here. We emphasize that accuracy, not stability, restricts the time step of the implicit midpoint rule applied to Schrödinger equations. High temporal smoothness, when available, leads to good accuracy also with larger time steps.

3 The Exponential Midpoint Rule

This method is obtained by formally replacing $r(z)$ by $\exp(z)$ in the formula (3) of the implicit midpoint rule:

$$\psi_{n+1} = \exp\bigl(-i\Delta t\, H(t_{n+1/2})\bigr)\psi_n \,. \tag{5}$$

Of course, instead of solving systems of linear equations, we now have to scope about how to compute the exponential of a large matrix. We will consider this aspect in the following sections and assume for the time being that the matrix exponential times a vector can be computed efficiently. We note that the above exponential midpoint rule again has a unitary propagator and it is time-reversible. It offers improved accuracy over the classical implicit midpoint rule: it satisfies a second-order error bound (4), but contrary to before, the constant hidden in the $\mathcal{O}$-notation is now independent of the time derivatives of the wave function $\psi(t)$[5,3]. The result[3] is

$$\|\psi_n - \psi(t_n)\| \le C\,\Delta t^2 \max_{t\in[t_0,t_{\rm end}]} \|D\psi(t)\|$$

under the assumption on the commutators ($[A,B] = AB - BA$)

$$\|[H(t), H(s)]\,\phi\| \le c\,\|D\phi\| \qquad \text{for all } t, s \text{ and } \phi.$$

Since the commutator of the Laplacian with a multiplication operator is a *first-order* differential operator, this condition holds with the gradient operator $D = \nabla_x$ in the spatially continuous case of $H(t) = (2m)^{-1}\Delta_x + V(t)$ with a smooth bounded potential $V(t)$, and with a discrete gradient in cases of spatially discretized problems[8].

This theoretical fact explains – and numerical experiments confirm – that much larger time steps than with the classical implicit midpoint rule can be taken to achieve the same accuracy, in particular in cases of low regularity of the wave function.

4 Strang Splitting

A standard approach to computing the exponential of $H = T + V$ is to use the symmetric splitting (known as Strang splitting or Marchuk splitting or symmetric Trotter splitting in different communities)

$$\exp(-i\Delta t(T+V))\psi \approx \exp(-i\Delta t\,T/2)\exp(-i\Delta t V)\exp(-i\Delta t\,T/2)\psi \,. \tag{6}$$

The right-hand side is often much cheaper to compute. For instance, this is the case when T is a spectral discretization of the negative Laplacian $-(2m)^{-1}\Delta_x$, which is diagonalized by fast Fourier transforms, and V is represented by a diagonal matrix. Only the exponentials of diagonal matrices, which are trivially computed, are required in this situation.

The symmetric splitting is a *second-order* scheme:

$$\|\exp(-i\Delta t T/2)\exp(-i\Delta t V)\exp(-i\Delta t T/2)\psi - \exp(-i\Delta t(T+V))\psi\| = \mathcal{O}(\Delta t^2)\,.$$

Here again, such an order statement must be taken with caution. This error bound is easily obtained by using the series expansion of the exponential, but then the $\mathcal{O}$-term depends on the norms of T and V. Since T is typically a discretized Laplacian, such an estimate is of no use here. A second-order error bound that allows for an unbounded T has been derived

only recently[8]. Under reasonable conditions on the commutators $[T, V]$ and $[T, [T, V]]$ and assuming bounds on V, it is shown that such an estimate holds with $\|T\psi\|$ appearing in the constant of the $\mathcal{O}(\Delta t^2)$ estimate. The spatial regularity of ψ thus enters the error. If only an energy bound $\psi^* H\psi \leq B$ is available, then the order of convergence of the splitting scheme may decrease to one. Numerical experiments confirm this theoretically predicted order reduction.

5 Chebyshev Approximation

When a computationally efficient splitting is not available, or when there is little spatial regularity in the problem, an alternative is to compute the exponential of H as a whole, using polynomial approximations to the exponential. In the Chebyshev approach, this requires bounds for the extreme eigenvalues $E_{\min}$ and $E_{\max}$ of H (which is here assumed to be given as a spatial discretization). One then uses a truncated Chebyshev expansion of $\exp(-ix)$ on the interval $[\Delta t E_{\min}, \Delta t E_{\max}]$:

$$\exp(-ix) \approx \sum_{n=0}^{m} c_n \, P_n(x) \, , \quad \text{where} \quad P_n(x) = T_n \left(\frac{2x - \Delta t E_{\max} - \Delta t E_{\min}}{\Delta t E_{\max} - \Delta t E_{\min}} \right)$$

with the usual Chebyshev polynomials $T_n(\xi)$ for the interval $[-1, 1]$. Then, one uses the approximation

$$\exp(-i\Delta t \, H)\psi \approx \sum_{n=0}^{m} c_n \, P_n(\Delta t \, H)\psi$$

which is computed efficiently using Clenshaw's algorithm. This requires m multiplications $H\phi$ of the Hamiltonian H with a vector. Concerning the quality of the approximation, there is nearly no error reduction for $m \leq \frac{1}{2}\Delta t(E_{\max} - E_{\min})$, but very rapid, superlinear error decay for m growing beyond that bound[15]. The error is not influenced by regularity properties of ψ, as opposed to the situation of the Strang splitting. However, refining the space discretization increases $E_{\max}$ and thus requires a higher degree m or a reduction of the time step Δt.

6 Lanczos Approximation

A different, and according to our numerical experience often more efficient approach to polynomial approximation of the product of the matrix exponential times a vector, is by using the *Lanczos process*[10,13]. This approach to computing the exponential was proposed in the context of quantum dynamics[12], and its convergence properties have meanwhile been analyzed[4]. The symmetric Lanczos process generates recursively an orthonormal basis $V_m = [v_1 \cdots v_m]$ of the mth Krylov subspace $K_m(H, \psi) = \text{span}\,(\psi, H\psi, \ldots, H^{m-1}\psi)$ such that

$$HV_m = V_m L_m + [0 \cdots 0 \, \beta_m v_{m+1}].$$

This requires m multiplications of H with a vector, where m is chosen much smaller than the dimension of the problem. The symmetric tridiagonal $m \times m$ matrix $L_m = V_m^T H V_m$ is

the orthogonal projection of H onto $K_m(H,\psi)$. This is used in the approximation[2,4,12,14]

$$\exp(-i\Delta t\, H)\psi \approx V_m \exp(-i\Delta t\, L_m) V_m^T \psi$$

with $V_m^T\psi = [10\cdots 0]^T$. The matrix exponential $\exp(-i\Delta t\, L_m)$ is computed cheaply from the eigendecomposition $L_m = Q_m D_m Q_m^T$, with diagonal D_m, via

$$\exp(-i\Delta t\, L_m) = Q_m \exp(-i\Delta t\, D_m) Q_m^T\,.$$

The Lanczos process is stopped if

$$\beta_m \Big| \Big(\exp(-i\Delta t L_m) \Big)_{m,m} \Big| < \texttt{tol},$$

where $(\cdot)_{m,m}$ denotes the (m,m) entry of the matrix, and `tol` is a user-specified tolerance. This stopping criterion is motivated[6] by a generalization of a residual bound which is the most popular stopping criterion for solving linear systems. The convergence behaviour as a function of m is similar to Chebyshev approximation of the same degree in cases where the eigenvalues of H are densely distributed in the interval $[E_{\min}, E_{\max}]$, but convergence can be much more rapid when there are eigenvalue gaps within this interval[4]. Moreover, this approach takes advantage of preferred eigendirections in the vector ψ. It does not require a priori estimates of the extreme eigenvalues. On the other hand, the Lanczos process needs the computation of scalar products of vectors that are not required in the Chebyshev approach.

7 Magnus Integrators

In the Magnus approach[11], the solution of (1) is represented as

$$\psi(t_n + \Delta t) = \exp(\Omega_n)\psi(t_n), \tag{7}$$

where Ω_n is given as a series composed of integrals of commutators of $A(t) = -iH(t_n + t)$, the *Magnus series*

$$\begin{aligned}\Omega_n \;=\; & \int_0^{\Delta t} A(\tau)\, d\tau - \frac{1}{2}\int_0^{\Delta t}\Big[\int_0^{\tau} A(\sigma)\, d\sigma, A(\tau)\Big]\, d\tau \\ & + \frac{1}{4}\int_0^{\Delta t}\Big[\int_0^{\tau}\Big[\int_0^{\sigma} A(\mu)\, d\mu, A(\sigma)\Big]\, d\sigma, A(\tau)\Big]\, d\tau \\ & + \frac{1}{12}\int_0^{\Delta t}\Big[\int_0^{\tau} A(\sigma)\, d\sigma, \Big[\int_0^{\tau} A(\mu)\, d\mu, A(\tau)\Big]\Big]\, d\tau + \ldots\,.\end{aligned} \tag{8}$$

For smooth bounded matrices $A(t)$ the remainder in (8) is of size $\mathcal{O}(\Delta t^5)$, and hence the truncated series inserted into (7) gives a higher-order approximation to the solution value $\psi(t_n + \Delta t)$ for small Δt. A simpler expression that agrees with the truncated series up to terms of size $\mathcal{O}(\Delta t^5)$, is given in terms of the univariate integrals[1]

$$B_k = \frac{1}{\Delta t^{k+1}} \int_{-\Delta t/2}^{\Delta t/2} t^k A(\tfrac{1}{2}\Delta t + t)\, dt$$

as $\Omega_n = \Delta t\, B_0 - \Delta t^2[B_0, B_1] + \mathcal{O}(\Delta t^5)$. The integrals B_k can be replaced by suitable quadrature, e.g., by the fourth-order Gauss or Simpson rule. Using this approximation in (7) gives a time-reversible, unitary method of order 4:

$$\psi_{n+1} = \exp(\widehat{\Omega}_n)\psi_n \quad \text{with} \quad \widehat{\Omega}_n = \Delta t\, B_0 - \Delta t^2[B_0, B_1]. \tag{9}$$

In Blanes et al.[1], methods of order 6 requiring 4 commutators, and methods of order 8 requiring 10 commutators are also constructed. See Iserles et al.[7] for a detailed review of Magnus integrators.

As with the previously considered methods, the order statements must be taken with caution in the case of unbounded operators $H(t)$. It turns out[3] that the above method retains fourth order independently of the norm of $H(t)$ in the situation of $H(t) = T + V(t)$ with T a discretization of the negative Laplacian (with maximum eigenvalue $E_{\max} \sim \Delta x^{-2}$) and with a smooth potential $V(t)$, under a rather mild time step restriction

$$\Delta t \sqrt{E_{\max}} \leq Const.$$

We remark that this holds in spite of the fact that here the Magnus expansion is generally a divergent series. Convergence of the Magnus series would require a more stringent time step restriction $\Delta t\, E_{\max} \leq c\ (\approx 1)$.

8 Integrators for Almost-Adiabatic Quantum Dynamics

A different situation from that considered so far occurs in the treatment of problems of the type

$$i\varepsilon\dot{\psi} = H(t)\psi \qquad (0 < \varepsilon \ll 1) \tag{10}$$

with a small parameter ε (which, in self-consistent field approaches, would correspond to the square root of the mass ratio of light and heavy particles, such as electrons and ions). Here it is assumed that $H(t)$ varies slowly compared to the fast time scale ε. All of the previously considered integrators require time steps $\Delta t \ll \varepsilon$. For $\varepsilon \to 0$ and $\Delta t > \varepsilon$, they do not approximate the adiabatic limit as given by the quantum-adiabatic theorem.

Numerical integrators that give good approximations to (10) with relatively large time steps $\Delta t > \varepsilon$, have recently been derived[9]. These integrators are devised for situations where $H(t)$ is expensive to evaluate, but the substantially occupied eigenstates and eigenvalues of $H(t)$ can be obtained at comparatively small additional computational cost. This situation occurs in particular in reduced, relatively low-dimensional models, which are often appropriate for the description of near-adiabatic behaviour. Let $H(t)$ be diagonalized as

$$H(t) = Q(t)\Lambda(t)Q(t)^T, \qquad \Lambda(t) = \mathrm{diag}(\lambda_k(t))$$

with an orthogonal matrix $Q(t)$. (This can be extended to the situation where only a few of the lower eigenstates are computed[9].) The numerical integrator is not applied directly to (10), but to an equivalent equation for the variable $\eta(t)$ defined by

$$Q(t)^T\psi(t) = \exp\Big(-\frac{i}{\varepsilon}\Phi(t)\Big)\,\eta(t) \quad \text{with} \quad \Phi(t) = \int_0^t \Lambda(\tau)\,d\tau\ .$$

Up to a rapidly rotating phase, η is the coefficient vector with respect to the eigenbasis representation of ψ. Then η solves the differential equation

$$\dot{\eta}(t) = \exp\left(\frac{i}{\epsilon}\Phi(t)\right) W(t) \exp\left(-\frac{i}{\epsilon}\Phi(t)\right) \eta(t) \tag{11}$$

with the skew-symmetric matrix $W = \dot{Q}^T Q$. The right-hand side of (11) is bounded (though highly oscillatory), and hence η is smoother than ψ. As long as the eigenvalues of $H(t)$ remain well-separated, $\eta(t)$ stays $\mathcal{O}(\varepsilon)$ close to the initial value $\eta(0)$.

The simplest method is based on freezing the slow variables η, Λ and W over a time step and integrating analytically over the highly oscillatory exponentials. This gives the method[9]

$$\eta_{n+1} = \eta_{n-1} + 2h\big(S(t_n) \bullet E(\Phi_n) \bullet W_n\big)\eta_n, \tag{12}$$

where the bullets $\bullet$ denote the entrywise product of matrices, $S(t)$ is the matrix with entries $\sin x_{kl}/x_{kl}$ with $x_{kl} = \Delta t(\lambda_k(t) - \lambda_l(t))/\varepsilon$, and $E(\Phi)$ is the matrix with entries $\exp(\frac{i}{\varepsilon}(\phi_k - \phi_l))$. W_n is a finite difference approximation to $W(t_n)$: $W_n = (2\Delta t)^{-1}(Q(t_{n+1}) - Q(t_{n-1}))^T Q(t_n)$, and Φ_n is the Simpson rule approximation to the integral $\Phi(t_n)$.

This method forms the basis for more accurate schemes also derived in the article[9] . That paper also gives an extension to adaptive time steps to treat avoided crossings of eigenvalues, where non-adiabatic behaviour with sudden energy redistributions occurs.

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