Comparison of Methods for Numerical Time-Integration of the Schroedinger Equation

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In this report we compare the time integration algorithm introduced by Flocard, Koonin and Weiss [1] to a simple leapfrog algorithm. We first analyze both methods analytically and then confirm the results numerically.

1 The FKW Method

We begin with the analysis of a time-independent Hamiltonian H. Given a state $\psi(0)$ at time zero, the time evolution is expressed by the formula

$$\psi(t) = e^{-iHt}\psi \tag{1}$$

In the standard method of numerical integration, one expands the above exponential in a Taylor series. As will be seen and was found by FKW, the 4^{th} order expansion is especially useful. For an N-order expansion, denote this truncated operator by e_N,

$$e_N^x = \sum_n^N \frac{x^n}{n!}. \tag{2}$$

Acting with $e_N^{-iH\Delta t}$ on a state, one obtains that state evolved to $t + \Delta t$. That is, $\psi(t + \Delta t) = e_N^{-iHt}\psi(t)$.

Let us consider an eigenstate φ, where $H\varphi = \lambda\varphi$. The exact time evolution is then given by $\varphi(t) = e^{-i\lambda t}\varphi$. The wave function calculated by the algorithm is $\varphi(n\Delta t) = (e_N^{-i\lambda\Delta t})^n\varphi$, and it is clear that this method will diverge if $|e_N^{-i\lambda\Delta t}|^2 > 1$. The change of the norm with each time step is given by the following formula for $N = 4$:

$$|e_{(4)}^x|^2 = e_{(4)}^x e_{(4)}^{-x} = 1 + \frac{8x^6}{4!^2} + \frac{x^8}{4!^2}, \tag{3}$$

where $x \to -i\lambda\Delta t$. Making the substitution, we find

$$|e_{(4)}^{-i\lambda\Delta t}|^2 = 1 + \frac{((\lambda\Delta t)^2 - 8)}{4!^2}(\lambda\Delta t)^6. \tag{4}$$

Thus the norm decreases provided $|\lambda\Delta t| < \sqrt{8} \approx 2.83$. In practice, the states of interest have small values of λ, and the decrease of norm for states of large λ do not affect the observables.

It is interesting to compare this with the 2^{nd} and 6^{th} order expansions. These are

$$|e^{-i\lambda\Delta t}_{(2)}|^2 = 1 + \frac{(\lambda\Delta t)^4}{4}, \tag{5}$$

and,

$$|e^{-i\lambda\Delta t}_{(6)}|^2 = 1 + \frac{180 - 24(\lambda\Delta t)^2 + (\lambda\Delta t)^4}{720^2}(\lambda\Delta t)^8, \tag{6}$$

The 2^{nd} order modulus is always greater than 1, and the 6^{th} order modulus is only less than 1 for a small range of $\lambda\Delta t$ values. Thus, with these values of N, the norm would eventually diverge for any value of Δt.

2 The Symplectic Method

From Variational Calculus one can derive the following method,

$$\psi(t + \Delta t) = \psi(t - \Delta t) + 2iH\Delta t\psi(t). \tag{7}$$

We call this the leapfrog method; it probably goes by another name in the literature. An example of its use is ref. [3]. It conserves probability perfectly (to within machine precision) when a certain condition is met. Probability conservation ensures that the norm of the state will not blow up, so the method can safely be used for a large number of time steps.

We now derive the condition. Equation 7 is invariant under the transformation $t \to t \pm n\Delta t$, and so such translations form a group with the representation $e^{i\lambda_{sym}n\Delta t}$ for some value of λ_{sym} that depends on the eigenstate. Returning to the eigenstate φ and substituting in Eq. 7, we arrive at the following relation,

$$\varphi(t + \Delta t) = e^{i\lambda_{sym}\Delta t}\varphi(t) = \varphi(t)e^{-i\lambda_{sym}\Delta t} + 2i\lambda\Delta t\varphi(t). \tag{8}$$

Solving for λ in terms of λ_{sym} leads to,

$$\sin(\lambda_{sym}\Delta t) = \lambda\Delta t. \tag{9}$$

from which it is clear that if $|\lambda\Delta t| > 1$ then $\mathrm{Im}(\lambda_{sym})$ will be nonzero. Since $\varphi(n\Delta t) = (e^{i\lambda_{sym}\Delta t})^n\varphi(0)$, this imaginary component will cause the state to blow up eventually. Therefore the condition for the leapfrog method to be stable is $|\lambda\Delta t| < 1$.

This restriction imposes time steps in the symplectic method of approximately 1/3 the size of those in the FKW method. However the leapfrog method only requires only a single application of the Hamiltonian operator per step, whereas FKW requires four.

3 Numerical Comparison of FKW and leapfrog methods

We test and compare the two methods on a simple 3-state system designed to mimic the spectral distribution of the atomic cluster systems to which we

Table 1: Summary of validity of integration after a time of 30,000.

$\Delta t(\times 10^{-3})$	$\Delta t \lambda_{max}$	A	B
1.0	0.51	$\surd$	$\surd$
1.2	0.61	$\surd$	$\surd$
1.4	0.71	$\surd$	$\surd$
1.6	0.81	$\surd$	$\surd$
1.8	0.91	$\surd$	$\surd$
1.9	0.96	$\surd$	$\surd$
2.0	1.01	$\surd$	.
5.0	2.53	$\surd$	.
5.5	2.78	$\surd$	.
5.6	2.83	$\surd$	.
5.7	2.88	.	.
5.8	2.93	.	.

intend to apply these methods later. In such systems, the highest states are approximately 500 times greater than the lowest in energy. We therefore choose as our test-Hamiltonian the following,

$$\hat{\mathbf{H}} = \begin{bmatrix} 2 & 1 & 0 \\ 1 & 255 & 250 \\ 0 & 250 & 255 \end{bmatrix}. \tag{10}$$

The eigenvalues of this matrix are [1.84, 5.16, 505.0]. The initial state ψ is a mixture of the three eigenstates in the proportions $\psi = 1\varphi_0 + 1\varphi_1 + 1 \times 10^{-2}\varphi_2$. We integrate to a total time of 30,000 in the dimensionless units of this problem. Because this system is small and computationally simple, we are also able to calculate the *exact* time-evolution by applying the exact evolution operator $e^{i\lambda_n t}$ to each eigenstate component φ_n individually. The leapfrog method requires knowledge of the $\psi(-\Delta t)$ state, which we put in from the exact solution.

For each method we test a range of time step sizes to determine the maximum step size for which the integration remains valid after the specified time interval. Validity is taken to mean that the norm of the wave function does not blow up, and that both the modulus and the phase of each component (in the basis in which the Hamiltonian is expressed) differ from the exact calculation by no more that 1% at the end of the integration.

The result for various step sizes are summarized in table 1. In all cases where the integration failed, failure was due to the norm blowing up, rather than an inaccuracy in the phase or modulus. The correspondence between these results and the convergence conditions derived above is remarkable. Both methods A fail at almost exactly the points predicted.

It should be noted that the leapfrog method is sensitive to the initial information $\psi(-\Delta t)$. An error in this information causes an oscillation with period

$2\Delta t$ and amplitude approximately equal to the magnitude of the error. However, this oscillation does not grow over time.

Yabana and Bertsch [2] used the FKW method for real-time DFT calculations, where the Hamiltonian depends explicitly on the time-dependent charge density $\rho(t)$. FKW advocate using an approximation to $\rho(t+\Delta/2)$ in the operator H, which makes the method energy-conserving. However, finding $\rho(t+\Delta/2)$ requires an additional application of the evolution operator, doubling the number of Hamiltonian operations per time step.

We test the necessity of the energy-conserving evolution by examining a real-time DFT code which *does not* make use of this modification. Our test was conducted on an H2 system, with a 4^{th} order expansion of the evolution operator. We increased the time step size until either energy or probability failed to be conserved satisfactorily. Specifically, we used a spatial mesh size of 0.3 Å, which gives a maximum kinetic energy of 800 eV. We found that the maximum safe value of Δt to be slightly larger than 0.003 eV^{-1}, corresponding to $||\mathrm{H}||\Delta t \approx 2.5$. This is consistent with the the requirement for probability conservation derived above. Thus there is no regime in which probability is conserved but energy is not, and so the improved algorithm to ensure energy conservation is unnecessary, at least in the treatment of the optical response of molecules.

4 Conclusion

This analysis shows that the excellent energy conservation property of the leapfrog algorithm only gives a slight advantage over the fourth-order power series method, as the norm blows up before energy non-conservation becomes a significant problem. Nevertheless, the leapfrog method is faster than the standard 4^{th} order by a factor of $\frac{4}{\sqrt{8}} \approx 1.414$ combining the smaller time steps with the fewer operations of the Hamiltonian per step.

[1] H. Flocard, S. Koonin, and M. Weiss, Phys. Rev. **C17** (1978) 1682.

[2] K. Yabana and G.F. Bertsch, Phys. Rev. **B54** (1996) 4484.

[3] J.J. Rehr and R. Alben, Phys. Rev. B**16**, 2400 (1977).