A Chebychev propagator with iterative time ordering for explicitly time-dependent Hamiltonians

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A propagation method for time-dependent Schrödinger equations with an explicitly time-dependent Hamiltonian is developed where time ordering is achieved iteratively. The explicit time dependence of the time-dependent Schrödinger equation is rewritten as an inhomogeneous term. At each step of the iteration, the resulting inhomogeneous Schrödinger equation is solved with the Chebychev propagator.

The iteratively time-ordering Chebychev propagator is shown to be robust, efficient, and accurate and compares very favorably with all other available propagation schemes. © 2010 American Institute of Physics. [doi:10.1063/1.3312531]

I. INTRODUCTION

The dynamics of the interaction of matter with a strong radiation field is described by time-dependent Schrödinger equations (TDSEs) where the Hamiltonian is explicitly time dependent. This description is at the core of the theory of harmonic generation,1,2 pump-probe spectroscopy,3 and coherent control.4,5 Typically, an atom or molecule couples to a laser pulse via a dipole transition,

\[
\hat{H}(t) = \hat{H}_0 + E(t)\hat{\mu},
\]

with \(E(t)\) as the time-dependent electromagnetic field, causing the explicit time dependence of the Hamiltonian. Simulating these light-matter processes from first principles imposes a numerical challenge. Realistic simulations require efficient procedures with very high accuracy.

For example, in coherent control processes, interaction of quantum matter with laser light leads to constructive interference in some desired channel and destructive interference in all other channels. In time-domain coherent control such as pump-probe spectroscopy, wave packets created by radiation at an early time interfere with wave packets generated at a later time. This means that the relative phase between different partial wave packets has to be maintained for a long time with high accuracy. As a result, numerical methods designed to simulate such phenomena have to be highly accurate, minimizing the errors in both amplitude and phase.

The difficulty of simulating explicitly time-dependent Hamiltonians emerges from the fact that the commutator of the Hamiltonian with itself at different times does not vanish.6

\[
[H(t_1),H(t_2)] \neq 0.
\]

Formally, this effect is taken into account by time ordering such that the time evolution is given by

\[
\hat{U}(T,0) = T e^{-i\hat{H}_0 T} e^{i\hat{H}_d T}.
\]

The effect of time ordering is to incorporate higher order commutators into the propagator \(\hat{U}(T,0)\). For strong fields \(E(t)\) and fast time dependences the convergence with respect to ordering is slow. Methods to incorporate the second order Magnus term \(\hat{M}^2\) have been developed either in a low order polynomial expansion8,9 or as a split exponential.10

A quantum dynamical propagator that fully accounts for time ordering is given by the \((t,t')\) method.11 It is based on rewriting the Hamiltonian in an extended Hilbert space where an auxiliary coordinate \(t'\) is added and terms such as \(E(t')\hat{\mu}\) are treated as a potential in this degree of freedom. The Hamiltonian thus looses its explicit dependence on time \(t\), and can be propagated with one of the available highly accurate methods for solving the TDSE with time-independent Hamiltonian.12

Most of the vast literature on the interaction of matter with time-dependent fields in general3,13-15 and on coherent control in particular4,16-19 ignores the effect of time ordering. Popular approaches include Runge–Kutta schemes,7,20,21 the standard Chebychev propagator with very small time step,22 and the split propagator.13,19,23 Naively it is assumed that if the time step is small enough the calculation with an explicit time-dependent Hamiltonian can be made to converge. The difficulty is that this convergence is very slow—second order in the time step if the Hamiltonian is stationary in the time interval and third order if the second order Magnus approximation is used.8,24 Additionally in many cases the error accumulates in phase25 so that common indicators of error such as deviation from unitarity are misleading.