New strategy for solving the time-dependent Schrödinger equation

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1. INTRODUCTION

- **Motivation**
  - New XUV sources (HOHG, FEL) → nonlinear processes in XUV regime.
    - interaction of atoms/molecules with ultra-short pulses.
  - Time dependent approaches usually easier to handle than time independent perturbation theory.

- **Difficulties linked to the numerical solution of the TDSE**
  - The continuum components of the wave packet expand in a rapidly increasing volume of space → large grid/basis size >> artificial reflections from the numerical boundaries.
1. INTRODUCTION

- Increasingly large spatial phase gradients develop in the wave packet with time.

  - dense grids or large bases

  \[ \Psi(x, t) = \int_{-\infty}^{\infty} dx' G(x, x', t) \Psi(x', t_{\text{final}}) \]

  \[ G(x, x', t) = \sqrt{\frac{1}{2i\pi t}} e^{i(x-x')^2 / 2t} \]

- The solution of the TDSE on a spatial grid or using a basis of square integrable functions leads to stiff systems of coupled first order differential equations.

  - problems of stability and accuracy

- The extraction of the information on the multi-electron continua requires the knowledge of the asymptotic behaviour of the corresponding wave function.
2. STATE OF THE ART (for two-electron systems)

- In all time-dependent approaches, the wave packet is time propagated on an extended spatial region during the interaction with the pulse (grid or spectral method).
- All methods differ by the way the information is extracted from the wave packet.

**Standard methods of collision theory**

The wave packet is propagated freely after the pulse. Taking its Fourier transform allows to generate a scattering wave function which is then analyzed by means of time independent techniques.

- **Exterior complex scaling (ECS)**
  Which maps an outgoing wave into a vanishing wave outside a physically unaltered region [1].

- **HRM-SOW method**
  The scattering wave function is propagated with respect to the hyperradius semi-classically all the way to the asymptotic region where the various channels are disentangled [2].

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2. STATE OF THE ART (for two-electron systems)

Projection on stationary states

The wave packet is propagated freely after the pulse until it reaches a region where the ionisation channels are assumed to be decoupled. It is then projected on an uncorrelated product of Coulomb functions [3].

The wave packet is projected at the end of the pulse on a multichannel scattering wave function representing the single continuum in order to calculate the single ionization probability.

The double ionisation probability is obtained by subtracting the single ionisation probability from the total ionisation probability.

The multichannel scattering wave function is generated by means of the Jacobi matrix method [4].


3. TIME SCALED COORDINATE (TSC) METHOD

- **outline of the method**

  \[ r_1 = R(t)\xi_1 \]
  \[ r_2 = R(t)\xi_2 \]

- **time dependent scaling**
- **freezing of the wave packet expansion**
- **phase transformation of the wave packet**
- **remove fast oscillations**
3. TIME SCALED COORDINATE (TSC) METHOD

- The idea is not new …

- **Mathematical physics**
  
  Solution of the multidimensional quantum harmonic oscillator with time-dependent frequencies [5]

- **Plasma physics**
  
  Expansion into vacuum of a one-dimensional, collisionless, two-species classical plasma and a quantum electron gas in planar geometry [6,7].

- **Atomic and molecular physics**
  
  Proper adiabatic representation of the Coulomb three-body problem [8] and dynamics of ionisation in ion-atom and atom-atom collisions [9].

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3. TIME SCALED COORDINATE (TSC) METHOD

Interaction of a model atom/molecule with an electromagnetic pulse [10,11]
Electron impact single and double ionisation of helium [12,13]
Double photoionisation of helium [14]

● **Astrophysics**

Similarity properties and scaling laws of radiating fluids in laboratory astrophysics context [15].

● **Quantum Optics**

Free expansion of a Bose-Einstein condensate [16,17]

3. TIME SCALED COORDINATE (TSC) METHOD

Basic formulation (1-D model)

unscaled TDSE: \[ i \frac{\partial}{\partial t} \psi(x,t) = \left( -\frac{1}{2} \frac{\partial^2}{\partial x^2} + V_I(x,t) \right) \psi(x,t) \]

- \( x = R(t)\xi \)
- \( \varphi(\xi, t) = \sqrt{R(t)} e^{-\left(\frac{i}{2}\right) mR\ddot{\xi}^2} \psi(x,t) \)

scaled TDSE: \[ i \frac{\partial}{\partial t} \varphi(\xi,t) = \left( -\frac{1}{2mR^2} \frac{\partial^2}{\partial \xi^2} + V(R\xi) + \frac{1}{2} mR\ddot{\xi}^2 \right) \varphi(\xi,t) \]

narrowing of the atomic potential \( \rightarrow \) shrinking of the bound states
increase of effective mass \( \rightarrow \) confinement of the wave packet
presence of an harmonic potential
3. TIME SCALED COORDINATE (TSC) METHOD

\[
R(t) = \begin{cases} 
1, & t \leq t_{sc} \\
1 + \left[ R_{\infty} (t - t_{sc}) \right]^n, & t > t_{sc} 
\end{cases}
\]

- \( R_{\infty} \) = asymptotic velocity
- \( t_{sc} \) = starting time for scaling
- \( m_{\text{eff}} = \text{effective mass} = mR^2 \)

confinement of wave packet becomes stationary after sufficient time and after subtracting scaled bound states
3. TIME SCALED COORDINATE (TSC) METHOD

- Formulation of the 1-D model

\[ V_I(x, t) = -V_0 e^{-\beta x^2} - iA_0 f(t) \sin(\omega t + \varphi) \frac{\partial}{\partial x} \]

\[ \psi(x, t) = \sum_n a_n(t) \varphi_n(x) \]

two types of basis:

- \( \varphi_n(x) = \) Hermite-Sturmian functions
  \[ \varphi_n(x) = N_n^\alpha e^{-\frac{\alpha x^2}{2}} H_n(\sqrt{\alpha} x) \]

- \( \varphi_n(x) = \) B-splines \( B^k_n(x) \) with exponential sequence of breakpoints
3. TIME SCALED COORDINATE (TSC) METHOD

- **Time evolution of the scaled ground state**

\[ V_0 = \beta = 1 \]  
\[ R_\infty = 0.1 \text{ a.u.} \]  
\[ n=4 \]

At large times, the width of the scaled ground state is \( \propto \frac{1}{R_\infty t} \)
3. TIME SCALED COORDINATE (TSC) METHOD

- Time evolution of the unscaled wave packet

![Graph showing time evolution of a wave packet with labels for various parameters such as $t_{final}$, $t_{final} + 300$ a.u., $I_{peak} = 10^{13}$ Watt/cm$^2$, $\omega = 0.7$ a.u., pulse duration = 6 cycles, $V_0 = 1$ a.u., and $\beta = 1$ a.u.]
3. TIME SCALED COORDINATE (TSC) METHOD

- Time evolution of the scaled wave packet

![Graph showing time evolution of the scaled wave packet](image)

- Parameters:
  - $t = t_{\text{final}} + 250 \text{ a.u.}$
  - $V_0 = 1 \text{ a.u.}$
  - $\beta = 1 \text{ a.u.}$
  - $R_x = 0.01 \text{ a.u.}$
  - $n = 4$
  - $t_{\text{sc}} = t_{\text{final}}$
  - $I_{\text{peak}} = 10^{13} \text{ Watt/cm}^2$
  - $\omega = 0.7 \text{ a.u.}$
  - Pulse duration = 6 cycles
3. TIME SCALED COORDINATE (TSC) METHOD

- The quadratic increase of the phase at large distances is cancelled by the phase transformation of the wave packet, no fast oscillations.
- The use of one exponential sequence of breakpoints introduces high frequencies in the problem, increases the stiffness.
- Scaled bound states should eventually be subtracted from the wave packet. The subtraction must be performed after the pulse and when the harm. pot. = 0.

!! multi-resolution techniques under development !!
4. TIME PROPAGATION

The stiffness problem

- The magnitudes of the eigenvalues of the system of first order ODE vary greatly → stiff system of equations.

- The problem of the stiffness due to the occurrence of large eigenvalues Cannot be dissociated from the spatial representation of the wave packet

- The problem of stiff systems is twofold: stability and accuracy.

In an explicit scheme, large eigenvalues force the step length to be excessively small.

In an implicit scheme where the stability problem is avoided, the components corresponding to the largest eigenvalues are inaccurate.

- Explicit methods require matrix-vector products at each time step → parallel calculations.

- Implicit methods require to solve large systems of algebraic equations at each time step.
4. TIME PROPAGATION

Fatunla’s method

- Fatunla’s method = explicit scheme for the solution of stiff systems of equations [7]:

\[ y' = f(x, y), \quad y = (y_1, y_2, \ldots, y_m) \]

- The solution \( y(x) \) is approximated by the function \( F(x) \)

\[ F(x) = (I - e^{\Omega_1 x})a - (I - e^{-\Omega_2 x})b + c \]

\[ \Omega_i = \text{diag}(\omega_1^{(i)}, \ldots, \omega_m^{(i)}) \]

4. TIME PROPAGATION

- The stiffness parameters are the solution of a simple quadratic system of algebraic equations.

- The stiffness parameters are related to the eigenfrequencies of the atom + field system.

- \( y_{n+1} \equiv y(x_{n+1}) \) is obtained from \( y_n \) by a simple recurrence relation involving matrix-vector products only parallel calculations.

- The local truncation error can be easily calculated allowing adaptative step sizes.
First illustration

\[
\begin{align*}
\Delta &= \left| \tau_{\text{exact}} - \tau_{\text{num}} \right| \\
\tau(x) &= \left| y_1(x) \right| = \sqrt{1 + \frac{1}{4} \alpha^2 x^2}
\end{align*}
\]

Perturbed motion on a circular orbit in the complex plane of the point \( y(x) = y_1(x) \) which spirals slowly outward such that its distance from the origin \( \forall x \) is given by:

\[
\begin{cases}
    y_1' = y_2 \\
    y_1(0) = 1 \\
    y_2' = -y_1 + \alpha e^{ix} \\
    y_2(0) = \left( 1 - \frac{\alpha}{2} \right) i
\end{cases}
\]
4. TIME PROPAGATION

- Second illustration: ionisation of atomic hydrogen in its ground state by a strong low frequency pulsed field.

The time propagation is performed in a basis of Coulomb Sturmian functions.

- photon energy: 0.057 a.u.
- peak intensity: $6 \times 10^{14}$ Watt/cm$^2$
- pulse duration: 10 optical cycles $\approx 25$ fs
4. TIME PROPAGATION

- Third illustration: scaled and unscaled TDSE for 1-D model atom

**unscaled TDSE:**
- 2000 Hermite functions
- \( \alpha = 0.008 \)

**scaled TDSE**
- 400 Hermite functions
- \( \alpha = 0.05 \)
- \( R_\infty = 0.01 \) a.u.
- \( n=4 \)
- scaled bound states subtracted
- propagation over 4000 a.u.

Despite stiffness, the step size stays reasonable but relative error on norm conservation hardly lower than \( 10^{-5} \)
4. TIME PROPAGATION

- **Predictor-corrector scheme**

  **Predictor** = Fatunla’s method
  **Corrector** = fully implicit Runge Kutta method of order 7

  \[
  y_{n+1} = y(t_{n+1}) = y_n + h \sum_{i=1}^{4} b_i H(t_n + c_i h) Y_i
  \]

  \[ h = t_{n+1} - t_n \]

  \[ b_i, c_i = \text{Runge Kutta coefficients} \]

  \[ Y_i = \text{estimation of } y(t_i) \]

  TDSE: \[ i \frac{d}{dt} y(t) = H(t) y(t) \]  
  \[ H(t) = (N \times N) \text{ matrix} \]
4. TIME PROPAGATION

- The $Y_i$ are the solution of a system of dimension $4N$:

\[
\begin{pmatrix}
Y_1 \\
\vdots \\
Y_4
\end{pmatrix}
= 
\begin{pmatrix}
y_n \\
\vdots \\
y_n
\end{pmatrix}
+ h
\begin{pmatrix}
\begin{array}{cccc}
a_{11}H(t_n + c_1 h) & \cdots & a_{14}H(t_n + c_4 h) \\
\vdots & \ddots & \vdots \\
a_{41}H(t_n + c_1 h) & \cdots & a_{44}H(t_n + c_4 h)
\end{array}
\end{pmatrix}
\begin{pmatrix}
Y_1 \\
\vdots \\
Y_4
\end{pmatrix}
\]

- In a predictor-corrector scheme, the vector in the RHS is replaced by the result provided by the predictor → no system to solve → lack of accuracy.

- Trick: Introduce a diagonal matrix $D$ → iterative procedure requiring to solve 4 systems of dimension $N$ at each iterative step. The starting solution is provided by the predictor.
4. TIME PROPAGATION

\[
\begin{pmatrix}
Y_1^{(j)} \\
\vdots \\
Y_4^{(j)}
\end{pmatrix}
- \begin{bmatrix}
\begin{array}{cccc}
\begin{array}{c}
\vdots \\
0
\end{array} & \cdots & 0
\end{array}
\end{bmatrix}
\begin{bmatrix}
\begin{array}{c}
d_{11} H(t_n + c_1 h) \\
\vdots \\
d_{44} H(t_n + c_4 h)
\end{array}
\end{bmatrix}
\begin{pmatrix}
Y_1^{(j)} \\
\vdots \\
Y_4^{(j)}
\end{pmatrix}
= 
\begin{pmatrix}
y_1 \\
\vdots \\
y_4
\end{pmatrix}
\]

\[
+ h
\begin{bmatrix}
\begin{array}{cccc}
(a_{11} - d_{11}) H(t_n + c_1 h) & \cdots & a_{14} H(t_n + c_4 h) \\
\vdots & \ddots & \vdots \\
a_{41} H(t_n + c_1 h) & \cdots & (a_{44} - d_{44}) H(t_n + c_4 h)
\end{array}
\end{bmatrix}
\begin{pmatrix}
Y_1^{(j-1)} \\
\vdots \\
Y_4^{(j-1)}
\end{pmatrix}
\]

Each system is solved by means of biconjugate gradient algorithm which is an iterative procedure: maximum 2 iterations are needed!

The overall procedure only requires matrix-vector products allowing parallel calculations.
5. OBSERVABLES

**Electron energy spectrum**

\[ \psi(x,t) = \sum_n a_n(t)\phi_n(x)e^{-iE_nt} + \int_{-\infty}^{+\infty} c(k)\phi_k(x)e^{-i\frac{k^2}{2}t} \, dk \]

- \( \phi_k(x \to \infty) = \frac{1}{\sqrt{2\pi}} e^{ikx} \)
- **stationary phase theorem**

\[ \psi(x \to \infty, t \to \infty) = c\left(\frac{x}{t}\right) \frac{1}{\sqrt{it}} e^{\frac{i x^2}{2t}} \]

- \( \varphi(\xi, t) = \sqrt{R(t)}e^{-(i/2)mR\xi^2} \psi(x, t) \)
- \( k = \frac{x}{t} \)

\[ c(k) = \sqrt{\frac{i}{R_\infty}} \varphi\left(\frac{k}{R_\infty}, t \to \infty\right) \]

The spectrum is \( \propto \) to the mod square of the scaled wave packet.
5. OBSERVABLES

- Effect of an inaccurate description of a scaled bound state

![Graph showing energy distribution and parameters related to bound state calculations.](Image)
5. OBSERVABLES

- The density of energy states in the continuum plays a crucial role in the accuracy of the above-threshold ionisation (ATI) spectrum.

- The accuracy of the ATI spectrum is limited by the precision of the time propagation scheme.
5. OBSERVABLES

Atomic Hydrogen

\[ I_{\text{peak}} = 10^{14} \text{ Watt/cm}^2 \]
\[ \omega = 0.114 \text{ a.u.} \]
pulse duration = 20 cycles

- : scaled TDSE, 600 Sturmian functions,
  \[ \alpha = 0.4, \ t_{\text{end}} = 20000 \text{ a.u.} \]

- : unscaled TDSE, 800 Sturmian functions,
  \[ \alpha = 0.4 \]
6. CONCLUSIONS AND PERSPECTIVES
6. CONCLUSIONS AND PERSPECTIVES

- The time scaled coordinate approach is reflection free and allows to control the increasingly large phase gradients that develop during the time propagation.

- Provided that the scaled bound states are extracted after the end of the pulse and when the harmonic potential has disappeared, Fatunla’s method or the predictor-corrector scheme allows to propagate accurately the scaled wave packet to the genuine asymptotic region where the various channels are decoupled.

- The electron energy spectra may be expressed in terms of the modulus square of the scaled wave packet.

- Our investigations suggest that the optimal way to represent spatially the scaled wave packet is based on multi-resolution techniques.
6. CONCLUSIONS AND PERSPECTIVES

Solution of the TDSE associated to the interaction of helium with an external field:

- Hamiltonian in hyperspherical coordinates.
- Time dependent scaling of the hyperradius.
- Use of the semi-classical boundary conditions in the double ionisation channels.