Time-Dependent Method in Laser Material Interactions

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Outline:

- What we are working on
  - *Understand* the mechanism of laser-material interactions
  - *Control* material properties in an ultra-short time scale

- Computational Methods
  - Working Equations: Time-dependent Schrodinger equation (PDE)
  - Many-electron effect: Model potential, Density-functional theory

- Examples
  - Mechanism of atomic ionization in mid-infrared Laser field
  - Control transparency of a material in attosecond domain
  - Others

- Future Plan
  - Develop a numerical tool for a many-electron system
  - Investigate dynamics in a many-electron system
Our Research Goals:

Control

Insulator $\Rightarrow$ conductor (1 fs)


PRA 87 (2013) 063413.
Theoretical Method: Working Equations

TDSE in differential form

\[ i \frac{\partial}{\partial t} \Psi(t) = H(t) \Psi(t) \quad \text{with} \quad \Psi(t = -\infty) = \Phi_0 \]

TDSE in integral form

\[ 
\Psi(t) = -i \int_{-\infty}^{t} e^{-i \int_{\tau}^{t} H(t') dt'} V_{in}(\tau) e^{-i H_0 \tau} \Phi_0 d\tau + e^{-i H_0 t} \Phi_0 \\
= -i \int_{-\infty}^{t} U(t, \tau) V_{in}(\tau) U_0(\tau, -\infty) \Phi_0 d\tau + U_0(t, -\infty) \Phi_0 
\]
Theoretical Method: Boundary

Dynamics-related wavefunction

\[ \Psi(t) = -i \int_{-\infty}^{t} U(t, \tau)V_{in}(\tau)U_0(\tau, -\infty)\Phi_0 d\tau + U_0(t, -\infty)\Phi_0 \]

Repartition of the wavefunction

\[ \Psi(r, t_j) = \Psi(r, t_j)(1 - f(r)) + \Psi(r, t_j)f(r) \]

[Chelkowski & Bandrauk, IJQC 60 (1996) 1685.]
Theoretical Method: Time Propagator I

Second order split-operator-method in the energy representation

$$\Psi(t + \Delta t) = U(t + \Delta t, t)\Psi(t) = e^{-iH\Delta t}\Psi(t)$$

$$\approx e^{-iH_0\Delta t/2}e^{-iV(t)\Delta t}e^{-iH_0\Delta t/2}\Psi(t) + O(\Delta t^3)$$


Discretize space in pseudo-spectral grid:

$$H_0 = H_{r_i,r_j}^0(\ell), \quad \Psi(r_i, \theta) = \sum_\ell R_\ell(r_i)Y_{\ell,m}(\hat{r}), \quad \Psi(r_i, \ell) = R_\ell(r_i)$$

Time-propagation $\rightarrow$ vector, matrix operations $\rightarrow$ blas

Easy refactor to modern computers, GPU cublas, MIC ??
Theoretical Method: Time Propagator I

Time propagators:

\[ U(t + \Delta t, t) = e^{-iH_0 \Delta t/2} e^{-iV_{in}(t) \Delta t} e^{-iH_0 \Delta t/2} \]

\[ U_v(t, t_i) = e^{-i \int_{t_i}^{t} (\mathbf{p} - \mathbf{A}(t'))^2 / 2 dt'} \]

\[ U_0(t, t_i) = e^{-iE_0(t-t_i)} \]

\[ \Psi(t) = \Psi(\mathbf{r}, t) + \Psi_c(\mathbf{p}, t) \]
Example 1: Mechanism of ATI in mid-IR Field

ATI: above-threshold ionization

Physical Process: \[ A + n \ h\nu \rightarrow A^+ + e \]

- understand the structure:
  interference between the returning and rescattering electrons
- Information encoded in the structure:
  inner-work: how the electron interacts with the parent core
Example 1: Mechanism of ATI in mid-IR Field


HA-PACS: 5 nodes, 20 hrs
Example 1: Mechanism of ATI in mid-IR Field

Separate the tunnel ionization from propagation:

\[ \Psi_k = U(\infty, t_{k+1}) \int_{t_k}^{t_{k+1}} U(t_{k+1}, t)V_{\text{ext}}(t)e^{-iH_0t}\Psi_0 dt \]

Low Energy Structure comes from multiple re-scattering.
Multiple re-scattering also exists for other wavelengths. But no experimental report on LES in 800 nm or shorter. Why?

Two conditions to observe LES:

- multiple scattering (exist for all laser wavelengths)
- tunnel ionization (dominant only for longer laser wavelengths)

### TABLE I. The ratio of the tunnel ionization probability to the total ionization probability ionized in a half cycle. ($I_0 = 10^{14}$ W/cm$^2$).

<table>
<thead>
<tr>
<th></th>
<th>0.5 $I_0$</th>
<th>1.0 $I_0$</th>
<th>1.5 $I_0$</th>
<th>2.0 $I_0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>400 nm</td>
<td>0.055</td>
<td>0.391</td>
<td>0.614</td>
<td>0.727</td>
</tr>
<tr>
<td>800 nm</td>
<td>0.570</td>
<td>0.812</td>
<td>0.931</td>
<td>0.969</td>
</tr>
<tr>
<td>1600 nm</td>
<td>0.970</td>
<td>0.995</td>
<td>1.000</td>
<td>1.000</td>
</tr>
</tbody>
</table>

![Graph showing ATI yields vs. photoelectron energy]
Example 2: Experiment Observations
IR assisted photoionization by APT or SAP

Physical Processes

SAP: 0.3 fs

EUV: APT: 10 fs

IR field: 45 fs

Observation: Ionization yield depends on the time-delay for He, not for Ar.

Johnsson et al., PRL 99, 233001 (2007)
Example 2: Interference between Different APs

Predictions: total yields

<table>
<thead>
<tr>
<th></th>
<th>He</th>
<th>Ar</th>
</tr>
</thead>
<tbody>
<tr>
<td>APT</td>
<td>Yes</td>
<td>No</td>
</tr>
<tr>
<td>SAP</td>
<td>No</td>
<td>No</td>
</tr>
</tbody>
</table>

For a given ATI energy

<table>
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<td>?</td>
</tr>
<tr>
<td>SAP</td>
<td>?</td>
<td>?</td>
</tr>
</tbody>
</table>
Example 2: For a specified ATI peak, oscillation *always* exists.

Conclusion: **New mechanism is needed!**
Example 2: Mechanism

Our proposed mechanism: [PRA 81 (2010) 021404(R)]

- IR field dressed atomic states -- Floquet states
- XUV excites an atom to a Floquet state through different sidebands.

\[
\Psi_\alpha(\mathbf{r}, t) = e^{-i\epsilon_\alpha t} \sum_{n=-\infty}^{\infty} e^{-i n \omega t} \phi_{\alpha,n}(\mathbf{r})
\]

- The interference between the transitions to different side bands results the oscillation.

\[
P(t_d) \propto \sum_\alpha |M_1^\alpha F_1 + M_2^\alpha F_2 e^{-i2\omega t_d}|^2
\]
Example 2: IR Assisted Photoionization
Example 2: IR Assisted Photoionization

Generate a super-fast optical switch

ON

OFF
Other Works

- Visualization of multiple-scattering in ATI spectra

- Control Molecular Dynamics from fs to as,
  PNAS, 111 (2014) 912.

- Attosecond-Resolved Evolution of a Laser-Dressed Helium Atom

- Laser enabled Auger decay in rare-gas atoms,
  PRL 106 (2011) 053002.

- Attosecond Streaking in the Low-Energy Region as a Probe of Rescattering,
  PRL 107 (2011) 183001.

- Breit interaction on dielectronic recombination in HCI

- Anomalous bumpy structures in the capture cross-section of antiprotons by helium atoms,
Future Plan: Limitation of the Present Method

Spherical coordinate: waste too much grid point

- $z_{max} \propto E_0 \cdot \lambda^2$
- $U_p \propto E_0^2 \cdot \lambda^2$

E0 = 0.1: 800 nm -> 30. a.u. 10,000 nm -> 4,000 a.u.

cylindrical: too specified (symmetry)

Cartesian: works for atoms, molecules and clusters

- $N \propto \lambda^3$
- $N_x \cdot N_y \cdot N_z$

50x50x10,000 $\rightarrow$ GPU 5GB
Future Plan: Develop a New Method

- Develop a general method to solve TDSE using split-operator-method in Cartesian coordinate + FFT

\[ \Psi(t + \Delta t) = e^{-iH\Delta t} \Psi(t) \approx e^{-iT\Delta t/2} e^{-iV(t)\Delta t} e^{-iT\Delta t/2} \Psi(t) \]

one time-step propagation: 4x 3DFFT + 3x Multiplications \( N \ln N \)

- eigen-value problems: \( \Delta t \rightarrow -i\Delta t \)
  sub-space \( \rightarrow \) time-propagation \( \rightarrow \) eigenvalue \( \rightarrow \) time-propagation..

\[ \{ e^{-H\tau} \psi_i(0) \} \rightarrow \{ \psi_i(\tau) \} \rightarrow \{ \langle \psi_i | H | \psi_j \rangle \} \rightarrow \cdots \cdots \]

- real time-dependent problems.

\[ \{ \psi_i(t) \} \rightarrow \{ \psi_i(t + \Delta t) = e^{-iH(t)\Delta t} \psi_i(t) \} \rightarrow \{ \psi_i(t + \Delta t) \} \]
\[ \{ \psi_i(t + \Delta t) \} \rightarrow \{ \rho(t + \Delta t) \} \rightarrow \{ V(t + \Delta t) \} \]

- one time-step for 64x64x4096 \( \approx 0.6 \text{ sec} \) (8-core Sandy Bridge)
Future Plan: Status

✧ Eigen-value problems: tested examples:
  • harmonic potential: ✓
  • H atoms with atomic model-potential: ✓
  • H$_2^+$ molecular ions with atomic model-potential: ✓
  • H$_2$ molecules with atomic model-potential: ✓

✧ Extend the above method to a many-electron system using DFT
  • Poisson Equation: FFT-3D \( \rho(\mathbf{r}) \rightarrow V_c(\mathbf{r}) \) ✓
  • Exchange-correlation potential: \( \rho(\mathbf{r}) \rightarrow V_{ex}(\mathbf{r}) \) almost
  • Propagation in atomic pseudo-potential: not yet
  • Boundary condition and physical insights: not yet
Future Plan: Study Dynamic Process

- Study the dynamics processes of a material in an intense laser pulse (from 200 nm to 10,000 nm)
  - Extract information from measurements.
  - Use the information for molecular holography.
  - Generate Coherent X-rays in the water window (below carbon K edge at 4.37 nm)
  - Search a way to control a quantum process in ultra-short time scale (femto or atto-s)
  - And more ….
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