Using Time-Dependent Density Functional Theory to produce High Harmonic Generation

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Outline of talk

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  - Time-dependent DFT

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  - High Harmonic generation
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Aims/motivation

• The process of high-order harmonic generation provides a practical scheme for producing attosecond bursts in the extreme ultra-violet from intense infrared lasers

• The mechanism for such generation of harmonics is rapid ionization followed by recombination

• An intense laser is focused into a gas target and the high-order non-linearity of the response of the atoms provides a narrow beam of high-order harmonics

• Modelling laser-atom interactions requires highly accurate solutions of the time-dependent Schrodinger equation

• We have developed a time-dependent density-functional theory approach to study attosecond pulse generation in rare gas atoms
Wave function method

- Hartree-Fock theory states that the many-electron wave function can be written as a product of one-electron wave functions
  \[ \Psi(r_1, r_2, \ldots) = \psi_1(r_1)\psi_2(r_2)\ldots\psi_N(r_N) \]

- The ground-state wave function is approximated by a Slater determinant
  \[ \Psi = \begin{vmatrix} \psi_1(r_1) & \psi_1(r_2) & \cdots & \psi_1(r_N) \\ \psi_2(r_1) & \psi_2(r_2) & \cdots & \psi_2(r_N) \\ \vdots & \vdots & \ddots & \vdots \\ \psi_N(r_1) & \psi_N(r_2) & \cdots & \psi_N(r_N) \end{vmatrix} \]

- The orbitals \( \psi_i \) are chosen to minimise the energy. The variational principle is used
  \[ E_{HF} = \min\{E[\psi_{HF}]\} \]

Failures of the Hartree-Fock approach
- Lack of electron correlation
- Very complex function depending on 3N variables
- Inefficient - only one Slater determinant to expand the many-body wave function

How do we include correlation
- Configuration Interaction: the wave function is written as a sum of Slater determinants
- Use Density Functional Theory
Density Functional Theory

What is DFT?

- It is a many-body theory that connects ground state properties of a system to the electron density (in an exact relationship) rather than the many-body wave function.
- Although DFT is formally exact, the exact functional is unknown, although good approximations are known.

Where can DFT be applied?

- Presently most successful approach in computing the electronic structure of matter - ranges from atoms, molecules and solids to quantum and classical fluids.
- Chemistry - DFT predicts a variety of molecular properties, such as structures, vibrational energy, ionisation energies, reaction paths etc...

Why use DFT?

- Computational costs are much lower (compared to the wave function method).

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1 A functional is a map between a set of functions and a set of numbers, usually denoted by $F[f(x)] = y$. 
TDDFT describes a system of interacting particles beyond the ground-state structure.

Density of interacting particles obtained from the density of an auxiliary system of non-interacting particles moving in an effective local single particle potential.

Density calculated via solution of Kohn-Sham equations:

\[
n(r, t) = \sum_{\sigma=\uparrow \downarrow} \sum_{i=1}^{N_{\sigma}} |\psi_{i\sigma}(r, t)|^2
\]

\[
i \frac{\partial}{\partial t} \psi_{i\sigma}(r, t) = \left[ -\frac{1}{2} \nabla^2 + V_{\text{coul}}(r, t) + V_H(r, t) + V_{x\sigma}(r, t) + V_{\text{laserr}}(r, t) \right] \psi_{i\sigma}(r, t)
\]

Poisson equation:

\[
\nabla^2 V_H(r, t) = -4\pi n(r, t)
\]

Exchange-only adiabatic Local Density Approximation (xLDA):

\[
V_{x\sigma}(r, t) = -2 \left( \frac{3}{4\pi} \right)^{1/3} n_{\sigma}^{1/3}(r, t)
\]
Grid-based approach

- Solution of the time-dependent Kohn-Sham equations is carried out in real space [1]
- A cylindrical geometry is used for the Kohn-Sham orbitals
  \[ r = \rho \cos \varphi i + \rho \sin \varphi j + zk \]
- Linearly polarised laser pulse along the \( z \)-axis allows us to treat \( \varphi \) analytically
- Finite-difference treatment of the \( z \) coordinate
  - allows for effective code parallelisation
- Lagrange mesh treatment of the \( \rho \) coordinate
  - accurately and efficiently maps the \( \rho \) space
- 18th-order Arnoldi propagator for time propagation [2]

Computer systems

Cray XT4
HEXAGON

XCmaster
• All electron calculations carried out

• Real-space mesh extent is

\[-44.84 \leq z \leq 44.84 \text{ a.u.}\]

\[\rho \leq 44.86 \text{ a.u.}\]

• Finite difference grid spacing

\[\Delta z = 0.02 \text{ a.u.}\]

(4485 grid points)

• Number of Lagrange-Laguerre points

\[N_\rho = 60\]

• Lagrange mesh scaling

\[h_\rho = 0.2027685\]

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<th>Atom</th>
<th>Present</th>
<th>Grabo et al</th>
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Polarisabilities

- For weak fields, leading-order perturbation theory can be used to determine the electric susceptibilities.

- The quadratic Stark effect is used to estimate the static polarisability, where the energy shift of the atomic state is given by

\[ \Delta E = -\frac{\alpha}{2} F^2 \]

where \( F \) is the static field and \( \alpha \) the polarisability constant.

- \( \alpha \) is calculated using the perturbation method within the Kohn-Sham scheme.

<table>
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<tr>
<th>Species</th>
<th>Polarisability (a.u.)</th>
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<tr>
<td></td>
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<td>Ne</td>
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<td>Ar</td>
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High Harmonic Generation

What is HHG?
HHG occurs when the strong electric field of the laser pulse interacts with the time-dependent laser induced dipole moment of the atom, causing non-linear scattering of the radiation.

3-step model:
1. Electron release by tunnelling
2. Acceleration in the laser field
3. Recollision with the parent ion and radiation of excess energy

Spectrum
The spectral density is calculated from the Fourier transform of the dipole acceleration:

\[ S(\omega) = \left| \int dt \, e^{i\omega t} \int dr \, \rho(r, t) \, \ddot{d}(t) \right|^2 \]

where the dipole acceleration is obtained using Ehrenfest’s theorem as

\[ \ddot{d}(t) = -\nabla \left[ V_{\text{coul}}(r, t) + V_{\text{H}}(r, t) + V_{\text{xc}}(r, t) \right] \]
HHG - Helium

(a) $1 \times 10^{14}$ W/cm$^2$

(b) $1 \times 10^{15}$ W/cm$^2$

(a) 5 pulse cycles

(b) 20 pulse cycles
Time-frequency analysis

- Analysis of time-frequency behaviour enables us to characterize the temporal and spectral structure of harmonics in both plateau and cutoff regimes.
- Time frequency analysis is performed using the Short-Time Fourier-Transform (STFT).
- A Hanning window is used (window length = 1/10 of an optical cycle).
Conclusions

• We employ time-dependent density functional theory to stimulate the response of rare-gas atoms to intense, ultra-short laser pulses

• We have implemented this as an algorithm on a parallel computer within Fortran 95 using MPI

• Static polarisabilities of rare gas atoms are calculated and found to be in good agreement with previous data

• HHG in helium is studied. The time profile of the emission spectra reveals new information on the dynamics of the interaction processes
Thanks for listening....