High harmonic generation and multi-photon ionization of atomic hydrogen in intense laser fields: a computational perspective

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Outline

- Expt.: HHG with 2-color laser for He(1s²)
- TDSE 3d: pseudospectral method for H(1s)
- Expt.: Attoclock measures tunneling time?
- TDSE 3d: dynamics of ionized wavepacket formation
Three-step model of HHG
Ottawa Experiment (Corkum)

- HHG plateau cut off at max. return kinetic energy
- IR laser: odd-order harmonics can go up to ~400
- Lower for Ti:sapphire laser (ω=0.057 au, 800 nm)
- New idea: add a perpendicular, weaker laser at 2ω;
- How is recombination affected?
- Use thin gas jet target: no phase-matching calculation required, single-atom response only
Thick target: propagation effects

PRL80, 3236 (F. Krausz et al., 750 nm laser): theoretical simulation matched well by experiment

![Graph showing intensity after propagation and single atom response]
HHG radiation is polarized along returning electron’s \( \mathbf{v} \): Resolve its components, know \( \theta \) vs phase delay.

Harmonic \( E_x \)-components: odd orders (inversion symmetry)  
\( E_y \)-components: only even orders (2\( \omega \) driving field)
HHG as a function of delay $0 < \tau < T/4$

Higher harmonic order: odd orders only!
Intermediate orders: even and odd, even stronger than odd!
Recollision angles $\theta$: Left=expt, right = class. simulation

Good: odd-only orders regime has near-zero angles $\theta$ (blue region, 0.1-0.3 fs delays)

Bad: simulation never gets larger angles than 45 degrees. (doesn’t go into red!)
Sample trajectories:

Start particles at the outer tunneling point

Vary the phase delay (different colors)

Trajectories become meaningless near nucleus

Re-collision angles tend to be small!

Coulomb interaction (in plane) is included
Quantum calculation: PRA88, 063419
will it get larger recollision angles?
(i.e., strong even vs odd harmonics at low order)

- Do H(1s), not He (more stable numerically)
- Solve TDSE, calculate \( <a_x>(t), <a_y>(t) \), Fourier an.

\[
a_x = -[\mathcal{H},[\mathcal{H},x]] = \left[\mathcal{H}, \frac{\partial}{\partial r}\right] \sin \theta \cos \phi
\]

- Power spectra:
- Red = x (odd),
- Blue = y (even)

One delay shown
TDSE H(1s) results for recollision $\theta$

Expt is for $0 < \Delta\phi < \pi/2$ only

QM: do get $\theta$ up to 75 deg

Some of the pattern is OK

Absolute values of HHG orders are lower in H(1s)

Should push the QM calc. to He in SAE model.

Expt should explore longer delays.

QM does better, as the complete wavepacket is taken into account?
Rotating electric field vector provides a time reference on the sub-fsec scale. Ursula Keller et al, ETH Zürich, Nature Physics, 7, 371 (2011).

Tunnel ionization at the peak of the multi-cycle laser pulse; follow classical trajectories for ionized electrons; Detect electrons and parent ions in two dimensions; Reconstruct: when was the electron released. Time delay?

Rotating E field vector = Clock handle

Attosecond resolution
TDSE simulations

• Q1: is it really tunneling ionization?
• Photoelectron momentum distributions show multi-photon absorption (MPI) features!
• Q2: Which state does MPI start from? (1s or excited)
• Q3: How does the ionized wavepacket build up?
• ‘time delay’ = result of prolonged packet formation?

• Idea: monitor the time evolution of $m$-level population
• Classically: freed electron at some distance picks up angular momentum due to torque exerted by rotating electric field.
**TDSE method:** 3d extension of PRA74, 031405 (XM Tong et al)

\[
    i\frac{\partial}{\partial t} \Psi(\vec{r}, t) = \left[ -\frac{\nabla^2}{2} - \frac{1}{r} + f(t)\vec{E}(t) \cdot \vec{r} \right] \Psi(\vec{r}, t) \quad f(t) = \text{envelope fn}
\]

\[
    \Psi(\vec{r}, t) = \sum R_{lm}(r, t)Y_{lm}(\Omega) \quad l=0..128, \ m=-l..l
\]

Pseudo-spectral method:
clever matrix representation, uses \( \Psi \) at grid points
Grid in SPC (\( r, \theta, \varphi \)) acts as collocation mesh

Avoidance of reflection: radial space is split into (I) and (II).
Regions (I) and (II) overlap with coherent wavefunction transfer.
In (II) a Volkov propagator is used (momentum space, VG)

- **TDSE (I)**
- **(II) Volkov**
- **mask function (acts as absorber for TDSE)**
Ionization probability grows over time

inner space (TDSE)

$T = 110 \text{ au}$!
Field $E_x$ pushes $e^-$ out to the left (tunneling?)
Then the field $E_y$ imparts momentum on free $e^-$
Classical naïve result: $[-A_x(t), -A_y(t)]$ parametric plot:
$\varphi_p = 270^\circ$ and $\mathbf{p} = [-A_x(o), -A_y(o)]$
m-level population over time

Length gauge (calc.)

Transformed to velocity gauge

Evolution of \( \langle L_z \rangle \) is a gauge-dependent result
Figure 1. \( P(m) \) of the inner wavefunction in length gauge (left column) and in velocity gauge (right column) for the 5e13W/cm\(^2\) driving laser field.

CP laser exerts torque on free electron -> angular momentum \( L_z \)
QM: electron is launched into high-A region: ‘ang. mom. potential’
6-cycle pulse, Elliptic Polarization

Small ellipticity, $\omega=0.057$ au, (800 nm)
Innermost cycle matters!
Central + 2 side peaks in intensity ->
(caused by ellipticity)
Intensity range: $10^{13}$-$10^{14}$ Wcm$^{-2}$
$E_0 < 0.1$ au
6-cycle EP result $I_0 = 10^{14}$ Wcm$^{-2}$

Ring structure = multi-photon ionization (MPI) ?
Why two structures on opposite sides ?
Three times higher peak electron yields compared to 3cyc CP
Test MPI hypothesis

\[ p^2/2 \text{ [au]} \]

Low orders suppressed

Spaced by \( \hbar \omega \)

\( \varphi_p \approx 270 \) peaks shifted in energy vs the \( \varphi_p \approx 90 \)

Idea: \( \varphi_p \approx 270 \) comes from \( t=0 \);
\( \varphi_p \approx 90 \) from two neighbor intensity peaks
Fitting the peaks

Energy [au]  $I_p = 5 \times 10^{13} \text{ Wcm}^{-2}$

Slope = 0.055

$\omega = 0.057 \text{ au}$

Record energy for order zero (ambiguous) vs peak intensity $I_p$
Understand ponderomotive shifts
Which $n$-level did the MPI electron come from?
Ponderomotive shift is $\sim I_p$ or $E_p^2$

$H(n=2)$

$H(n=3)$

$I_p$ is the peak intensity for $t=0$, or side peaks (16 data; 8 $I_o$ values)
Data: intercepts of MPI peak fits, absolute photon nr. is unknown
Time delay = tunneling time ??

In LG $m>5$: definitely ionization (not temporary bound population)
In VG bound states acquire high $m$ values (oscillate with field)
This is NOT tunneling from the H(1s) state!
Conclusions

- TDSE simulations support the idea of MPI in few-cycle CP or EP laser pulses after pre-excitation
- The idea of pre-excitation was proposed on a classical level by P.Corkum (years ago)
- Ponderomotive shifts (slope in the intercept energy vs $I_p$ line fit) are in line with steady-state formula
- HHG spectra for H(1s) are not as strong as the experimental He(1s²) data, but on the right track
- Simulations coupled with analysis provide insights into electron dynamics
- MPI idea extends PRA89, 021402 (2014).
Wavepacket formation