Attosecond Pulse Production
From Excited Molecules


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Motivation

Mechanisms of attosecond pulse excitation have been understood:

Nonlinear response of atoms and molecules at ionization by superstrong femtosecond pulse

Pulse duration obtained:
- 170 as (R. Lopez-Martens et al., PRL, 2005) – pulse train

Next step: Search for ways to control efficiency and spectra in attosecond pulse production (especially single attosecond pulse)

One of the ways:
To use excited atoms and molecules for preparation and control (“Engineering”) of electronic wave packets providing the maximum efficiency and flexibility of bremsstrahlung at electron-ion recollision

On this road, more classical than quantum-mechanical intuition may sometimes be helpful
To maximize the amount of electrons contributing to bremsstrahlung we engage

- Fast single cycle ionization
- Electrons with large collision radii rather than Corkum’s electrons
- Initial electronic excitation in atoms
- Vibrational excitation in molecules
- Orientation of molecule against laser field
- Orientation of molecule against direction of pulse propagation
- All together

Fast single cycle ionization
How to generate single attosecond pulse?
Use ionization in linearly polarized field with rapidly increasing amplitude

- Almost full ionization within a single optical cycle produces a quasiclassical electron wave packet
- Recollision with a higher energy provides deeper penetration in the X-Ray range
- Recollision of the electron wave packet as a whole gives a single attosecond burst


Experiment: (Z.Chang, A.Rundquist, H.Wang et al, 1997)

$\lambda_0 \sim 800$ nm, $\tau_0 \sim 26$ fs $\rightarrow$ He $\rightarrow$ $\lambda \sim 2.7$ nm, $\tau \leq 3$ fs

$E(t) = E_0 f \exp(\beta \omega t) \sin(\omega t)$
Electrons with large collision radii

Full 3D modeling – theory and computer experiment

Schroedinger equation

\[ \frac{i}{\hbar} \frac{\partial \Psi}{\partial t} = H\Psi = \frac{1}{2} \left( \frac{\hat{p}}{c} + \frac{\hat{A}(t)}{c} \right)^2 \Psi + V(x, y, z)\Psi \]

Here

\[ \hat{A}(t) = c \int_{-\infty}^{t} \hat{E}(t')dt'; \quad \hat{E}(t) = \hat{E}_0 f(t) \sin(\omega_0 t) \]

\[ f(t) = \exp\left(2\omega_0 t / \pi\right) \]

\[ E_0 = 0.36; \quad \omega_0 = 0.114 \]

Electric field:

Potential:

\[ V(x, y, z) = -\left(x^2 + y^2 + z^2\right)^{1/2} \]
Ionization from 2S state

Electric field:

Temporal and spectral response of ionized atom

weak interaction of the main part of electrons having large collision radii

strong interaction of the small part of electrons in the vicinity of Coulomb singularity
Ionization from 2S state

Electric field:

Electrons with large collision radii versus Corkum’s electrons?
Wave packet \( \psi_\omega = (1 - N) \exp\left(-\left(\vec{r} - \vec{R}\right)^2/R^2 + i\vec{P}\cdot\vec{x}\right) \)

Radiation spectrum for \( |N\psi_{\omega} + (1 - N)\psi_e|^2 \)

\( R = 3 \)

\( R = 6 \)

Temporal and spectral response of ionized atom

\[
E(t) = E_0 \exp\left[ -5 \cdot \left( \frac{\omega_0 t}{2\pi} - 1 \right)^4 \right] \sin(\omega_0 t) \quad E_0 = 2
\]
Temporal and spectral response of ionized atom

\[ E(t) = E_0 \exp \left( -\frac{1}{2} \left( \frac{\omega_0 t}{2\pi} - 1 \right) \right) \sin(\omega_0 t) \]

For \( E_0 = 1 \)

For \( E_0 = 0.8 \)
$E(t) = E_0 \exp\left(-\frac{\omega t}{2\pi} - \frac{1}{2}\right) \sin(\omega t)$

$E(t) = E_0 \exp\left(-\frac{\omega t}{2\pi} - \frac{1}{2}\right) \sin(\omega t)$
Temporal and spectral response of ionized atom

\[ E(t) = E_0 \exp \left[ -5 \left( \frac{\omega t - 1}{2\omega t} \right)^2 \right] \sin(\omega t) \]

\[ E_0 = 0.2 \]

\[ E_0 = 0.4 \]

\[ E_0 = 2 \]
Electronic excitation in atoms

Ionization from initially excited s-states of H

\[ \psi_n(r, t=0) = \frac{1}{n^{3/2}} \exp(-r/n) F\left(1-n, 2, \frac{2}{n} r\right); \quad P = |\psi|^2 \]

Initial electron wave packet

\[ n = 1 \quad n = 2 \quad n = 3 \]
Three-step strong-field model for the whole electronic wave packet:

1) Emerges from the atom being almost undisturbed
2) Is accelerated by the strong optical field and spreads
3) Recollides with ion and emits attosecond burst

The profile of attosecond burst can be found analytically for electrons weakly interacting with Coulomb center

Schroedinger equation

\[\begin{align*}
\frac{i}{\hbar} \frac{\partial \psi}{\partial t} &= -\frac{1}{2} \frac{\partial^2 \psi}{\partial r^2} - \frac{1}{r} \frac{\partial \psi}{\partial r} \\
\psi &= \frac{e^0}{r} \\
\frac{\partial \rho}{\partial t} &= i \frac{\partial^2 \rho}{2 \partial r^2}
\end{align*}\]

Solution in integral form

\[\begin{align*}
\varphi(r,t) &= \frac{1}{\sqrt{2\pi i \hbar}} \int_0^\infty \varphi(x,0) \left[ \exp \left( \frac{i}{2\hbar} (x-r)^2 \right) - \exp \left( \frac{i}{2\hbar} (x+r)^2 \right) \right] dx \\
\Psi_n(r,t) &= \frac{1}{r \sqrt{2\pi i \hbar}} \int_0^\infty x \Psi_n(x,0) \left[ \exp \left( \frac{i}{2\hbar} (x-r)^2 \right) - \exp \left( \frac{i}{2\hbar} (x+r)^2 \right) \right] dx
\end{align*}\]

Electron probability density at the center of the packet

\[\begin{align*}
\psi_n(0,t) &= A \sum_{m=0}^{n-1} C_m \left[ B_1 F \left( -n, m - n + \frac{3}{2}, z \right) + B_2 (-z)^{n-m-\frac{1}{2}} F \left( -m - \frac{1}{2}, n - m + \frac{1}{2}, z \right) \right]
\end{align*}\]

\[\begin{align*}
A &= -\frac{\exp \left( -z \right)}{\sqrt{\pi^3 n^3}} \\
B_1 &= \frac{n!}{\Gamma \left( m + \frac{3}{2} \right) \Gamma \left( n - m - \frac{1}{2} \right)} \\
B_2 &= \frac{n!}{\Gamma \left( m - n + \frac{3}{2} \right) \Gamma \left( m - \frac{1}{2} \right)} \\
C_m &= (-1)^{n-m} C^{2m+1} \frac{2}{2n^2}
\end{align*}\]
Asymptotic shape of the packet at $t \to \infty$

$$|\psi_n(r, t \to \infty)|^2 = \frac{2n^3 t^5}{\pi^2 (t^2 + n^2 r^2)^{n+2}} \left[ \sum_{m=0}^{n-1} C_{2m+1}(-1)^m (2m+1)^{n-m-1}(n^2 r^2)^m \right]^2$$

![Graphs showing the asymptotic shape for different values of n.](image)

Analytical solution for attosecond atomic response

$$d_n^*(t) = E(t) - R(t), \quad R(t) = \iint |\psi(r, t)|^2 \frac{\partial V}{\partial z} \, dx \, dy \, dz$$

$$R_n(t) = -\frac{2}{\pi n^2} \left[ \arctan \left( \frac{\lambda K_n(\lambda)}{n \left( n^2 - 1 (1 + \lambda^2) \right)^{n+1}} \right) \right]$$

$$\lambda = \frac{nz(t)}{t}$$

$$K_1(\lambda) = 3\lambda^4 + 8\lambda^3 - 3$$

$$K_2(\lambda) = 30\lambda^8 + 140\lambda^6 - 128\lambda^4 + 500\lambda^2 - 30$$

$$K_3(\lambda) = 105\lambda^{12} + 700\lambda^{10} - 1043\lambda^8 + 13584\lambda^6 - 11053\lambda^4 + 4340\lambda^2 - 105$$

![Graphs showing the analytical solution for different values of n.](image)
Spectra of atomic nonlinear response

Analytical result

Numerical result (n=2)

Atomic nonlinear response

Spectra of atomic response
Atomic nonlinear response

\[ W = \int R(t)^2 dt \quad W_2/W_1 \approx 35 \quad I_2(100)/I_1(100) \approx 10^4 \]
How short an attosecond pulse can be?

Besides the increased efficiency, the attosecond pulse is shorter for excited states. What is the shortest pulse that can be produced by this mechanism?

Optimal excitation state $n \approx 4-5$
Maximum collision velocity $V \approx 25-30 \ (v/c \approx 1/5)$

$t_{\text{min}} \approx 10 \ \text{as}$
Time-frequency spectra of attosecond bursts for $n=1, 2, 3, 4$ electronic states

Limitation due to Lorentz force effect

3S state
Vibrational excitation in molecules

Enhancement of XUV yield due to slower spreading of electron wave packet

Small ion spacing

Large ion spacing

Transverse polarization of optical field

Electron density

Electron density
Attosecond burst production in high intensity regime ($H_2^+$, ionization from excited states)

Potential curves

Eigenfunctions

HHG in stretched molecules


Pump-probe scheme for HHG in molecules

Enhanced attosecond burst production in a stretched molecule

\[ R(t) = \iint |V(\vec{r}, t)|^2 \frac{\partial V}{\partial y} \, dx \, dy \]


Wave packets: initial (up) and before recollision with ion core (down)

Initial electron transverse momentum distribution
3. Orientation of molecule against laser field
Alignment dependence of HHG in molecules

Theory

Experiment
Enhancement of efficiency: \( I(N=250, \theta=90^\circ) / I(N=250, \theta=0^\circ) \sim 10^{10} \)
Single attosecond pulse production in a stretched molecule

Orientation dependence

$H_2^+, D=20$

\[ \theta = 90^\circ \quad \theta = 60^\circ \quad \theta = 0^\circ \]
Single attosecond pulse production in a stretched molecule

Bond-length dependence

$H_2^+$, $\theta=0^\circ$

$D=14$

$D=20$

$D=28$

By recording attosecond pulse spectra, one can image molecule structure!
Orientation dependence

Bond-length dependence

Angle-averaged spectrum

\[ \omega_{\text{max}} = \frac{2\pi V}{d} = \frac{2\pi v d}{2\pi} = \frac{V d}{t} \]

- \( d \) – initial spacing of nuclei
- \( V \) – velocity of recollision
- \( t \) – out-of-molecule travel time

Putting spectral maximum into “water window”
Orientation of molecule against direction of pulse propagation

Optimization of electronic, vibronic excitations and Lorentz force effect

$H^2+, D=20$
$2S$ states
CONCLUSION

The choice of an initial state of atoms and molecules can have a significant effect on attosecond pulse production.

Optimal preparation of an excited molecule medium can lead to highly enhanced IR-XUV frequency conversion and provide efficient control of the spectral and temporal distributions of attosecond radiation.

Strong bond-length and orientation dependence of emission spectra can be used for continuously tunable coherent X-ray production and for probing molecular vibration-rotational dynamics.

Compromising excitation type, molecule orientation and Lorentz force effect can result in production of single pulse with $\approx 10$ attosecond duration.