ABCs of high harmonics and attoseconds

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nonlinear frequency conversion: \( \omega_q = q \omega_{800}, q \gg 1 \)

- SF physics, the semi-classical view and sub-cycle response
- attosecond formation by Fourier synthesis
- measurement methods
- unique information from applications
first international attosecond school

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Majorana Center, Erice, Sicily: http://www.ccsem.infn.it
what is & why are attosecond interesting?

modified from LCLS/SLAC website
challenges of breaking the fs barrier

• sub-femtosecond ⇒ short wavelengths (XUV, x-rays)
  single-cycle 25 as pulse ⇒ 7.5 nm (∼165 eV photon energy)

• uncertainty principle: $\Delta \nu \Delta t \approx 1$ ⇒ need bandwidth!!
  single-cycle 25 as pulse ⇒ $20 \times 10^{15}$ Hz frequency spread

• control the phases of the field, i.e. mode-locking

• attosecond metrology
• sub-femtosecond $\Rightarrow$ short wavelengths (XUV, x-rays)
  single-cycle 25 as pulse $\Rightarrow$ 7.5 nm ($\sim$165 eV photon energy)

• uncertainty principle: $\Delta \nu \cdot \Delta t \cong 1$
  $\Rightarrow$ need bandwidth!!

• single-cycle 25 as pulse $\Rightarrow$ 20 $\times$ $10^{15}$ Hz frequency spread

• control the phases of the field, i.e. mode-locking

• attosecond metrology

Q0: can I produce an attosecond EM pulse in the optical regime?
basic measurements and observables

- non-linear
- non-resonant
- non-perturbative
what is a strong-field?: the atomic gauge

Coulomb law:
\[ E = \frac{q}{r^2} \sim 5 \times 10^9 \text{ V/cm} \equiv 1 \text{ au} \]

what laser intensity gives an equivalent field strength?

\[ I_p = \frac{\varepsilon_0 c E_p^2}{2} \equiv 3 \times 10^{16} \text{ W/cm}^2 \]
motion of the free electron

classical equations:
\[ F = m \ddot{x} = -eE \sin(\omega t + \phi_0) \]
\[ m \dot{x} = (eE/\omega) \cos(\omega t + \phi_0) + mv_0 \]

cycle-averaged energy
\[ U_p \equiv \langle E_k \rangle = 1/T \int \frac{1}{2}mx^2 dt = (eE)^2/4m\omega^2 \]

ponderomotive or quiver energy: \[ U_p \propto \lambda^2 I/4 \]
displacement: \[ \alpha \propto \lambda^2 E \]

for 0.8 \( \mu \)m (red) laser at \( 10^{15} \) W/cm\(^2\) (0.18 au field)
\[ U_p = 60 \text{ eV} \]
\[ \alpha \sim 50 \text{ au (25 A)} \]

think in ponderomotive units !!!
the simpleman’s picture of ionization

- Gallagher, PRL 61, 2304 (1988)
- Van Linden van den Heuvell & Muller, in Multiphoton Processes (1988)

**electric field**

\[ E = E_0 \sin \omega t \]

**velocity**

\[ v(t) = \frac{E_0}{\omega} [\cos \omega t - \cos \phi_0] + v_0 \]

*quiver drift*

For tunneling, \( v_0 = 0 \)
simpleman prediction: electron distribution

\[ v(t) = \frac{E_o}{\omega} [\cos \omega t - \cos \phi_o] \]

\( \text{quiver} \) drift

* in the experiment, we detect the drift energy not quiver !!

\[ E(t) = \frac{1}{2}v^2 = 2U_p \cos^2 \phi_o \]

\( \text{maximum drift energy} = 2U_p. \)

ac-tunneling: Ammosov, Delone & Krainov, Sov. Phys. JETP 64, 1191 (1986)
simpleman prediction: electron distribution

quantum analogy: strong-field approximation or KFR theory:
Keldysh V S, Sov. Phys. JETP 20,1307(1965)
Faisal F H J, Phys. B 6,L89 (1973)

\[ \uparrow V \xrightarrow{x \to} \]

\[ E/U_p \]

\[ \text{electron counts} \]

\[ \text{maximum drift energy} = 2U_p. \]

\[ \text{tunneling } e^E \]

ac-tunneling: Ammosov, Delone & Krainov, Sov. Phys. JETP 64, 1191 (1986)
simpleman’s comparison to experiments

xenon 30 TW/cm²
$U_p = 3$ eV
bad news?
$\gamma = 1.5$ (multiphoton)

helium 1 PW/cm²
$U_p = 50$ eV
good news!
$\gamma = 0.5$ (tunneling)
position versus initial phase: $x(t) = E_o/\omega^2 \left( \sin \omega t - \sin \phi_o + (\phi_o - \omega t) \cos \phi_o \right)$
2nd approximation: semi-classical rescattering

Schafer, Yang, DiMauro & Kulander, *PRL*, 70, 1599 (1993)

- electron-core interaction $\sim \frac{1}{2}$ cycle
- electron gains field energy, max $3.17U_p$
- physics is inherently sub-cycle (attosecond!)
semi-classical description of harmonic emission

I. The electron tunnels through the distorted Coulomb barrier

II. The free electron is accelerated by the field, and may return to the atomic core

III. The electron recombines with the atom, emitting its energy as a photon
• classical trajectories have different release time (phase), propagation time and return times & energy.
• control of wave packet dynamics

Fourier synthesis of harmonic comb

FFT

odd harmonics separated by $2\omega$

log(photons)

wavelength (nm)

photons
Fourier synthesis of harmonic comb

Q1: What does the harmonic spectrum look like?
Fourier synthesis of electron comb

 FFT

above-threshold ionization (ATI) peaks separated by $\omega$
Fourier synthesis of electron comb

above-threshold ionization (ATI) peaks separated by $\omega$

Q2: What does the ATI spectrum look like?
origin of single-atom dispersion: attochirp

Electron position

$x(t_i) = 0$
$v(t_i) = 0$

Return energy

Short traj. chirp > 0
Long traj. chirp < 0

*modified from talk by B. Mahieu (CEA)
return energy: $E(t = t_r) = 2U_p (\cos^2 \phi_r + \cos^2 \phi_o - 2\cos \phi_o \cos \phi_r)$

\[
\text{cutoff energy } \sim 3.17U_p + I_p \propto 3U_p
\]
high energy electrons & photons

- elastic scattering yields energetic \((10U_p)\) electrons
- recombination (1-photon matrix element) with initial state produces high harmonics
- inelastic \((e, 2e)\) scattering \(\Rightarrow\) multiple electron ejection

Q3: What type of collision produces 10$U_p$?
(a) forward scattering
(b) back scattering

- the short-range physics is important
- wave packet spread reduces the effective rescattering
- the recollision occurs in less than an optical cycle

elastic rescattering: intensity scaling

ponderomotive potential: \( U_p \propto \lambda^2 I \)

helium, 0.8 \( \mu \)m

Q4: Why is it that as intensity increases, rescattering is less efficient?
elastic rescattering: intensity scaling

Rutherford (Coulomb) scattering

$$\frac{d\sigma}{d\Omega} = \frac{1}{16\varepsilon_0^2} \frac{1}{\sin^4(\phi/2)}$$

higher return energies produces less rescattered electrons since

$$\varepsilon_o \to 3U_p \propto I$$
HHG frequency comb for different $\lambda$, constant $I$

argon HHG, neon PES detector

$0.8 \, \mu m$

$1.3 \, \mu m$

$2 \, \mu m$

$110 \, \text{TW/cm}^2$

$\lambda^2$-scaling produces sufficient number of photons $\sim 200 \, \text{eV}$
monochromatic model for harmonics

\[ I(t) = \left| \sum_{q_{\text{odd}}} A_q e^{-i(q\omega_0 t + \varphi_q)} \right|^2 \]

\[ \Delta \varphi = \varphi_{q+2} - \varphi_q \]

Δφ = 0

train at T/2 and pulses are sub-cycle (attosecond)
monochromatic model for harmonics

\[ I(t) = \sum_{q_{\text{odd}}} A_q e^{-i(\omega_0 t + q \varphi)} \]

\[ \Delta \varphi = \varphi_{q+2} - \varphi_q \]

Q5: What happens in the time domain for this phase?

constant phase difference results in transform-limited pulses
attosecond Fourier synthesis

monochromatic model for harmonics

\[ I(t) = \left| \sum_{q_{\text{odd}}} A_q e^{-i(q\alpha_0 t + q \phi)} \right|^2 \]

\[ \Delta \phi = \phi_{q+2} - \phi_q \]

need a control and method to measure this!
attosecond engineering step 1: spectral filtering

- intrinsic time-structure is dominated by the beating between the strong low-order harmonics
- select the plateau region by spectral filtering
attosecond engineering step 2: select trajectory

**harmonic spectrum**

![Harmonic Spectrum Graph]

**temporal profile**

![Temporal Profile Graph]

**Contributions from 2 Dominant Trajectories**

- Short trajectory
- Long trajectory

Select the short trajectory by spatial filtering.

Bellini et al. PRL (1998)
attosecond pulse train generation

intense ultra-fast laser
0.8-2 μm

argon jet

Al filter

aperture

to experiment

selection in argon:
harmonic order 13 to 31
isolated attosecond pulse generation

confine emission to a half-cycle using CEP few-cycle pulses
• polarization gating
• amplitude gating
• bichromatic fields

see review paper by Popmintchev et al. Nature Photonics 2010
Reconstruction of Attosecond Beating By Interference of Two-photon Transitions

sideband signal:

\[ I_{q+1}^{SB} \propto \cos \left( 2\omega_{IR} \tau - \Phi_{q+1}^{SB} \right) \]

sideband phase:

\[ \Phi_{q+1}^{SB} = \varphi_{q+2} - \varphi_{q} - \Delta \varphi_{q+1}^{atomic} \]

\[ \Delta \varphi_{q+1} \]

intense optical field
absorption & emission

one-photon absorption

photoelectron spectrum

I_p
metrology: RABBITT measurement

- in combination with the harmonic amplitudes, the average attosecond pulse can be reconstructed

P. Johnsson et al., PRL 95, 013001 (2005)
OSU attosecond beamline/end-station

2π magnetic bottle e-spectrometer

toroidal mirror

HHG arm

probe arm

BS

1-2 µm

Ar HHG, Ne PES detector

1.3 µm

2 µm RABBITT spectrogram
RABBITT: $\lambda$-dependence of the attochirp

- the attochirp decreases with $\lambda$ at constant intensity
- the spectral phase is more complex than classical predictions
- can produce 100 as pulses from 30-150 eV
streaking metrology: isolated attosecond pulses

- attosecond pulse duration less than optical period
- attosecond pulse releases a photoelectron with some initial energy
- the photoelectron’s energy is shifted and broadened by the optical field
MPQ streaking for isolated pulse metrology

\[ p_f = p_i + A(\phi) \]

from Drescher et al., Nature 419, 803 (2002)
attosecond spectroscopy: scattering phase

a slow free particle
(k = 0.5 a.u.)

Q6: Does the particle’s momentum change?
atomic delay: Wigner delay

- Wigner, Eisenbud, Smith (1950)
- the phase shift cannot be measured
- the relative delay can be measured by comparison, e.g. valence & core
- compared the s and p photoionization for Ne (MPQ) and Ar (Lund) and measured 20 and 100 as, respectively.

RABBITT measures a group delay

\[ \tau_{\text{RABBITT}} = \tau_{\text{chirp}} + \tau_{\text{atomic}} \]

\[ \tau_{\text{atomic}} = \tau_{\text{Wigner}} + \tau_{\text{cc}} \] (source term)
attosecond spectroscopy: Wigner delay

attoseconds pulses can measure detailed electronic information

- source terms are removed by pairwise comparison
- the atomic delay of Ne, Ar and Kr are referenced to accurate He calculation
- measure the energy dependence of the Wigner delay
- delay decreases with increasing energy
- delay is sensitive to many-body effects, e.g. resonances

collaborators: M. Dahlström (Stockholm) A. Kheifets (Canberra)
• in 1962, Cooper predicted a minimum in the photoabsorption caused by a sign change (π-phase jump) in the dipole matrix element
• the Cooper minimum has been extensively studied in the frequency domain, particularly in argon
• the phase jump is not measured but inferred from other observables
HHG spectroscopy: Cooper minimum in argon

- In HHG, the field-driven electron wave packet probes the Cooper structure via the complex recombination dipole.
- HHG gives you access to the phase.

Collaborators: M. Gaarde (LSU)  
K. Schafer (LSU)