Attosecond Science: Challenges and Prospects

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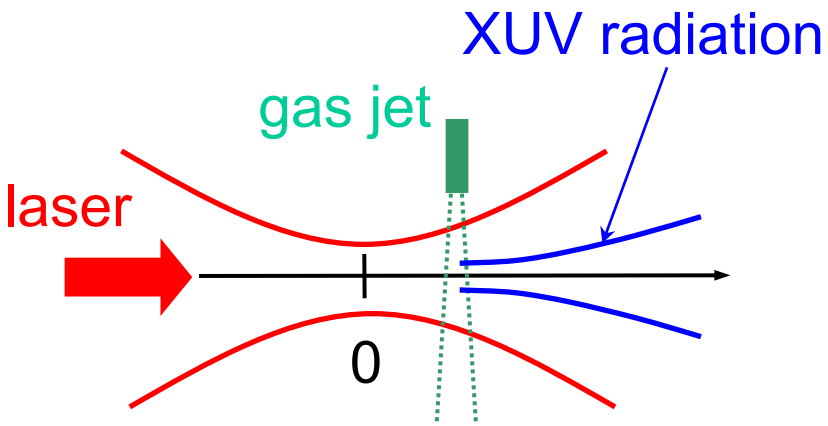
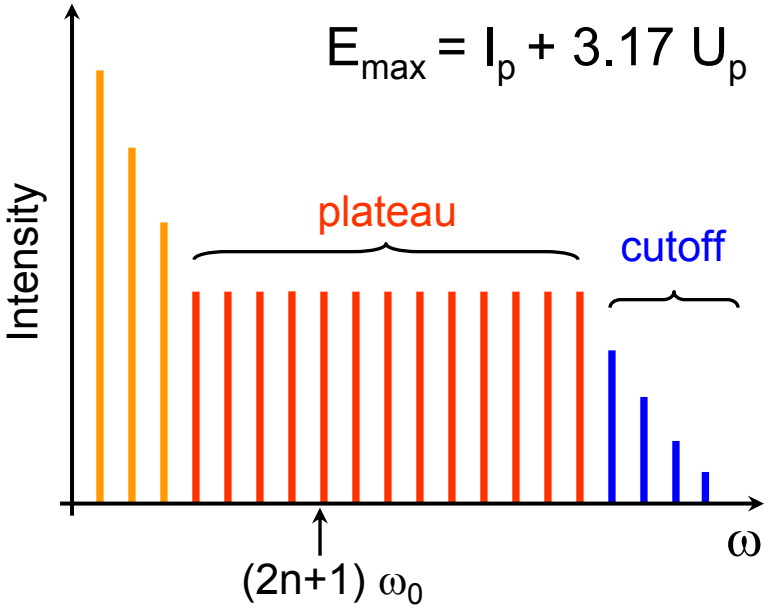
Outline

- Temporal gating schemes for the generation of isolated attosecond pulses
- Status and prospects of attosecond spectroscopy and control

High-order Harmonic Generation

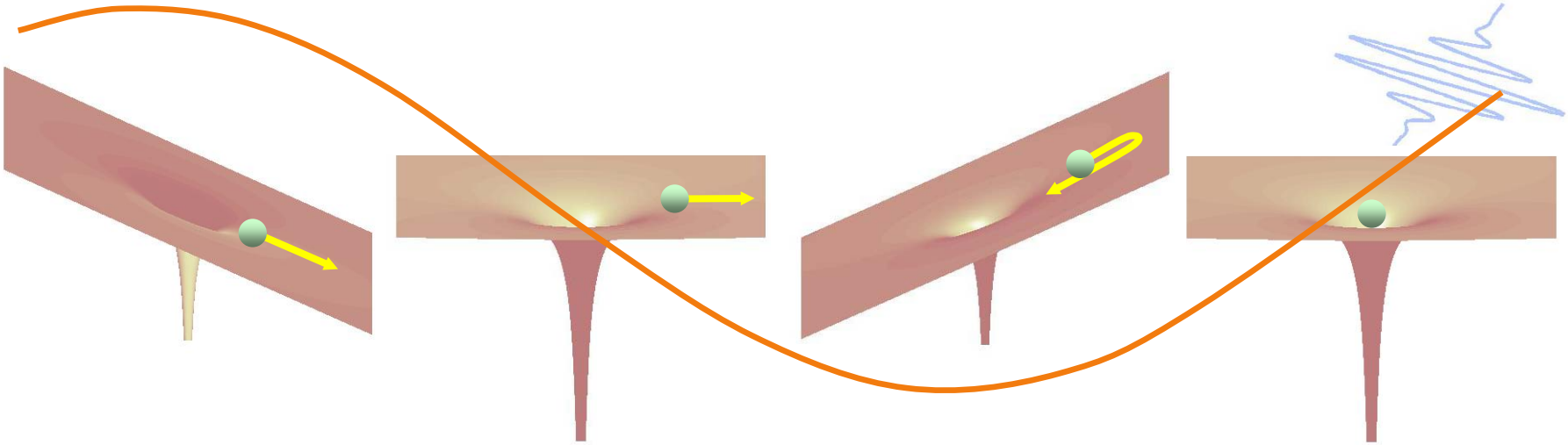
- An intense ultrashort light pulse is focused on a gas jet





➔ Odd harmonics of the visible light are generated up to the soft-X-ray region

Attosecond Source: High-order Harmonic Generation



Step 1
Ionization

Step 2
Motion after ionization

Step 3
Recollision

➔ maximum photon energy: $h\nu_{max} \propto I \lambda_L^2$

“Intrinsic” tools in Attosecond Technology

- Attosecond optical pulses always associated to attosecond electron pulses

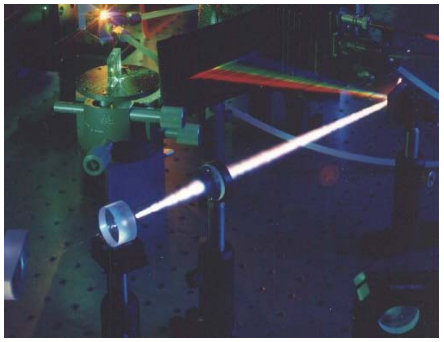
  electrons give access to spatial resolution:
  electron wavelength (~1Å)

  optics gives electron collision physics a systematic method for measuring dynamics

- Attosecond photon or electron pulses always synchronized to a visible pulse with controlled waveform

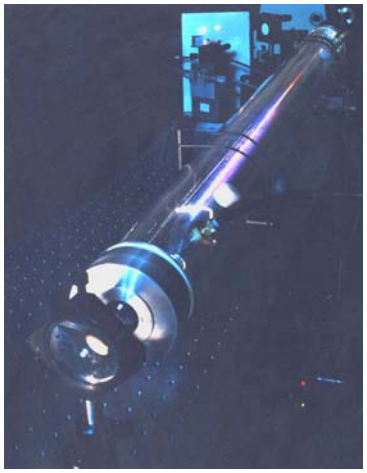
  extension of conventional ultrafast spectroscopy and strong field coherent control from the cycle-averaged into the sub-cycle domain of visible light

Basic “laser” tools for attosecond technology



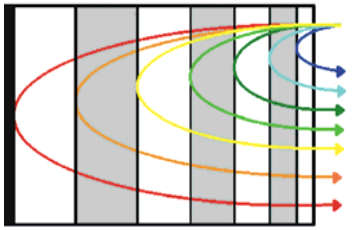
Chirped-Pulse Amplification

Strickland, G. Mourou, Opt. Commun. **56**, 219 (1985)



Hollow-fiber compression
Sub-6-fs high-peak power light pulses

M. Nisoli, S. De Silvestri, O. Svelto, Appl. Phys. Lett. **68**, 2793 (1996)

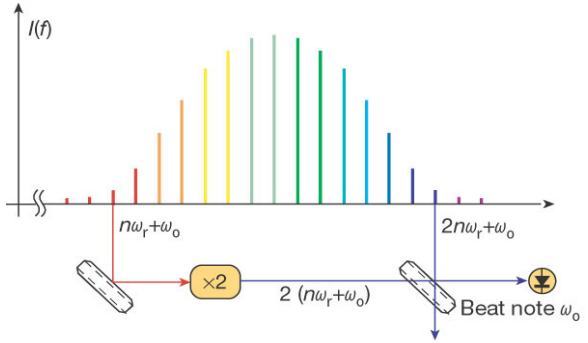


Ultrabroadband dispersion control with chirped mirrors

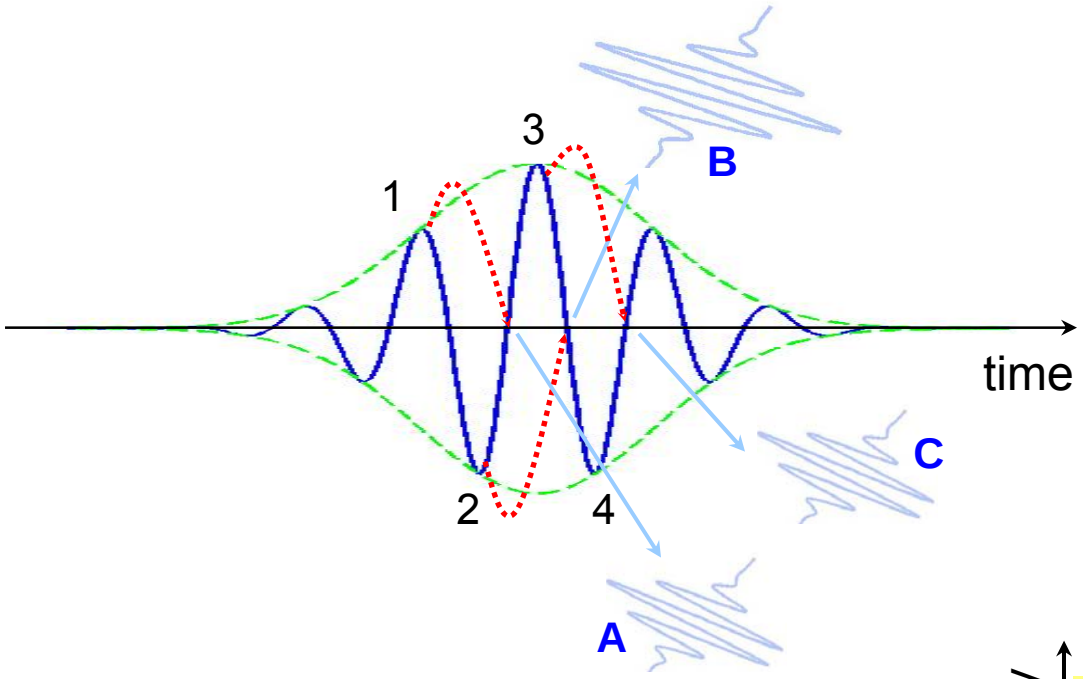
R. Szipocs, K. Ferencz, Ch. Spielmann, F. Krausz, Opt. Lett. **19**, 201 (1994)

Carrier-envelope phase stabilization

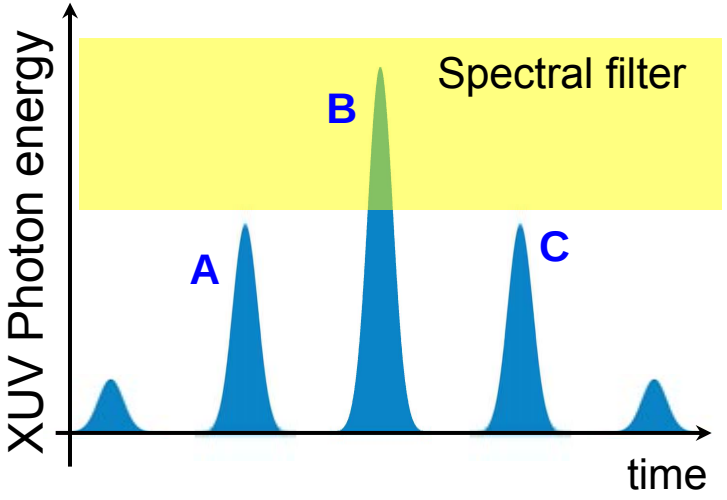
T.W. Hansh et al., 1997,1999
H. Telle et al., Appl. Phys. B **69**, 327 (1999)
D. Jones et al., Science **288**, 635 (2000)
A. Baltuska et al., Nature **421**, 611 (2003)



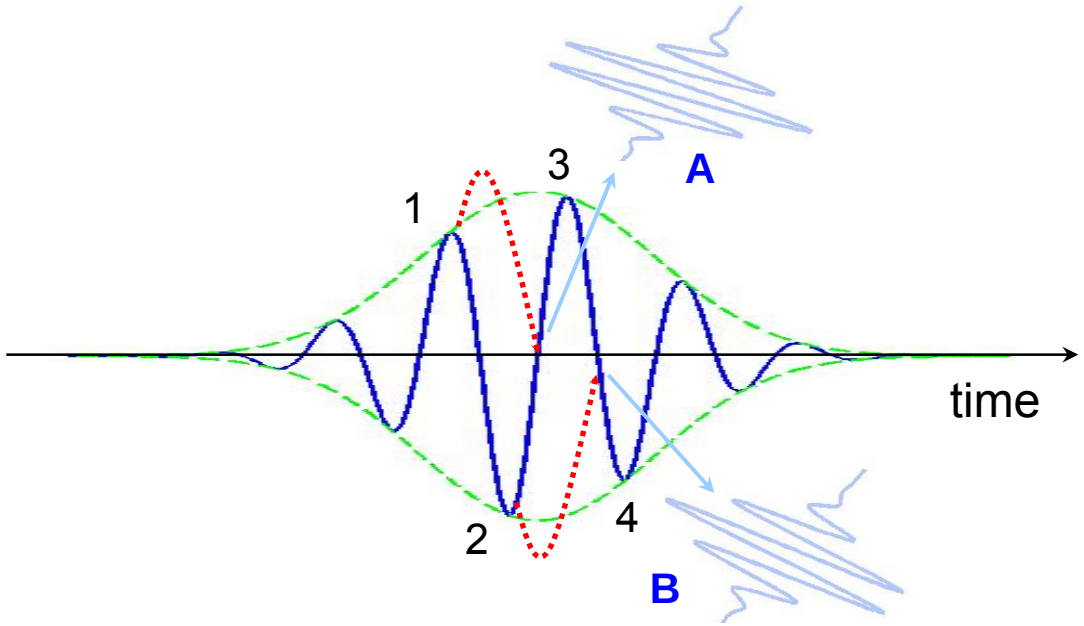
Spectral selection of cutoff harmonics



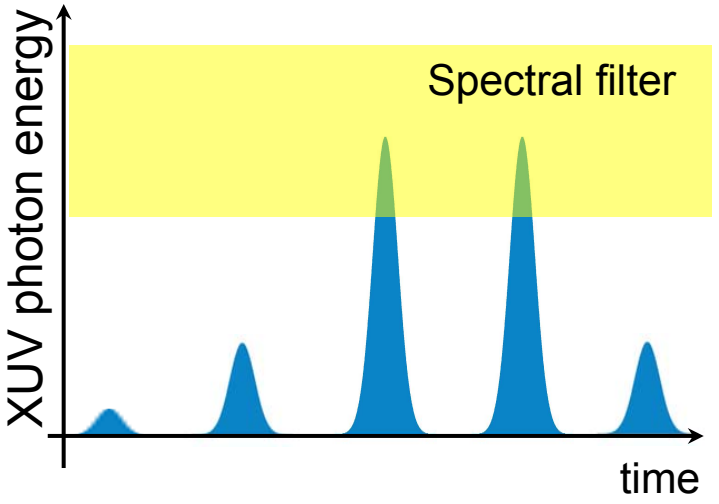
- Spectral selection of cutoff photons leads to generation of isolated attosecond pulses
- Requirements: sub-5-fs driving pulses (linear polarization)



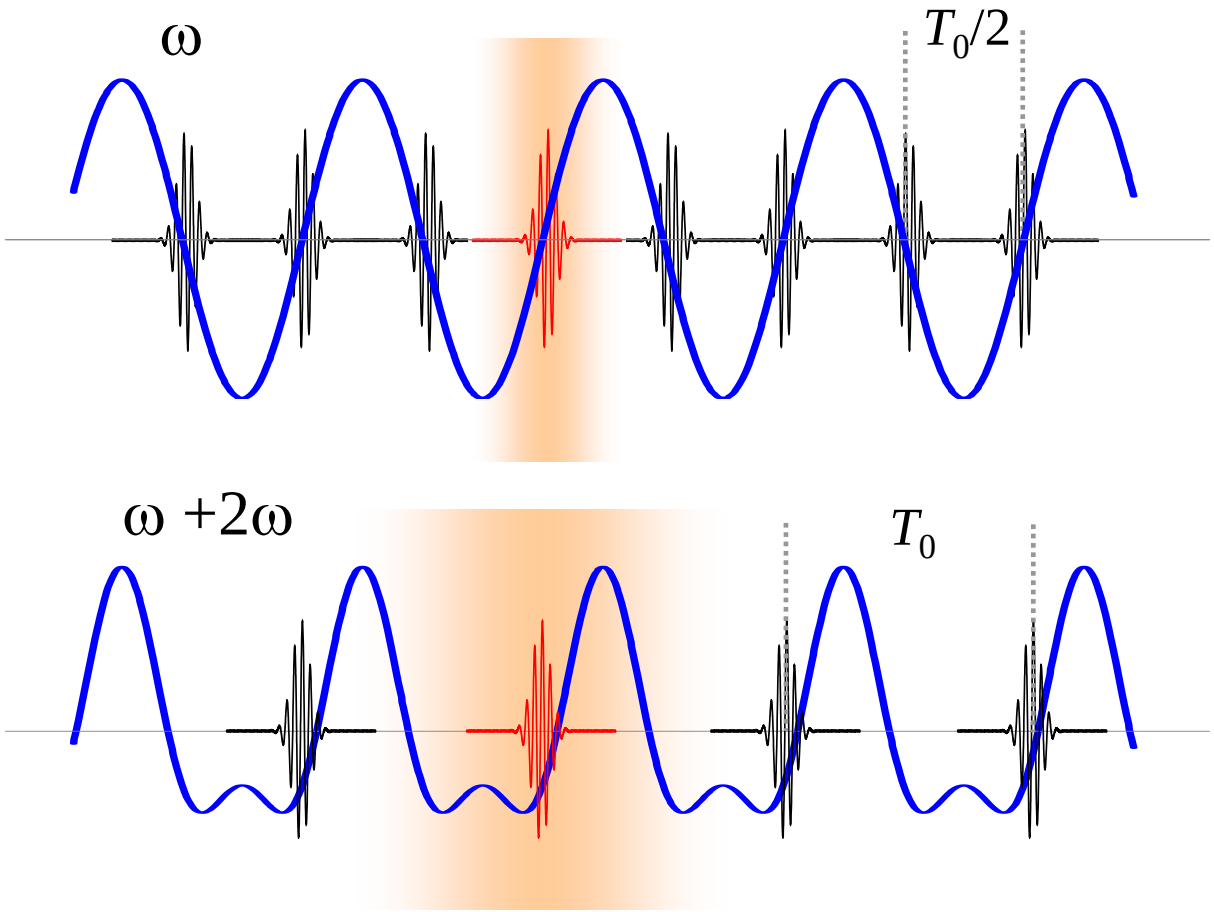
Spectral selection of cutoff harmonics



- Spectral selection of cutoff photons leads to generation of one or two attosecond pulses
- Requirements: sub-5-fs *phase-stabilized* driving pulses (linear polarization)



Temporal gating

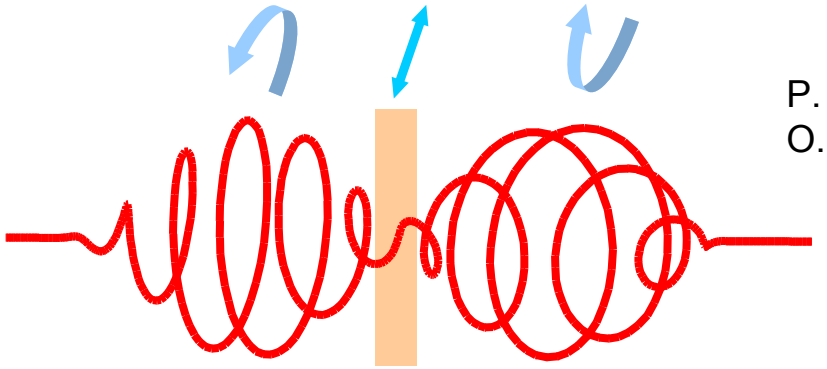


Temporal gating schemes

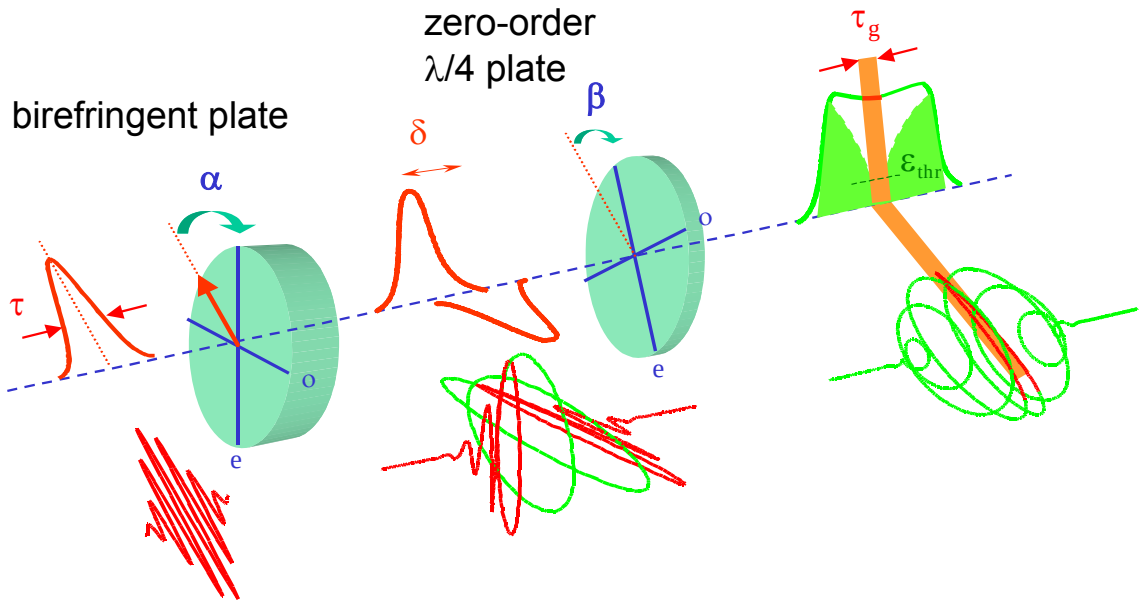
- Polarization gating:
  - ➔ One-color scheme
  - ➔ (Generalized)-Double-Optical gating (DOG and GDOG)
- Two-color gating:
  - ➔ intense IR pulses + intense visible (VIS) few-cycle pulses
- Ionization gating:
  - ➔ Few-cycle pulses with above saturation intensity and controlled electric field

Polarization gating

- Time-dependent polarization

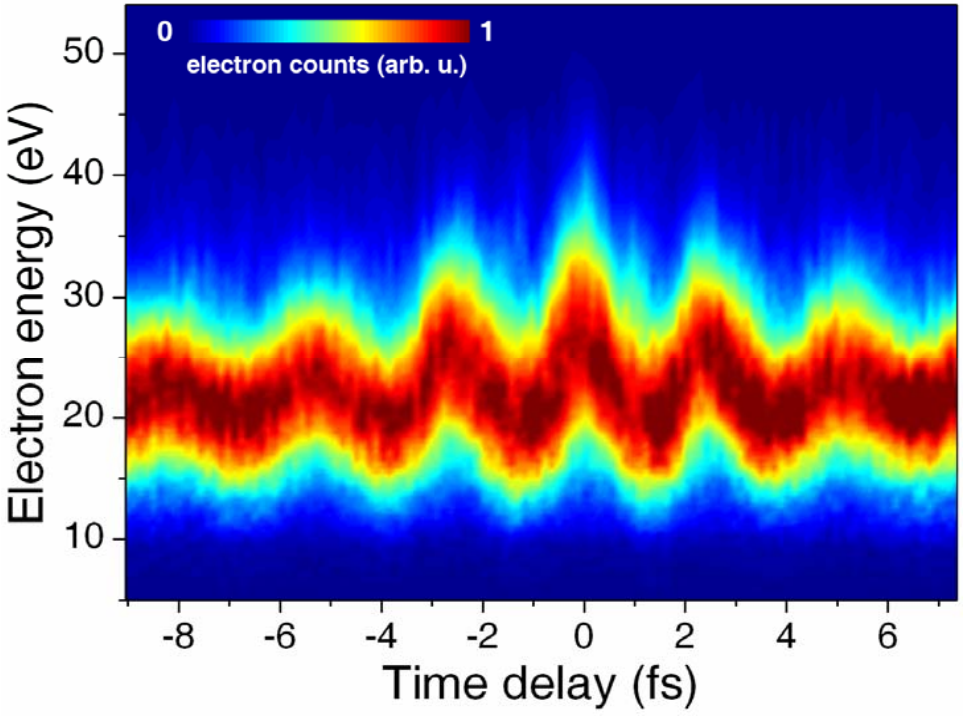


P. Corkum *et al.*, Opt. Lett. **19,** 1870 (1994)
O. Tcherbakoff *et al.*, Phys. Rev. A **68,** 043804 (2003)

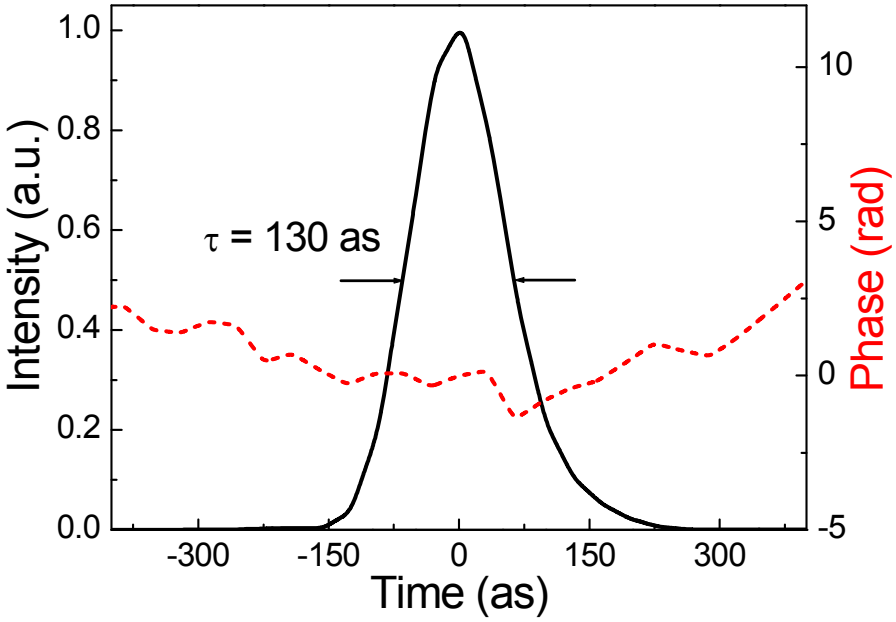


Temporal characterization

- Dispersion compensation by Aluminum foils



Retrieved Intensity profile and phase



➔ Good dispersion compensation

➔ Near-single cycle pulse

G. Sansone *et al.*, Science **314**, 443 (2006)

Temporal gating schemes

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- Two-color gating:
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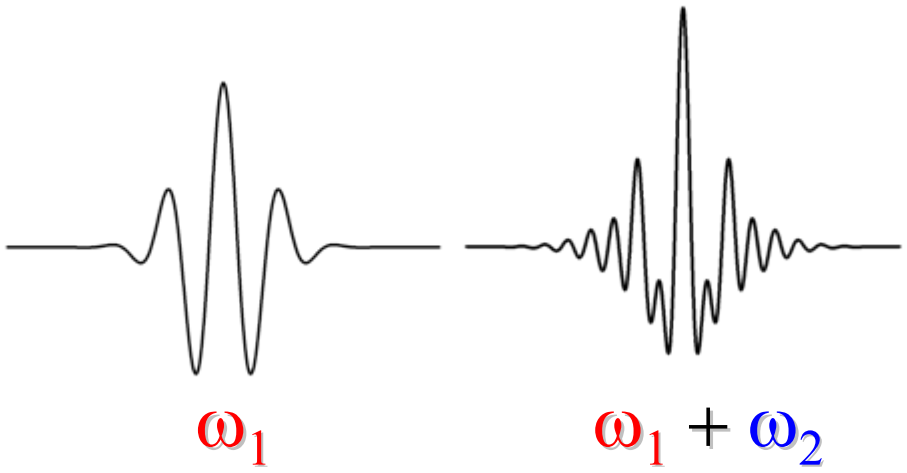
Two-color gating



Driving field: $\omega_1 + \omega_2$

$\omega_2 = 2\,\omega_1 + \delta\omega$: spectrally detuned second harmonic

New periodicity of the electric field can lead to isolation of single attosecond pulses

H. Merdji *et al*., Opt. Lett. **32**, 3134 (2007)

Key parameters:

central wavelength of the two components

intensity of the pulses

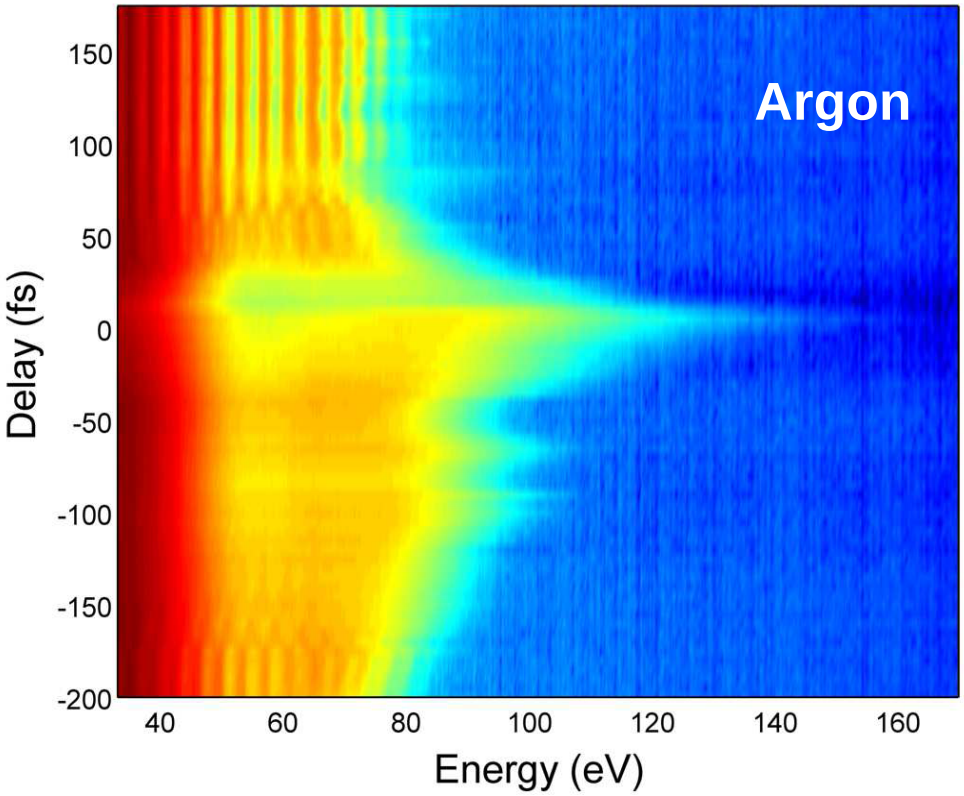
temporal overlap

gas target position

Intense ultrashort two-color driver

Intense IR pulses: 1.45 μm, 20 fs, $I_{IR} = 2 \times 10^{14}$ W/cm^2

Intense VIS pulses: 0.8 μm, 13 fs, $I_{VIS} = 8.5 \times 10^{14}$ W/cm^2

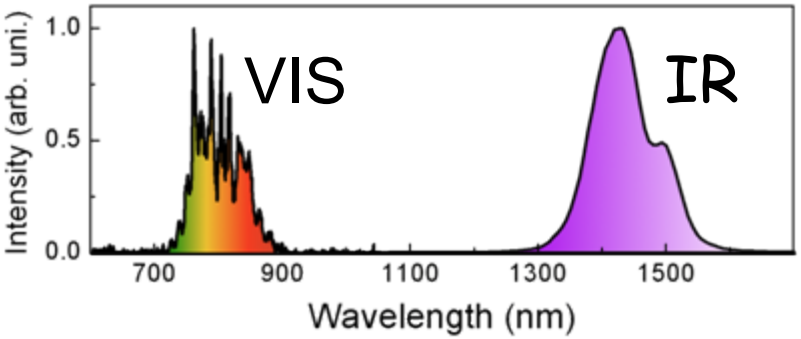


- ➔ τ = 0: dramatic cutoff extension and continuum generation
- ➔ outside overlapping region harmonic spectrum is dominated by VIS pulse
- ➔ IR component: responsible for cutoff extension
- ➔ VIS component: increase of conversion efficiency

F. Calegari *et al*., Opt. Lett. **34**, 3125 (2009)

Two-color vs one-color

F. Calegari et al., Opt. Lett. **34**, 3125 (2009)

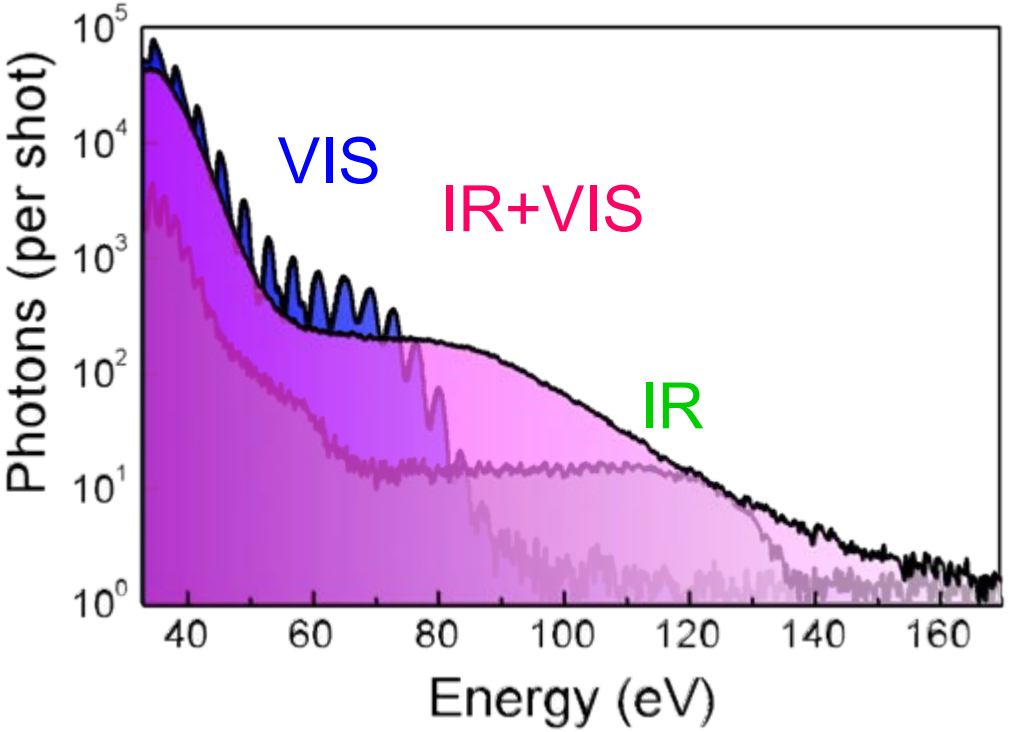


Argon

Maximum photon energy with
VIS: 100 eV
(high-conversion efficiency)

Maximum photon energy with
IR: 140 eV
(low-conversion efficiency)

Maximum photon energy with
VIS+IR: 160 eV



Temporal gating schemes

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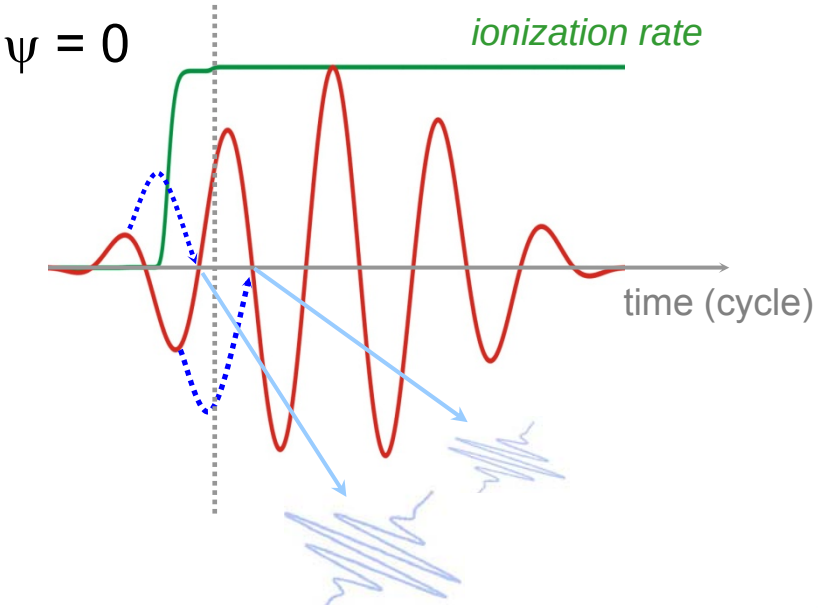
- Ionization gating:
  - ➔ Few-cycle pulses with above saturation intensity and controlled electric field

Sub-cycle ionization dynamics



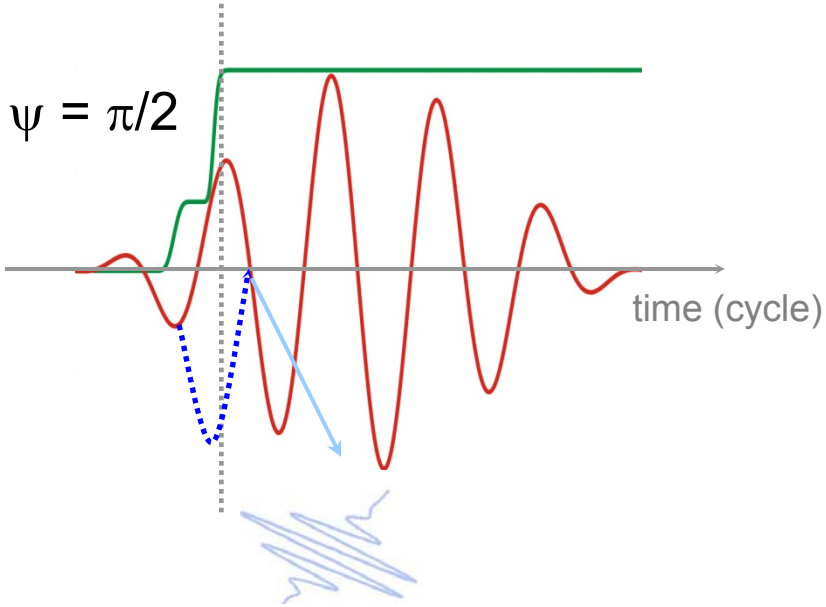
High-energy few-cycle pulses:

complete depletion of neutral atom population on the pulse leading edge

for some CEP values confinement of the XUV emission within a single event

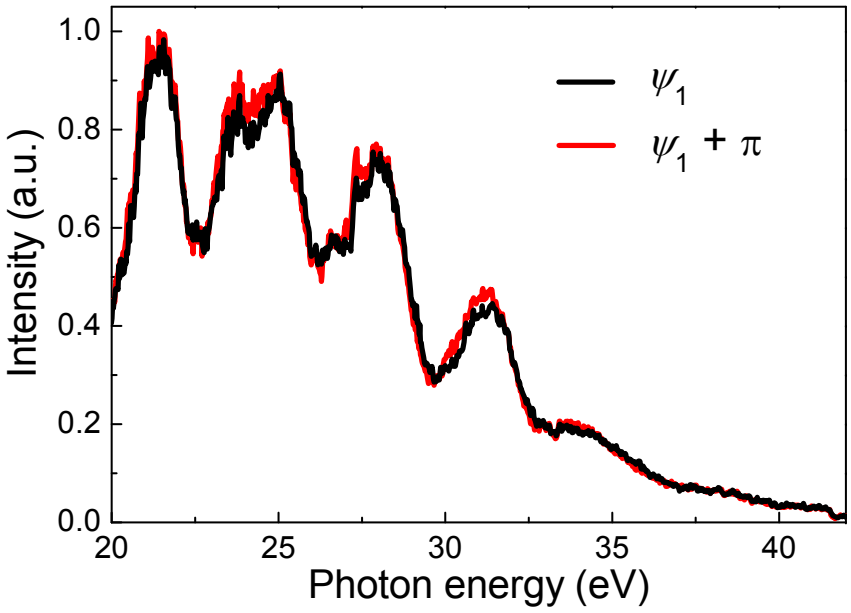
Requirements:

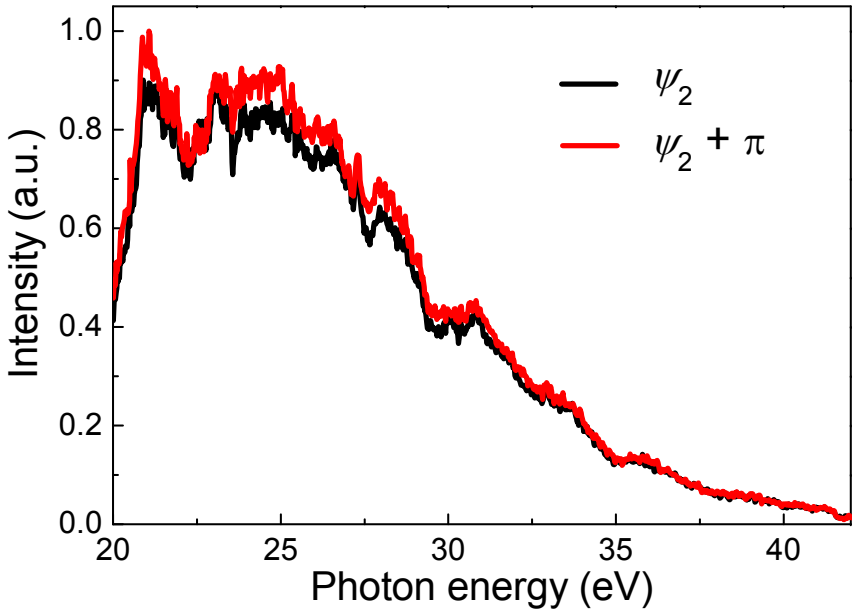
few cycle pulses
peak intensity > saturation intensity
CEP control
low gas pressure
spatial filtering after the gas cell



XUV spectra vs CEP: Xenon

- pulse duration τ = 5 fs; peak intensity 2.3×10^{15} W/cm^2; 2.5-mm xenon cell



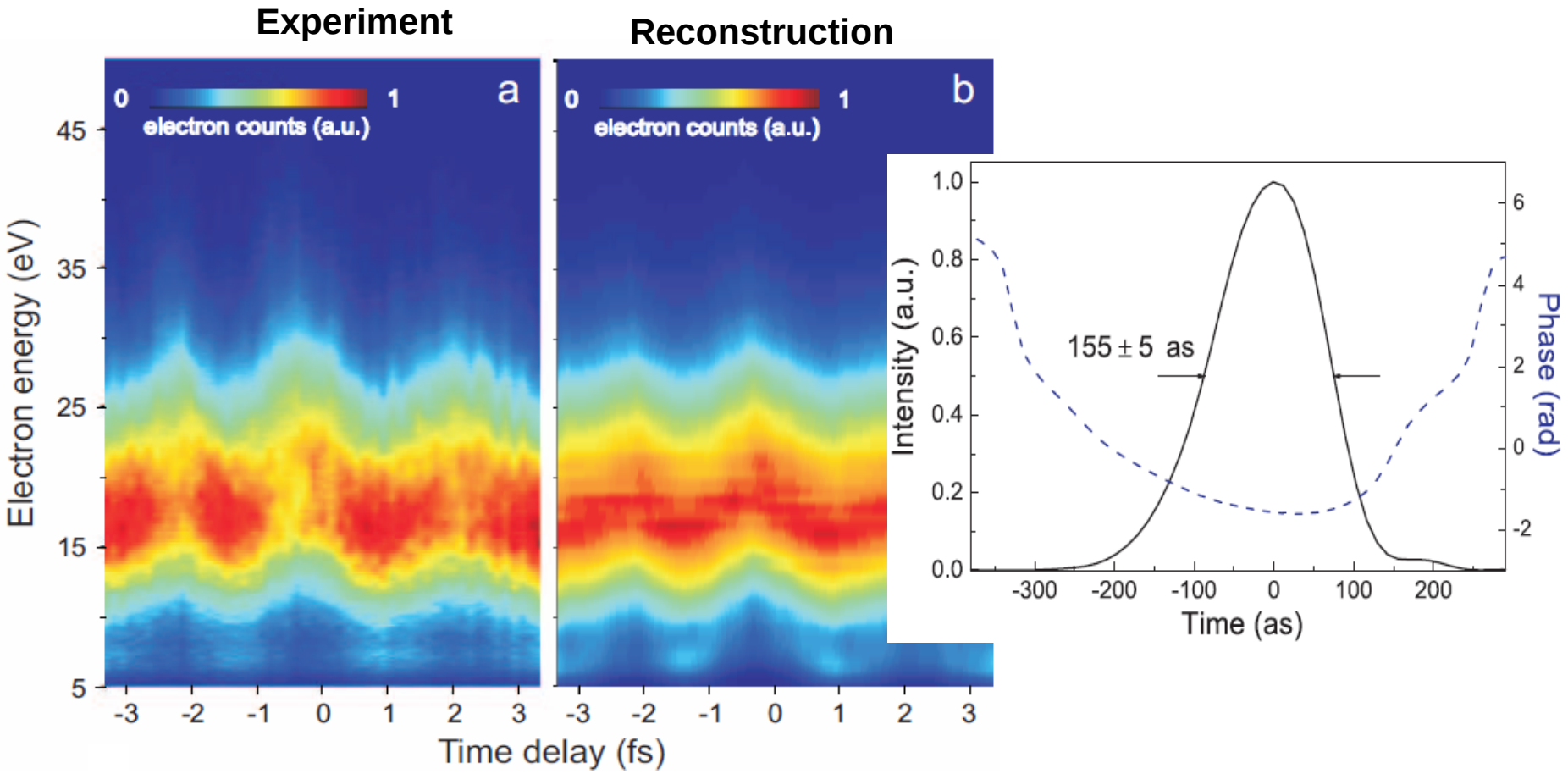


- ➔ Periodic change of amplitude and shape for $\Delta\psi = \pi$
- ➔ CEP drives transition from double to single emission
- ➔ Measured pulse energy *on target* 2.1 nJ

F. Ferrari *et al.*, Nature Photon. **4**, 875 (2010)

Temporal characterization

- FROG CRAB measurement



➔ Isolated attosecond pulses

➔ Near-single cycle pulses

F. Ferrari *et al.*, Nature Photon. **4**, 875 (2010)

Applications of Attosecond Pulses

- Status and prospects of attosecond spectroscopy and control
  - ➔ isolated atoms: a few examples (Kr, Ne, Xe, He, Ar)
  - ➔ simple diatomic molecules: 1 example (H_2/ D_2)
  - ➔ condensed matter: 1 example (tungsten crystal)
  - ➔ complex (bio)molecules and supramolecular assemblies
  - ➔ nanostructures
- Use of synthesized (waveform-controlled) pulses to steer electrons in molecules on the electronic time scale

Charge migration in molecules

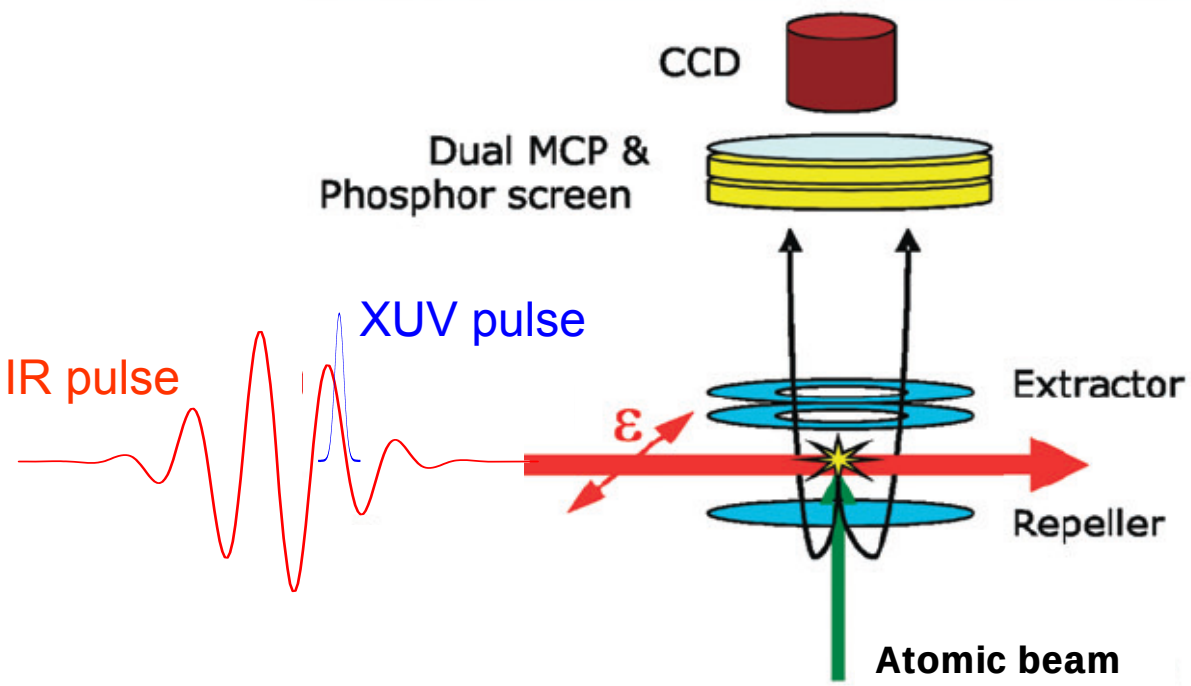
➔ Method: measurement of angular asymmetries in momentum distributions of fragments resulting from dissociative ionization

- Excitation of D_2 with isolated attosecond pulses in the presence of few-cycle IR laser field

  ➔ observation of electron localization following attosecond molecular photoionization

Velocity Map Imaging

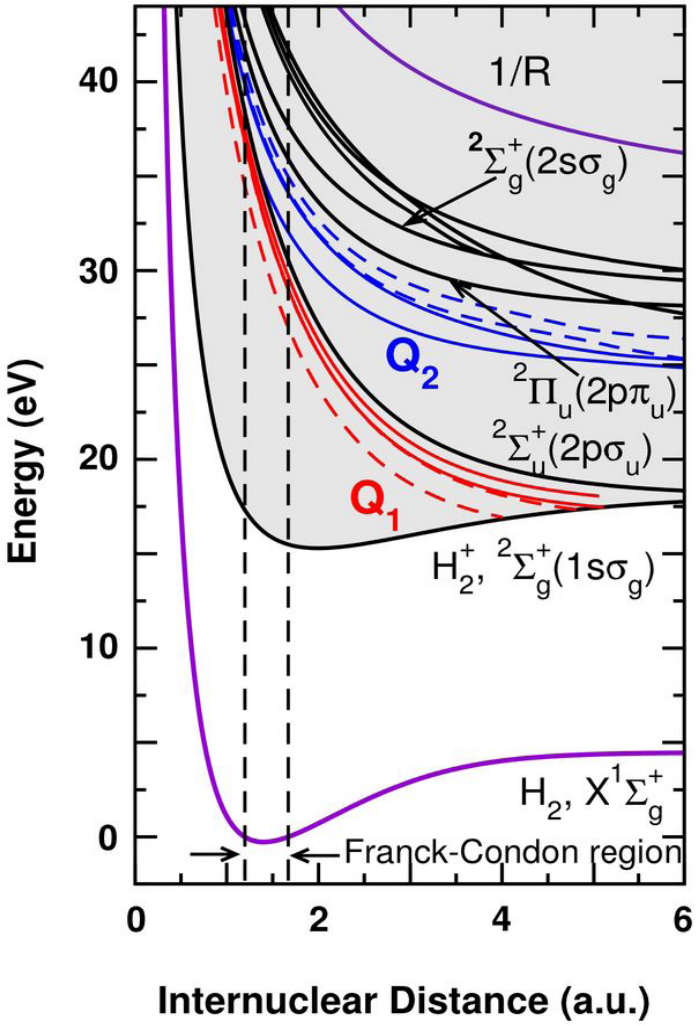
- ➔ very high collection efficiency (up to 100%)
- ➔ energy AND angular information

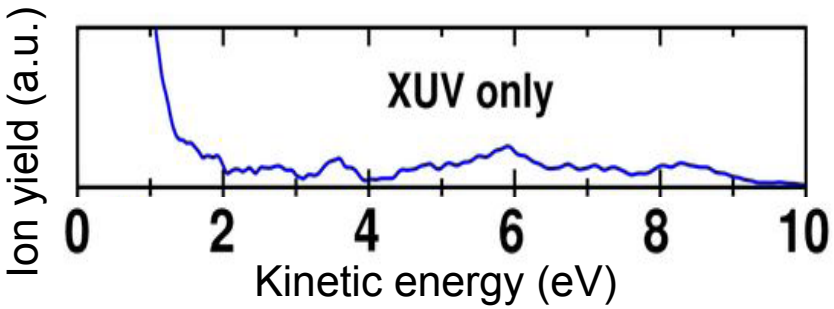


A.T.J.B. Eppink and D.H. Parker, Rev. Sci. Instrum. **68**, 3477 (1997)

Excitation by isolated attosecond pulses

- Several pathways lead to dissociative ionization
  (XUV spectrum between 20 and 40 eV)





< 1 eV: direct dissociative ionization via $1s\sigma_g$ state

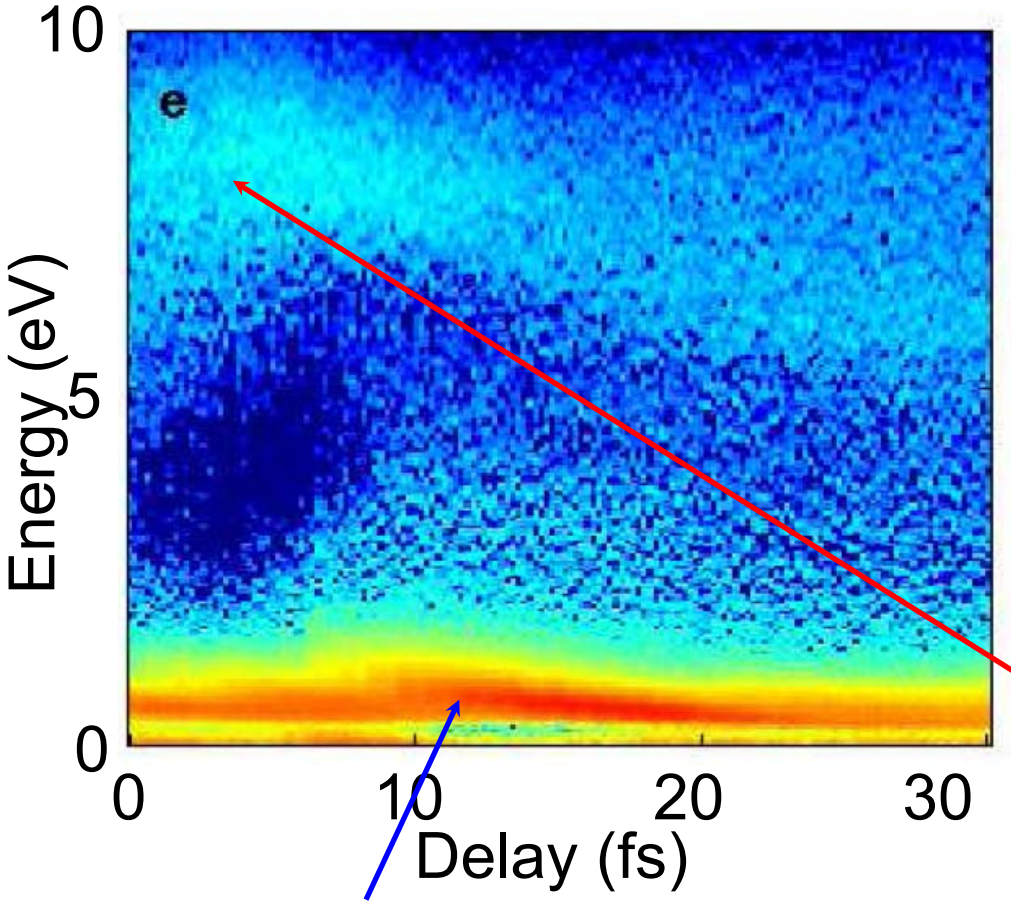
0 - 10 eV (primarily 2-7eV): auto-ionization of doubly excited Q_1 state to $1s\sigma_g$

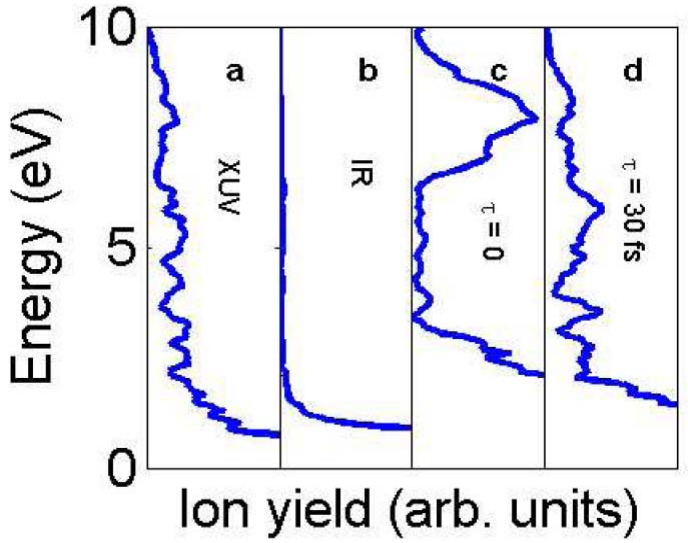
> 5 eV: direct excitation of $2p\sigma_u$

1 – 8 eV: auto-ionization of Q_2 state to $2p\sigma_u$ and $1s\sigma_g$

Isolated as pulse + few-cycle IR pulse

- D^+ kinetic energy distribution vs time delay





IR-induced ionization of Q_1 states producing $2p\sigma_u$ state

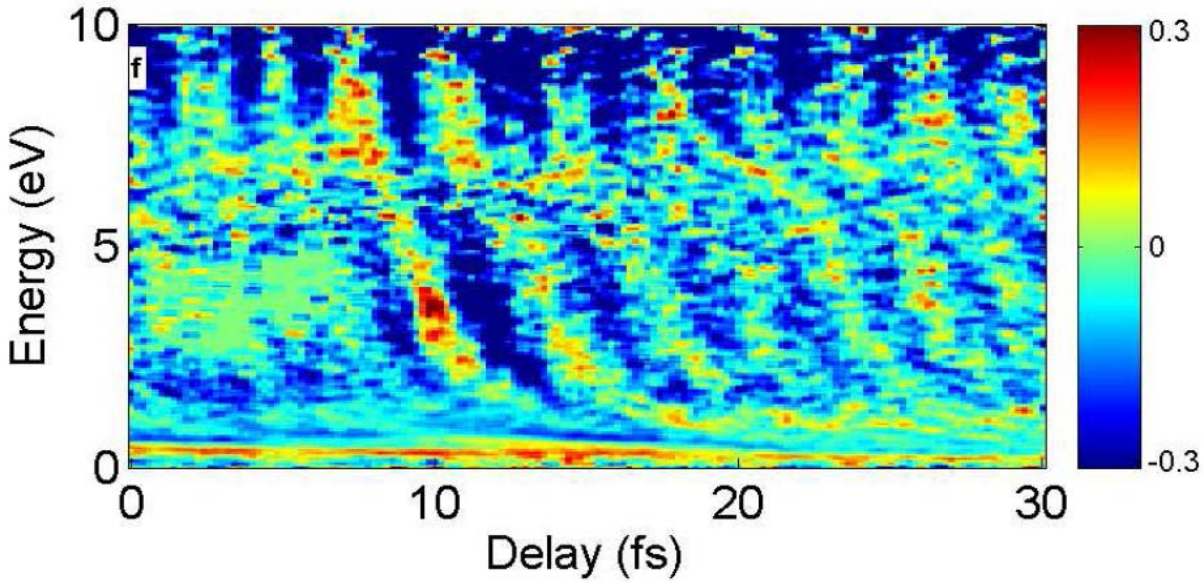
increase of excitation cross-section of $2p\sigma_u$ continuum due to IR-induced mixing of $2p\sigma_u$ and $1s\sigma_g$ states

Bond softening induced by IR pulse (maximum when bound WP is at the outer turning point of the potential curve)

G. Sansone *et al.*, Nature **465**, 763 (2010)

Electron localization in D_2

- Asymmetry parameter: $A(E_k,\tau)=\dfrac{N_L(E_k,\tau)-N_R(E_k,\tau)}{N_L(E_k,\tau)+N_R(E_k,\tau)}$



- ➔ First example of a molecular attosecond pump-probe experiment
- ➔ Electron localization is shown to rely on two mechanisms
- ➔ Prospect: investigation of complex molecular dynamics beyond the Born-Oppenheimer approximation.

G. Sansone *et al.*, Nature **465**, 763 (2010)

From diatomic to complex (bio)molecules

- Attosecond-scale electronic dynamics in molecules affect chemical changes

  L.S. Cederbaum, J. Zobeley, Chem. Phys. Lett. **307**, 205 (1999)
  F. Remacle, R.D. Levine, PNAS **103**, 6793 (2006)

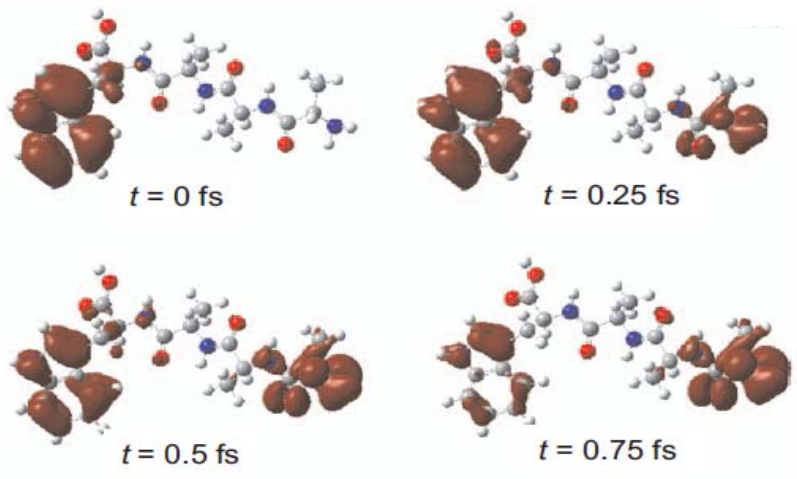
- When *charge migration* is the crucial step, the time-scale relevant to chemistry is set by electronic motion
  - ➔ electron delocalization in aromatic molecules
  - ➔ photosynthesis
  - ➔ long-range electron transfer in biomolecules
  - ➔ biological energy conversion processes

- Molecular electronics and molecular photovoltaics

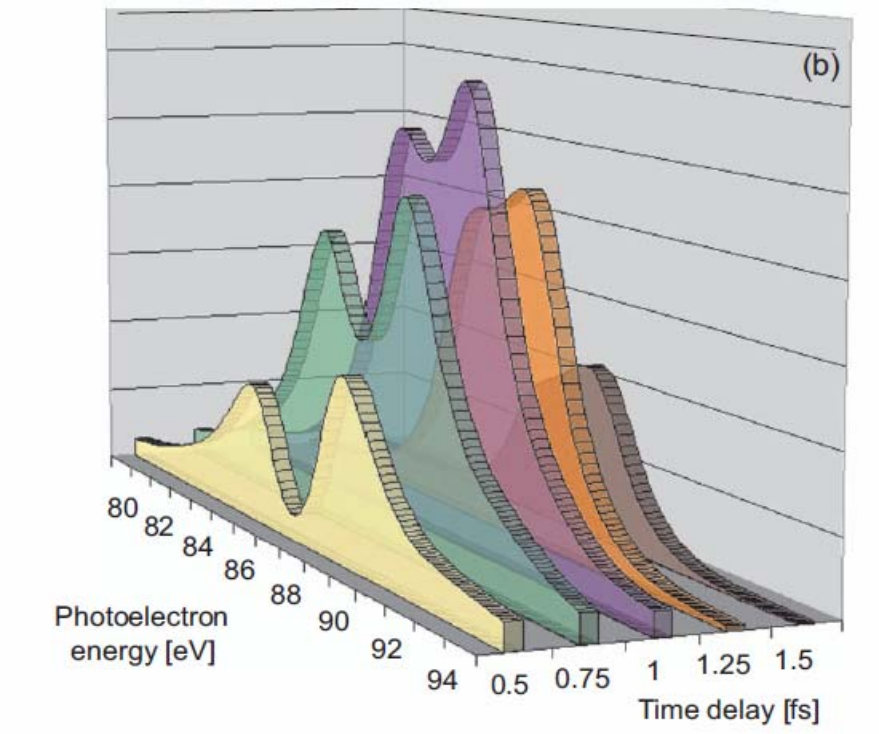
  Control of electronic current in ever smaller semiconductor nanostructures and molecular systems

Charge migration in small peptides

- Charge localization by sudden ionization of small peptides: the resulting hole is localized and is not stationary (the stationary orbitals of the cation are delocalized)



proposed experimental technique: measure of kinetic energy distribution of photoelectrons released by a time-delayed sub-fs XUV pulse (250 as, 95 eV)



F. Remacle, R.D. Levine, Z. Phys. Chem. **221**, 647 (2007)

Conclusions

- Generation and characterization of isolated attosecond pulses by temporal gating techniques
- Application to molecular physics
  - ➔ observation of electron localization following attosecond molecular photoionization
  - ➔ first experimental observation and control of attosecond dynamics in molecules

Involved people

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Post-doc:

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Anne L'Huillier (Lund, Sweden)
Matthias Kling (MPQ, Garching, Germany)
F. Kelkensberg, W. K. Siu (AMOLF, Amsterdam, The Netherlands)
Frank Lépine (University of Lyon, France)