Istituto di Fotonica e Nanotecnologie

From phase stable pulses to Attosecond Science

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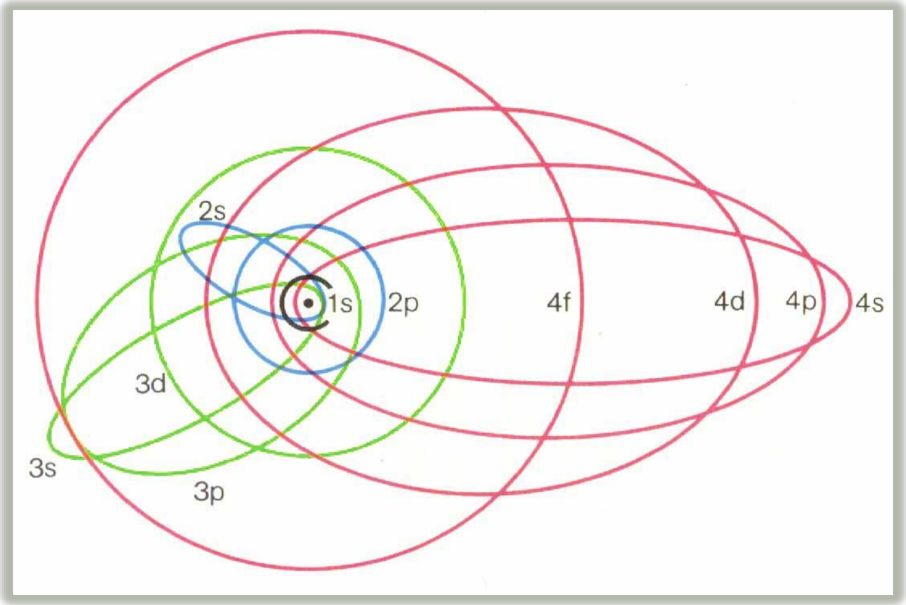
ICTP WINTER COLLEGE
Trieste, 24 February 2016

Why is this attosecond business interesting??

real-time observation and direct control of electronic motion in atoms, molecules and solids!!



Bohr-model of hydrogen atom:

electron in the ground state moves in a circular classical orbit about the nucleus in ~ 150 as

Attosecond resolution is required!

Outline

I. Generation of attosecond pulses

II. Attosecond metrology

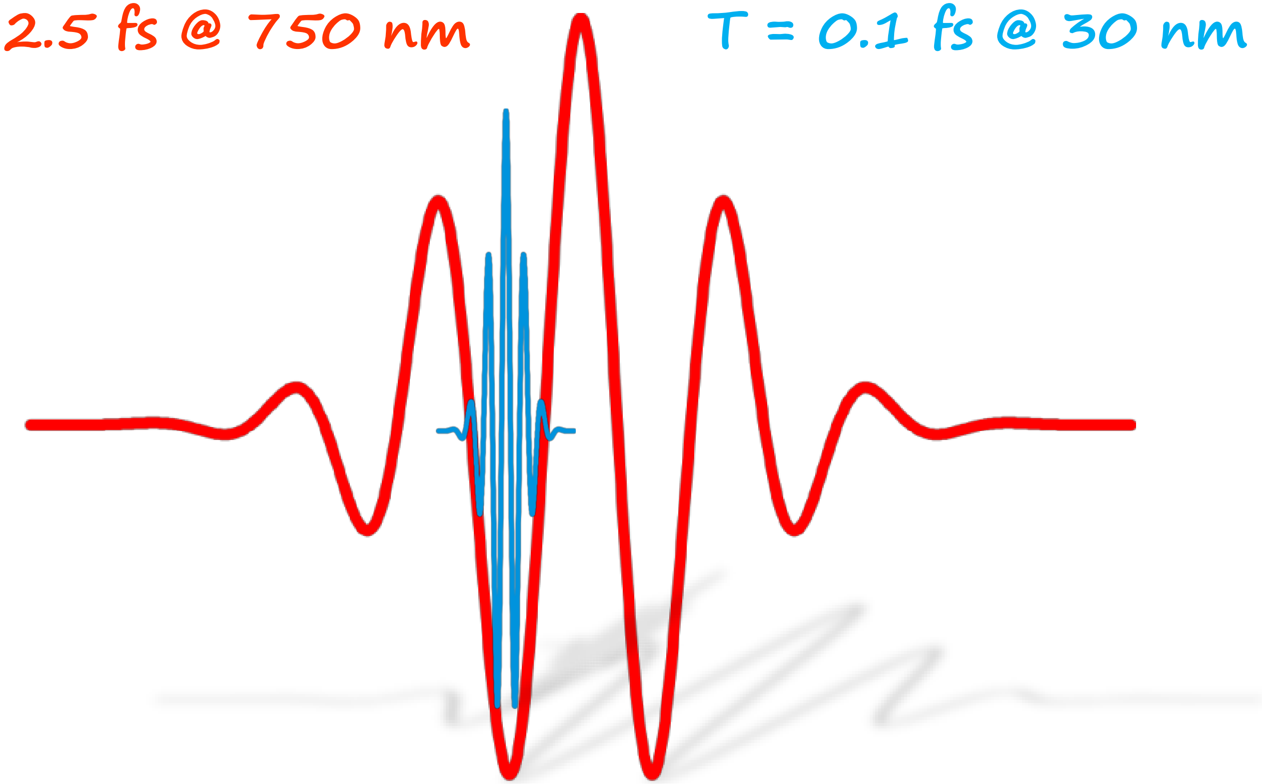
III. Applications

I.
Generation of attosecond pulses

Sub-femtosecond pulses

T = 2.5 fs @ 750 nm

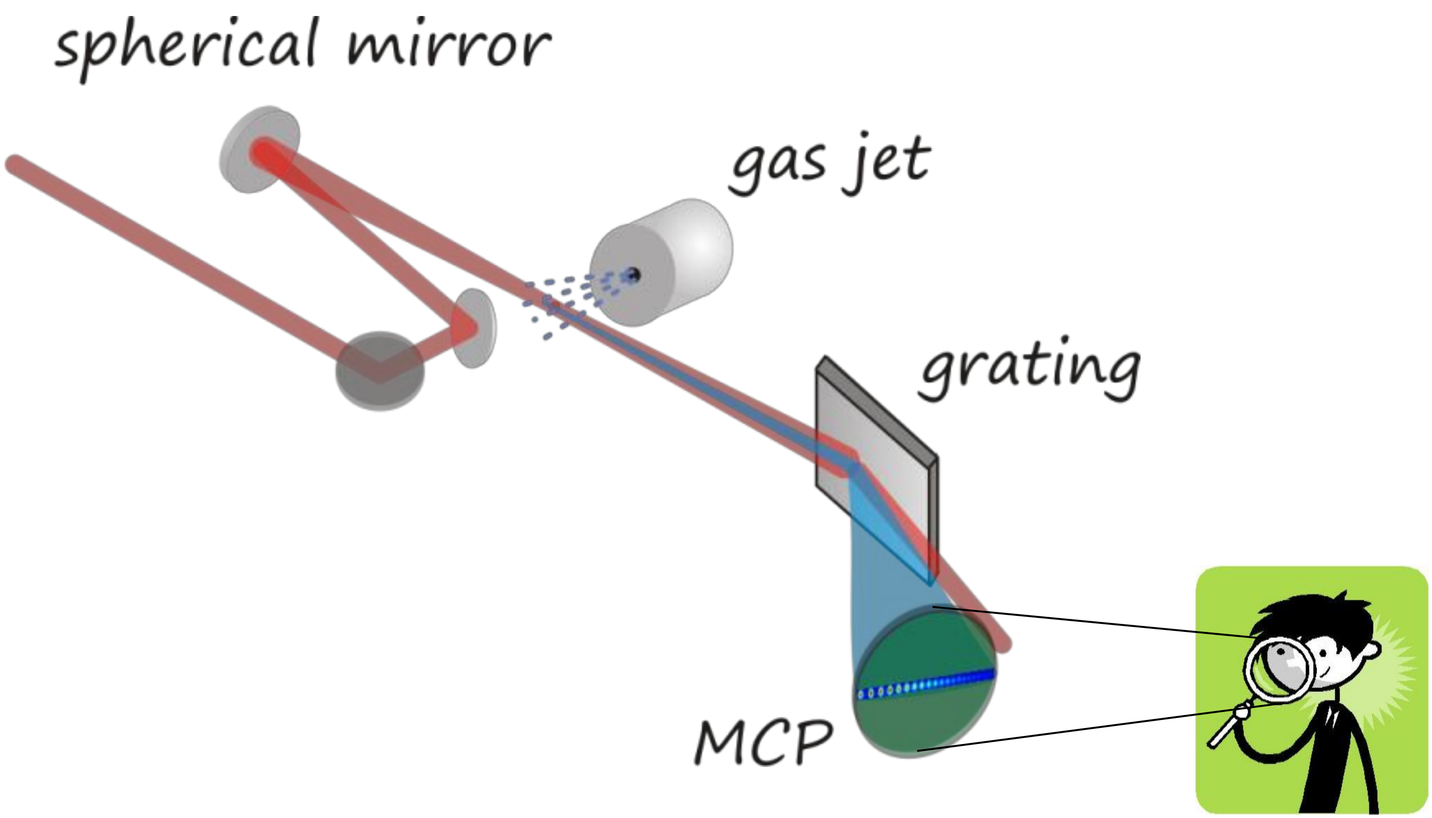
T = 0.1 fs @ 30 nm

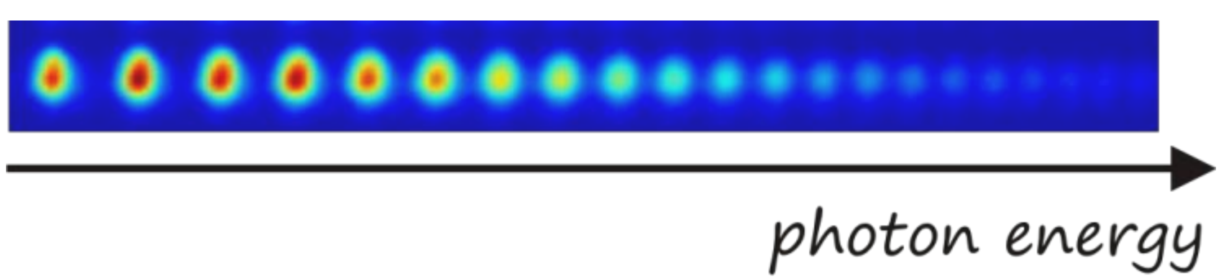


light pulses in the XUV are required

→ high-order harmonic generation

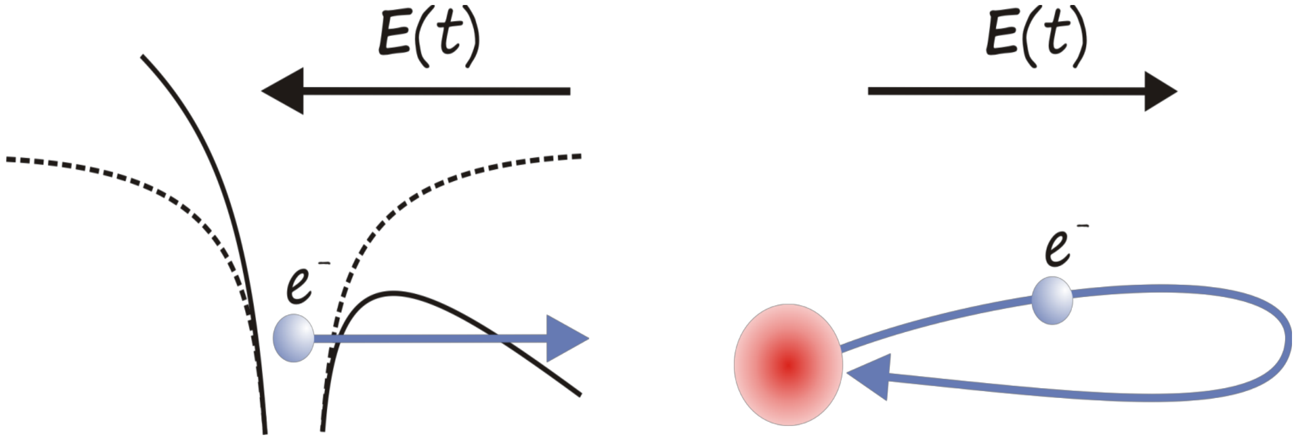
High-order Harmonic Generation





3 step model

P. B. Corkum, Phys. Rev. Lett. 71, 1994 (1993)

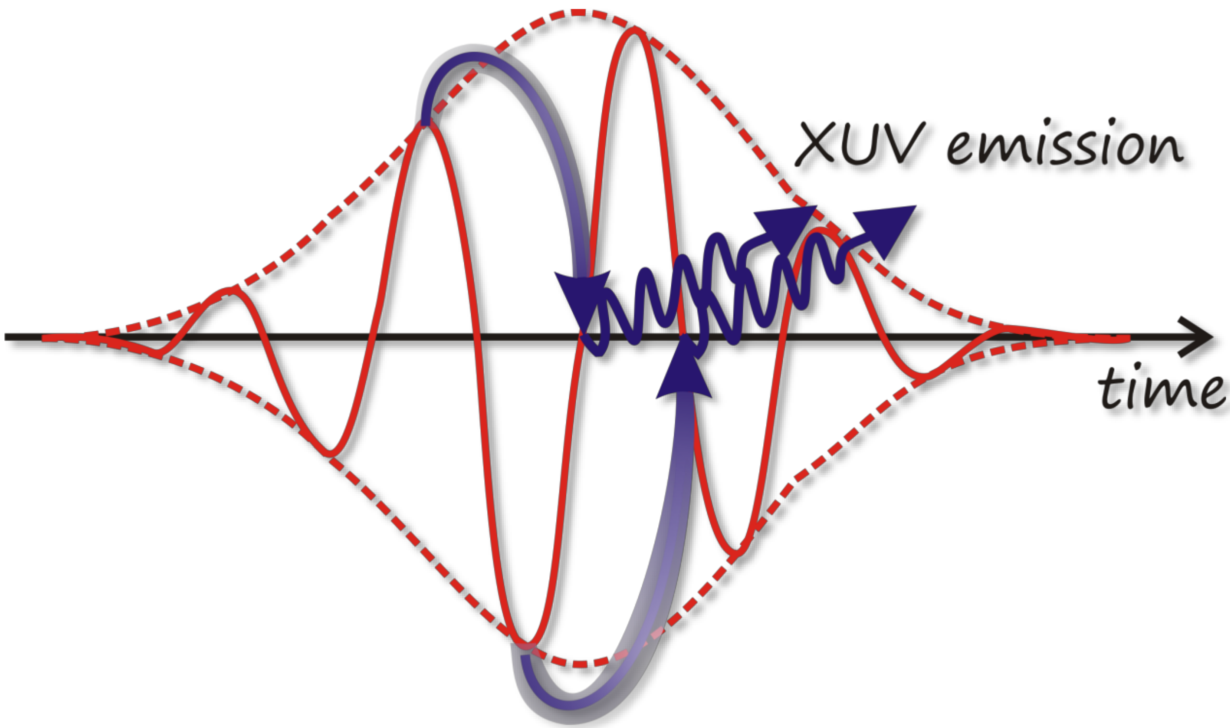


I. Ionization: the laser field detaches an electron from the valence shell via tunnel ionization

II. Propagation: the freed electron is accelerated by the laser field

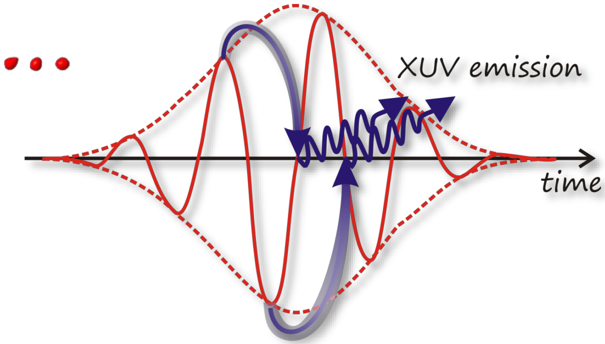
III. Recombination: the energy gained by the electron is released through the emission of a XUV photon

High-order Harmonic Generation

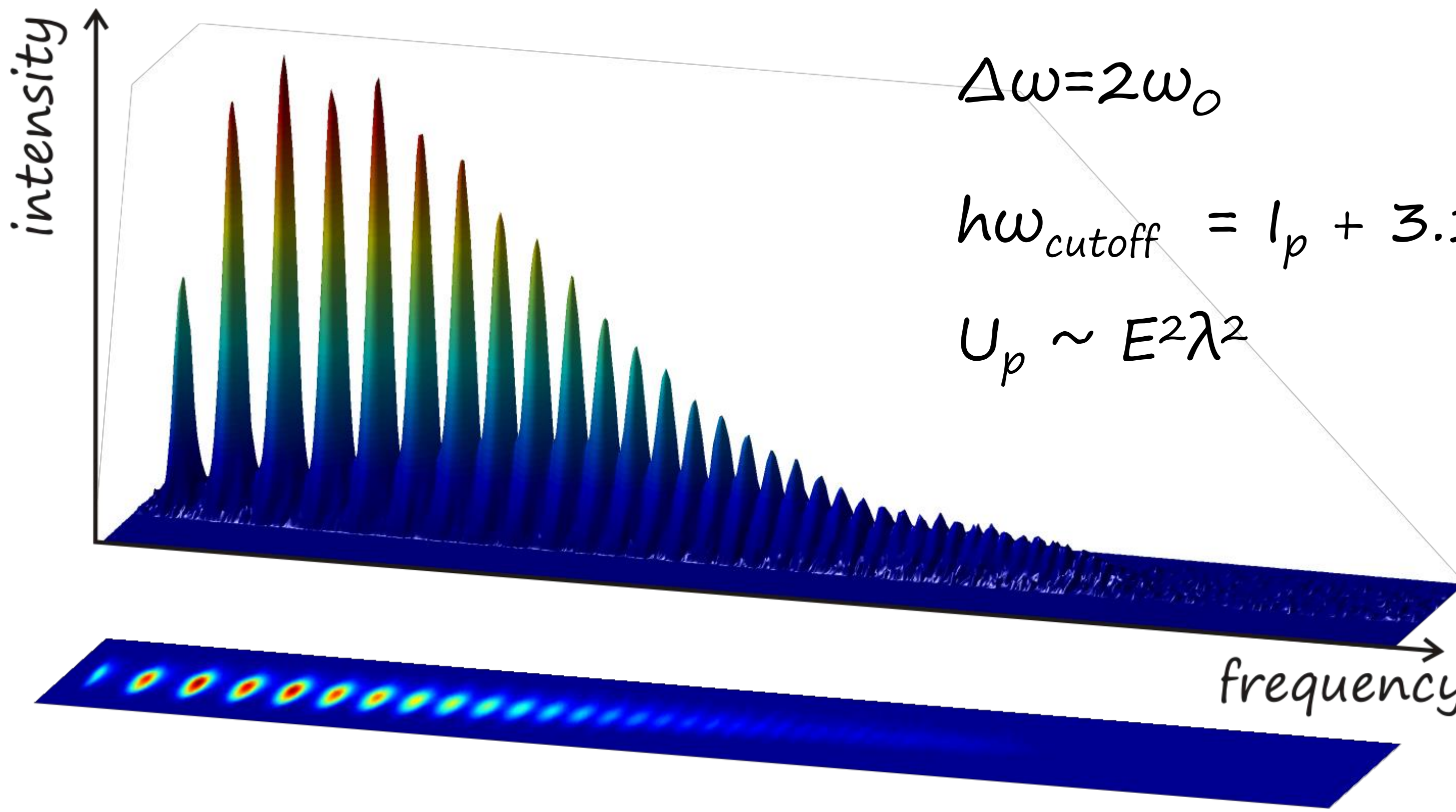


The process is repeated periodically every half cycle

In the spectral domain...



odd harmonics of the fundamental frequency

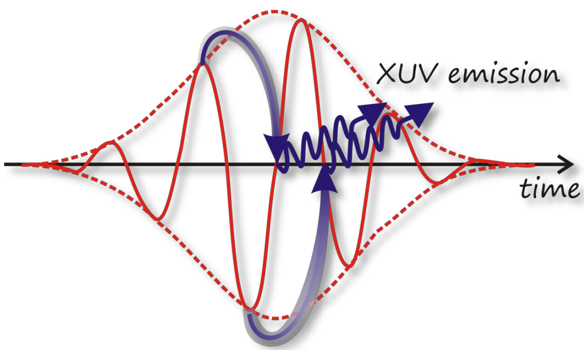


$\Delta\omega = 2\omega_0$

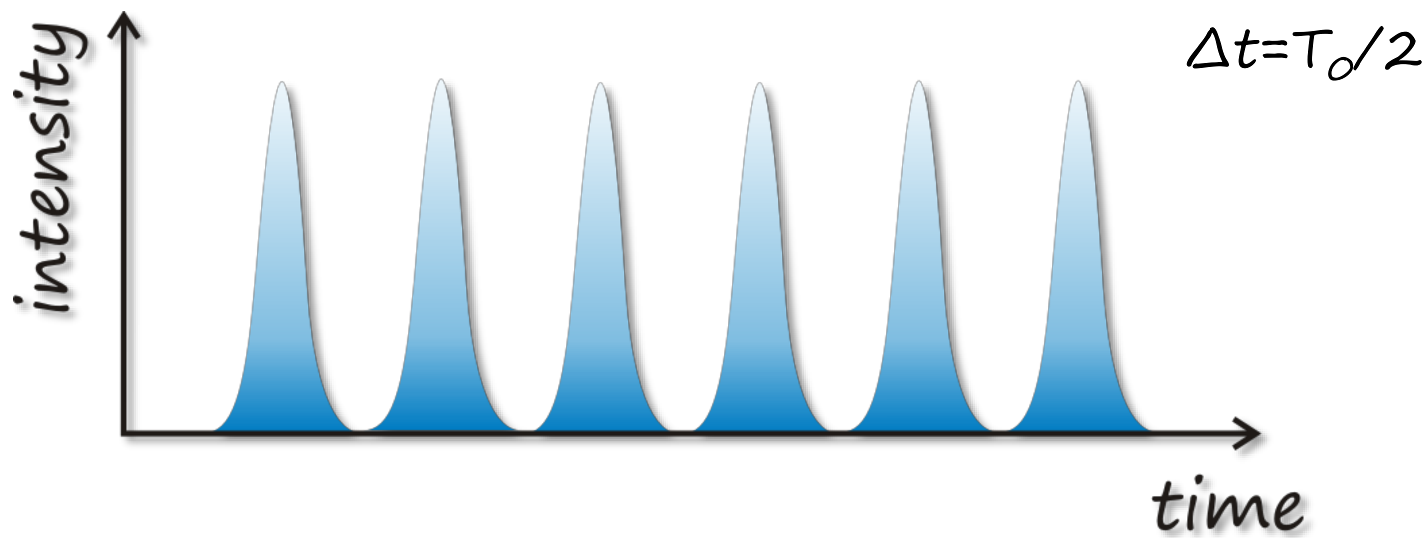
$h\omega_{cutoff} = I_p + 3.17\ U_p$

$U_p \sim E^2\lambda^2$

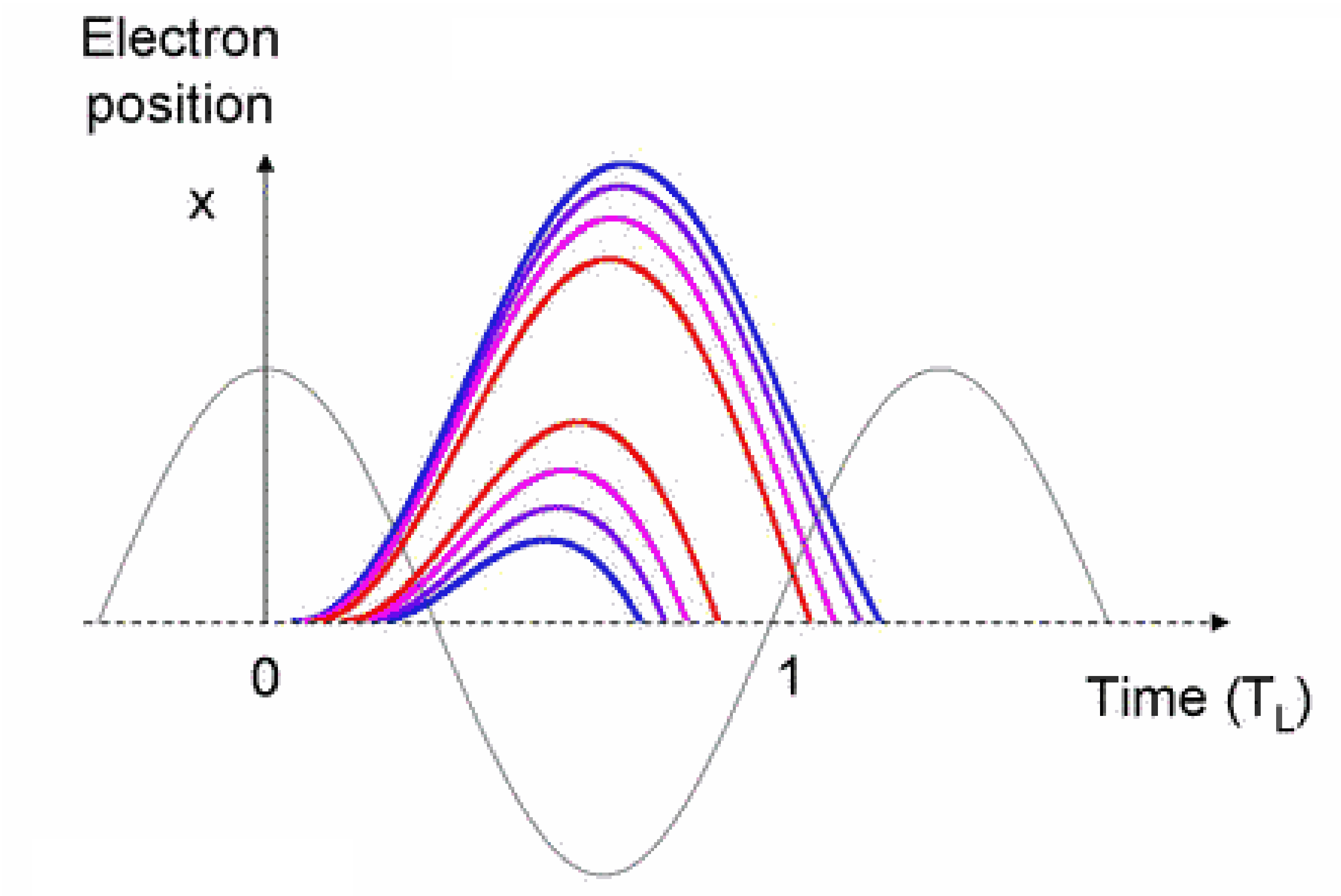
...and in the temporal domain



train of attosecond pulses



Electron trajectories



Lewenstein quantum model

- strong field approximation
- single active electron
  (from the outermost valence shell)

I. ionization   II. propagation

$$\vec{\mathcal{E}}_{XUV}[\Omega] \propto -(m\omega)^2 \int_{\mathbb{R}} \int_{\mathbb{R}+} \int_{\mathbb{R}^3} \boxed{\frac{e}{\hbar^4} \vec{E}(t-\tau) \cdot \vec{d}\left[\vec{k}(\vec{p}, t-\tau)\right]} \boxed{\vec{d}\left[\vec{k}(\vec{p}, t)\right]^*} \boxed{e^{-\frac{i}{\hbar}S(\vec{p},t,\tau)+i\Omega t}} \mathrm{d}\vec{p}\mathrm{d}\tau'\mathrm{d}t'$$

III. recombination

$$S(\vec{p}, t, \tau) = \int_{t-\tau}^{t} \frac{\left[\vec{p} + e\vec{A}(t')\right]^2}{2\mu} \mathrm{d}t' \qquad \vec{d}(\vec{k}) = \langle e^{i\vec{k}\cdot\vec{r}} | \vec{r} | \Gamma_0(\vec{r}) \rangle$$

Attosecond dynamics probed by HHG

- saddle point approximation (SPA)

I. ionization — II. propagation — III. recombination

$$\vec{\mathcal{E}}_{XUV}[\Omega] \propto -(\Omega)^2 \int_{\mathbb{R}} \int_{\mathbb{R}+} \int_{\mathbb{R}^3} \boxed{\frac{e}{\hbar^4} \vec{E}(t-\tau) \cdot \vec{d}\left[\vec{k}(\vec{p}, t-\tau)\right]} \boxed{\vec{d}\left[\vec{k}(\vec{p}, t)\right]^*} \boxed{e^{-\frac{i}{\hbar} S(\vec{p},t,\tau) + i\Omega t}} \mathrm{d}\vec{p}\mathrm{d}\tau'\mathrm{d}t'$$

$$\vec{\mathcal{E}}_{XUV}[\Omega] \propto -(\Omega)^2 \vec{E}(t_s - \tau_s) \cdot \frac{e}{\hbar^4} \vec{d}\left[\vec{k}(\vec{p_s}, t_s - \tau_s)\right] \vec{d}\left[\vec{k}(\vec{p_s}, t_s)\right]^* \int_{\mathbb{R}} \int_{\mathbb{R}+} \int_{\mathbb{R}^3} \boxed{e^{-\frac{i}{\hbar} S(\vec{p},t,\tau) + i\Omega t}} \mathrm{d}\vec{p}\mathrm{d}\tau'\mathrm{d}t'$$

- coupling between:

ionization time $t_s - \tau_s$

$$\frac{\partial S(p_s, t_s, \tau_s)}{\partial(-\tau)} = 0 = \frac{\left[\vec{p_s} + e\vec{A}(t_s - \tau_s)\right]^2}{2\mu} + I_p$$

recombination time t_s

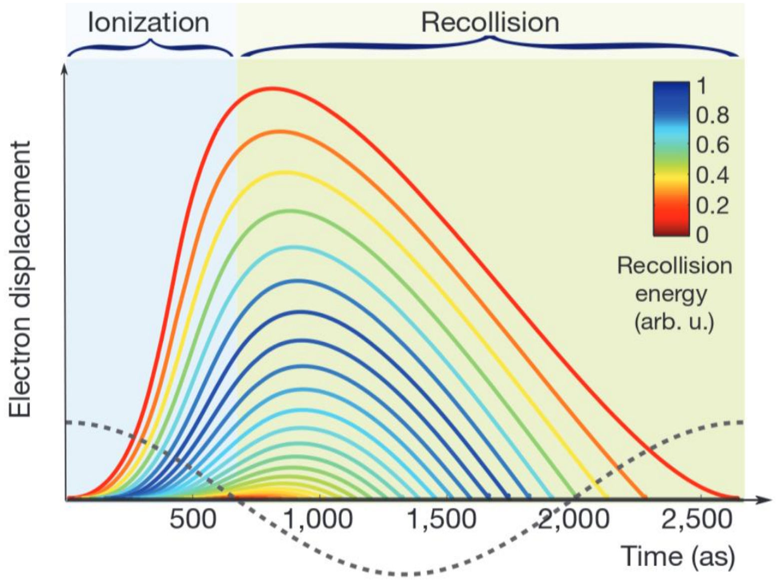
$$\frac{\partial S(p_s, t_s, \tau_s)}{\partial t} = 0 = \frac{\left[\vec{p_s} + e\vec{A}(t_s)\right]^2}{2\mu} + I_p - \hbar\Omega$$

photon energy $\hbar\Omega$

$$\vec{\nabla}_{\vec{p}} S(p_s, t_s, \tau_s) = 0 = \int_{t_s - \tau_s}^{t_s} \frac{\left[\vec{p_s} + e\vec{A}(t')\right]}{\mu} dt'$$

Attosecond dynamics probed by HHG

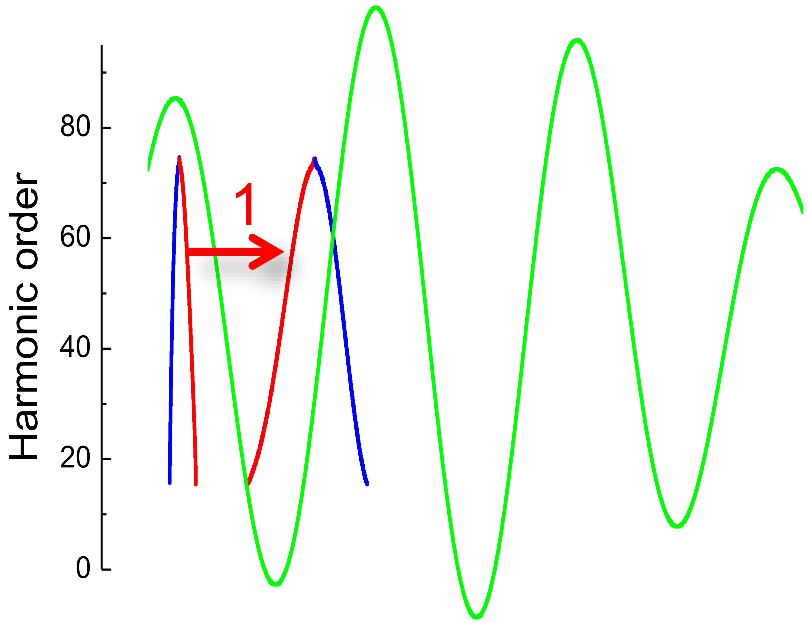
Each saddle point solution defines a quantum trajectory

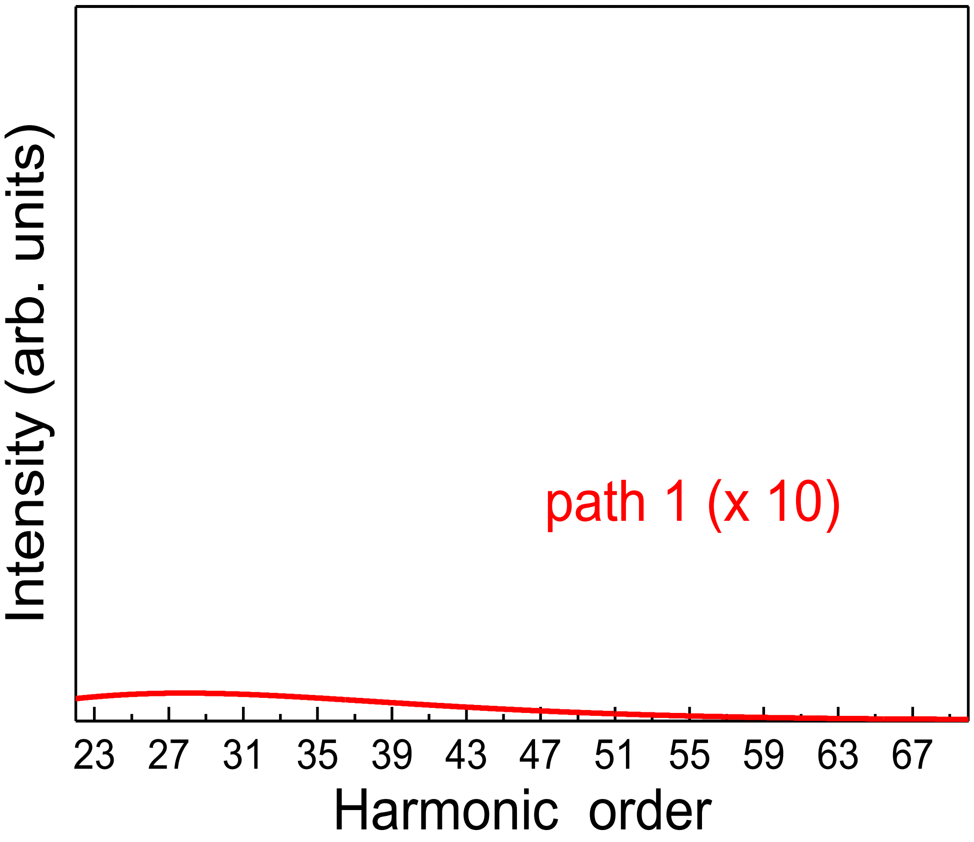


Shafir et al. Nature 485, 343 (2012)

THE ATTOSECOND NATURE OF THE PROCESS IS MAPPED INTO THE HHG SPECTRUM

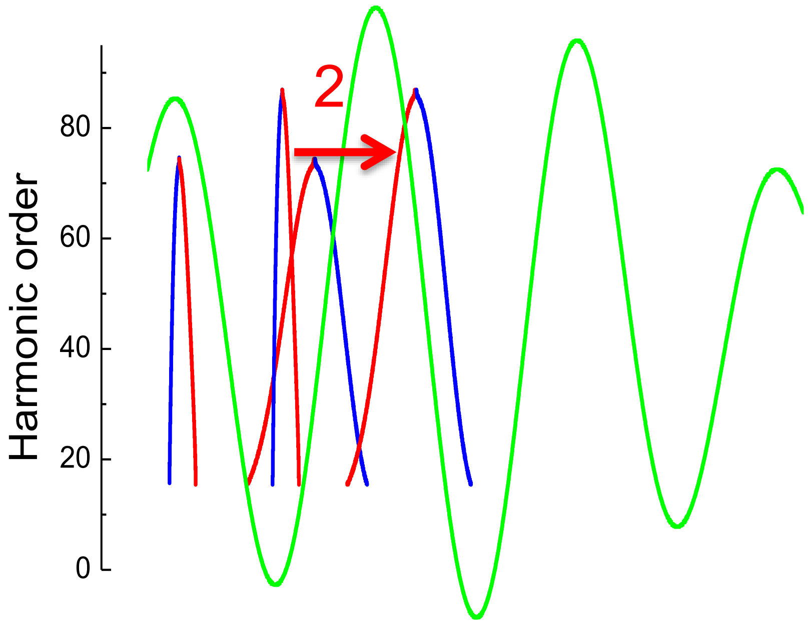
HHG quantum paths

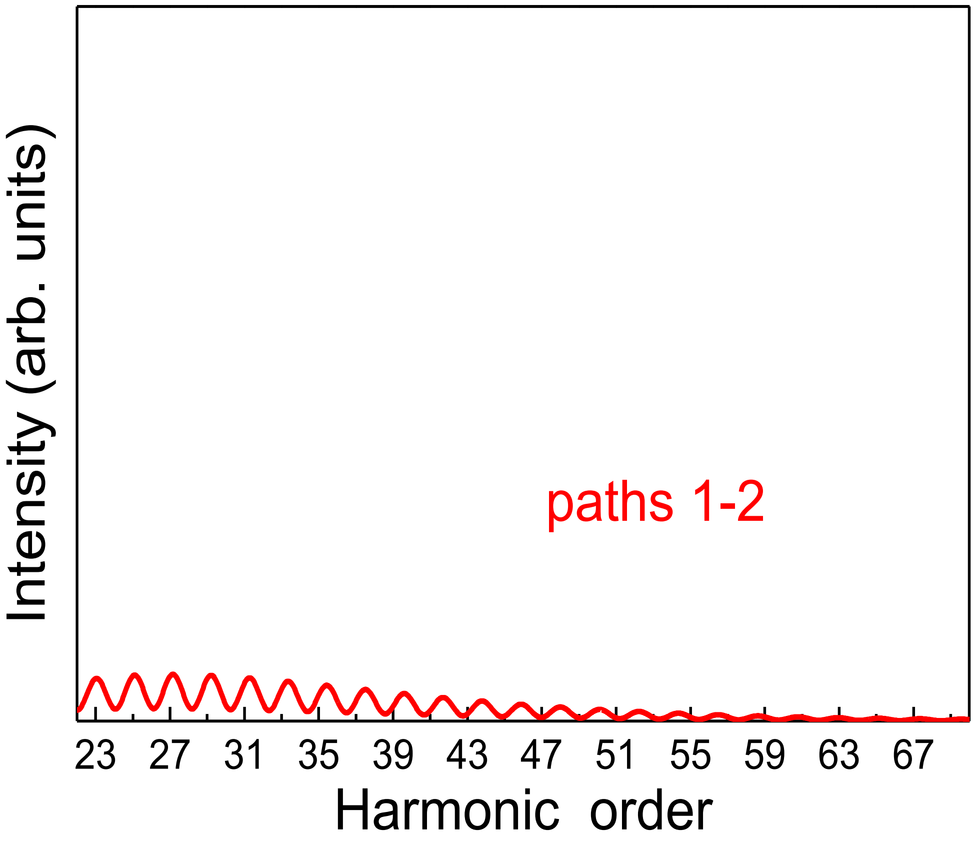




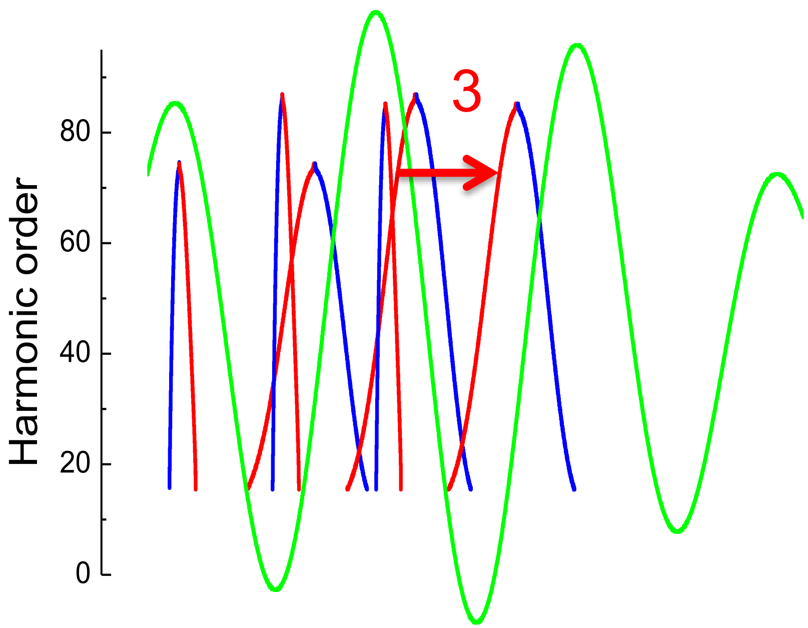
G. Sansone et al., Phys. Rev. A 70, 013411 (2004)

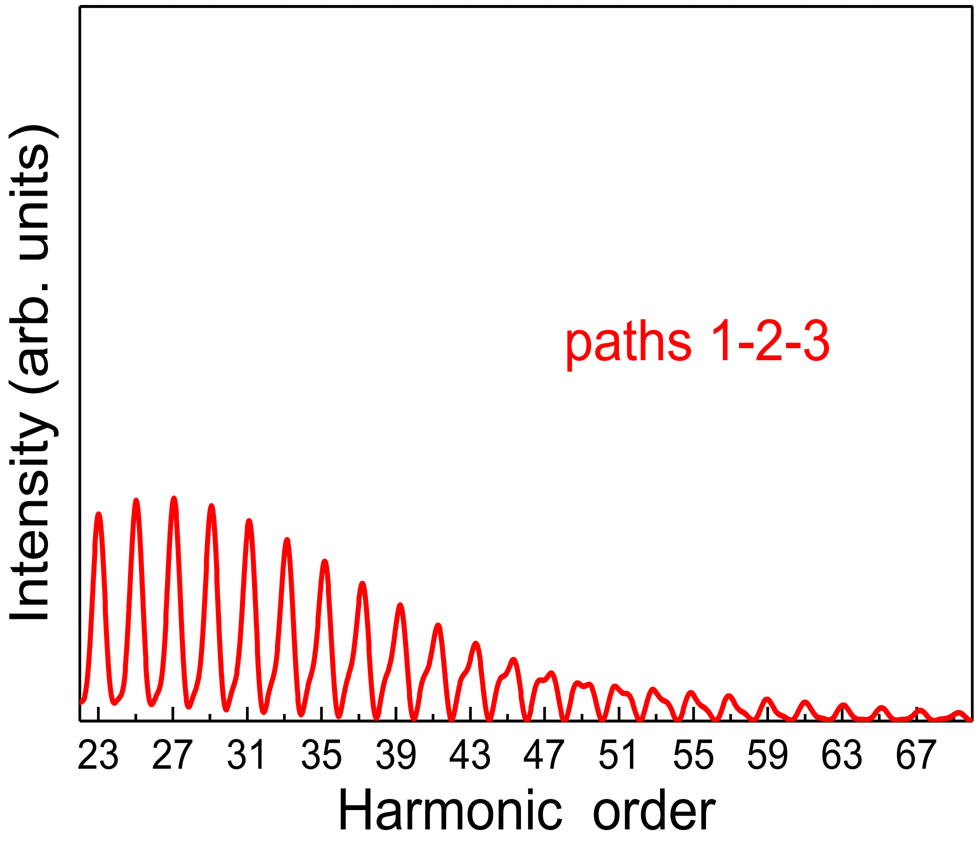
HHG quantum paths



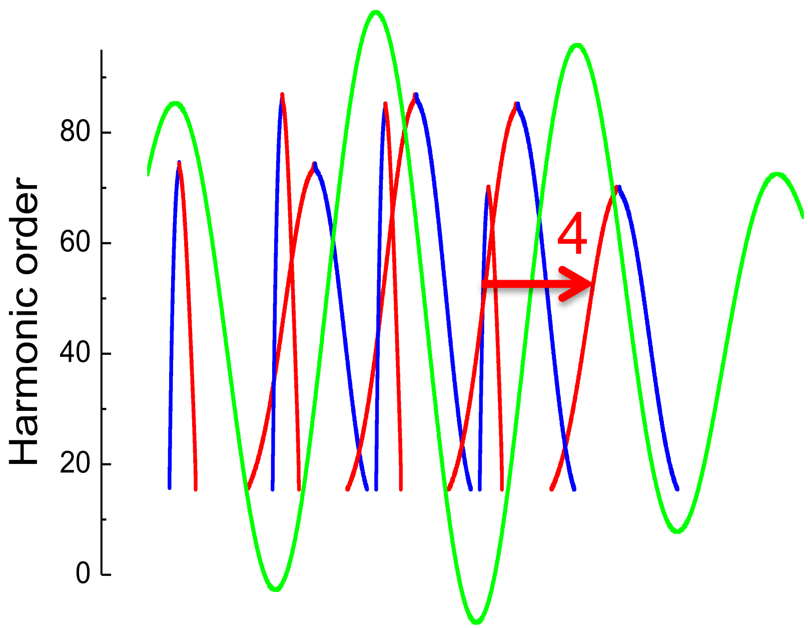


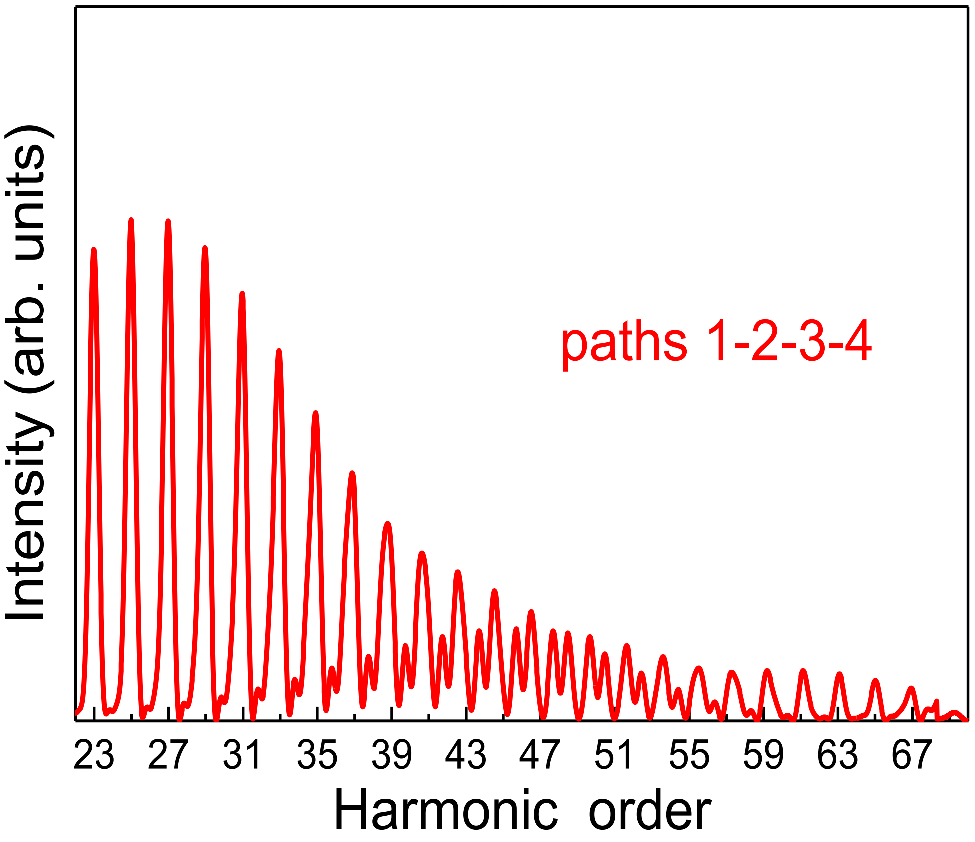
HHG quantum paths



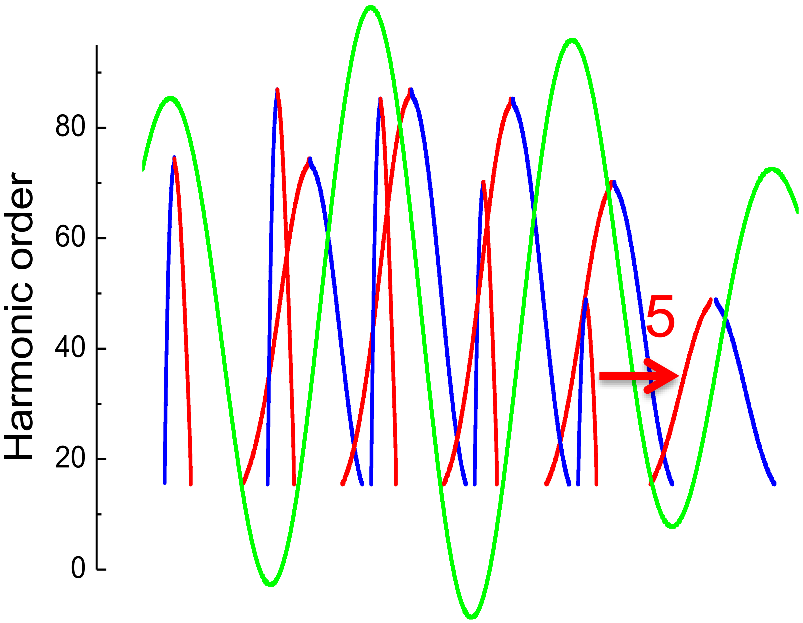


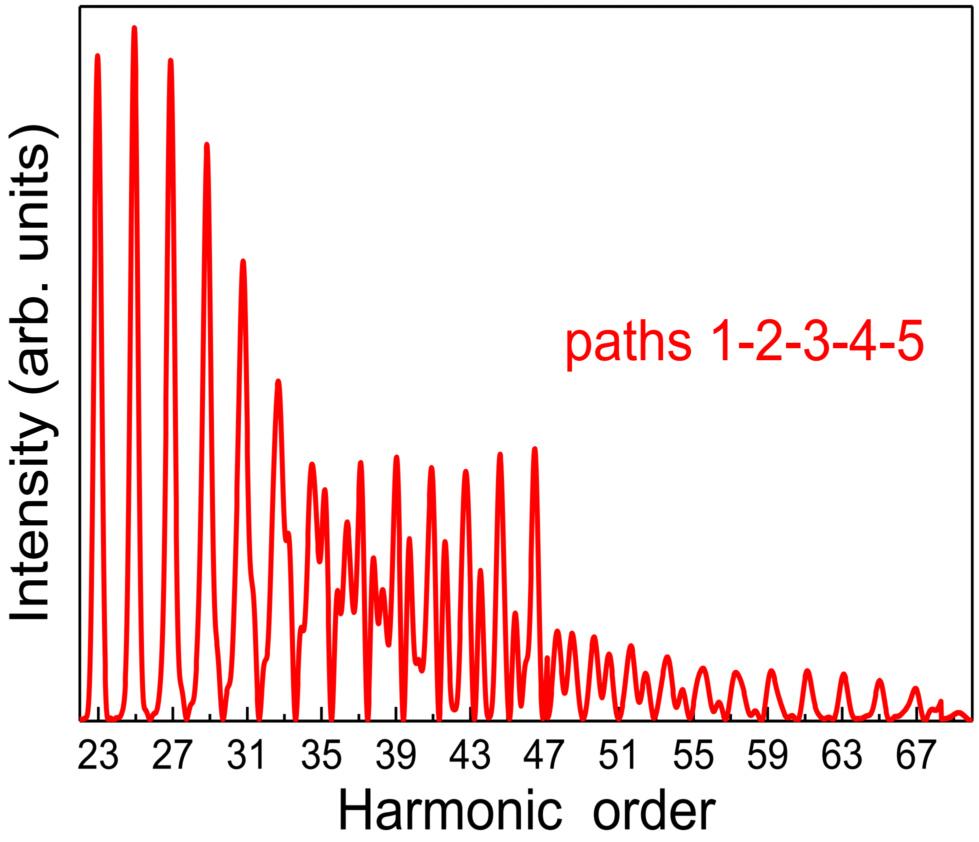
HHG quantum paths



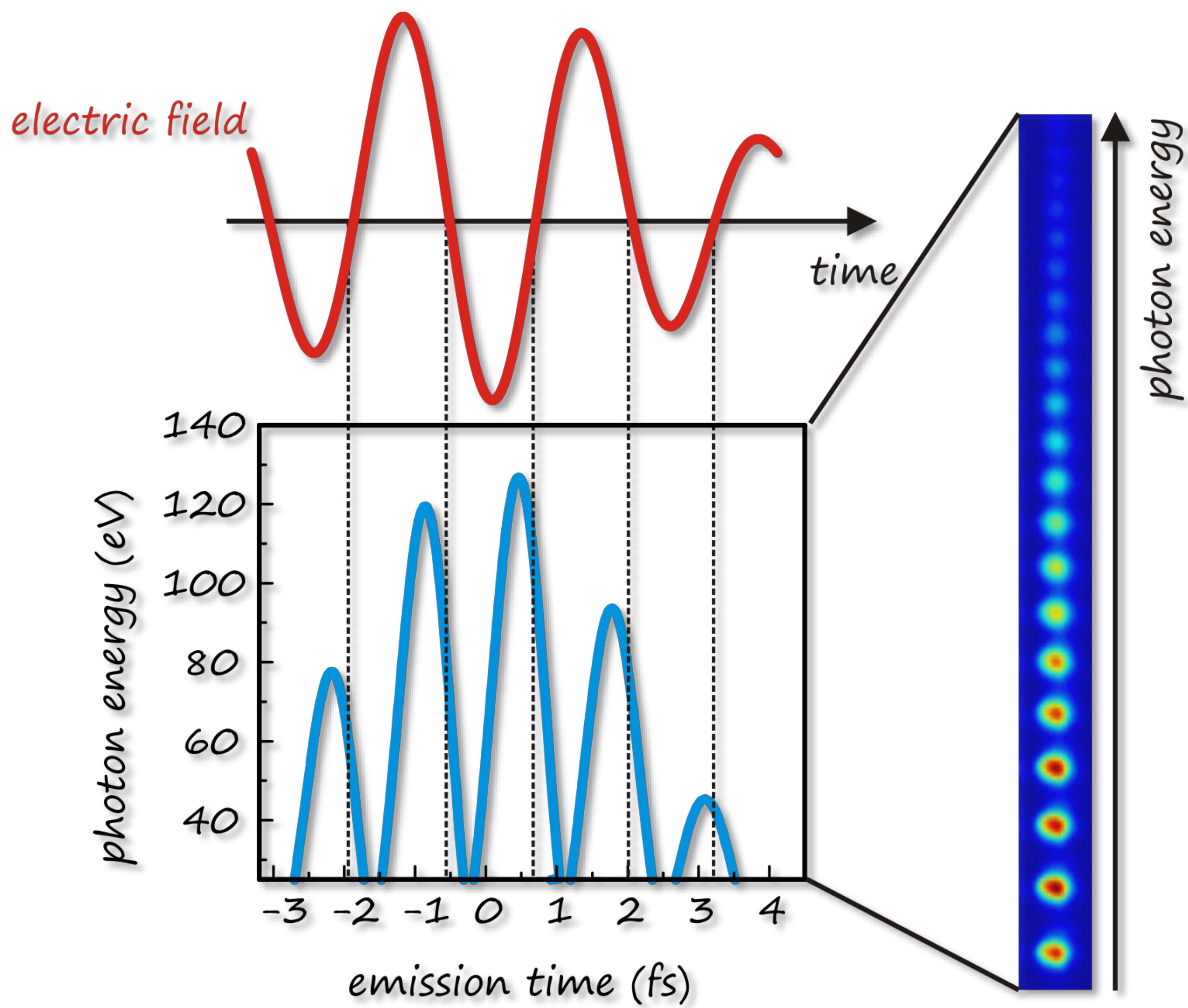


HHG quantum paths





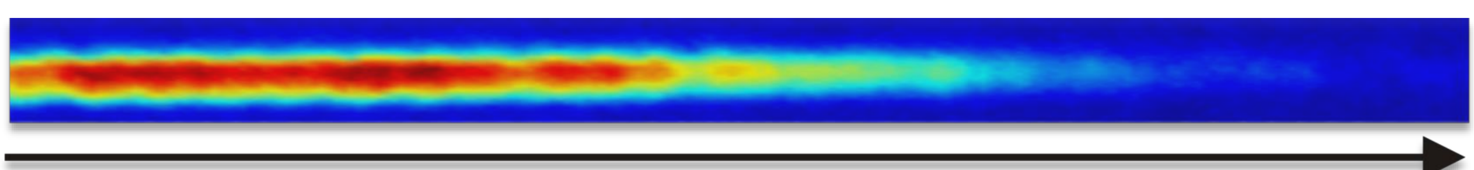
from a train of attosecond pulses...



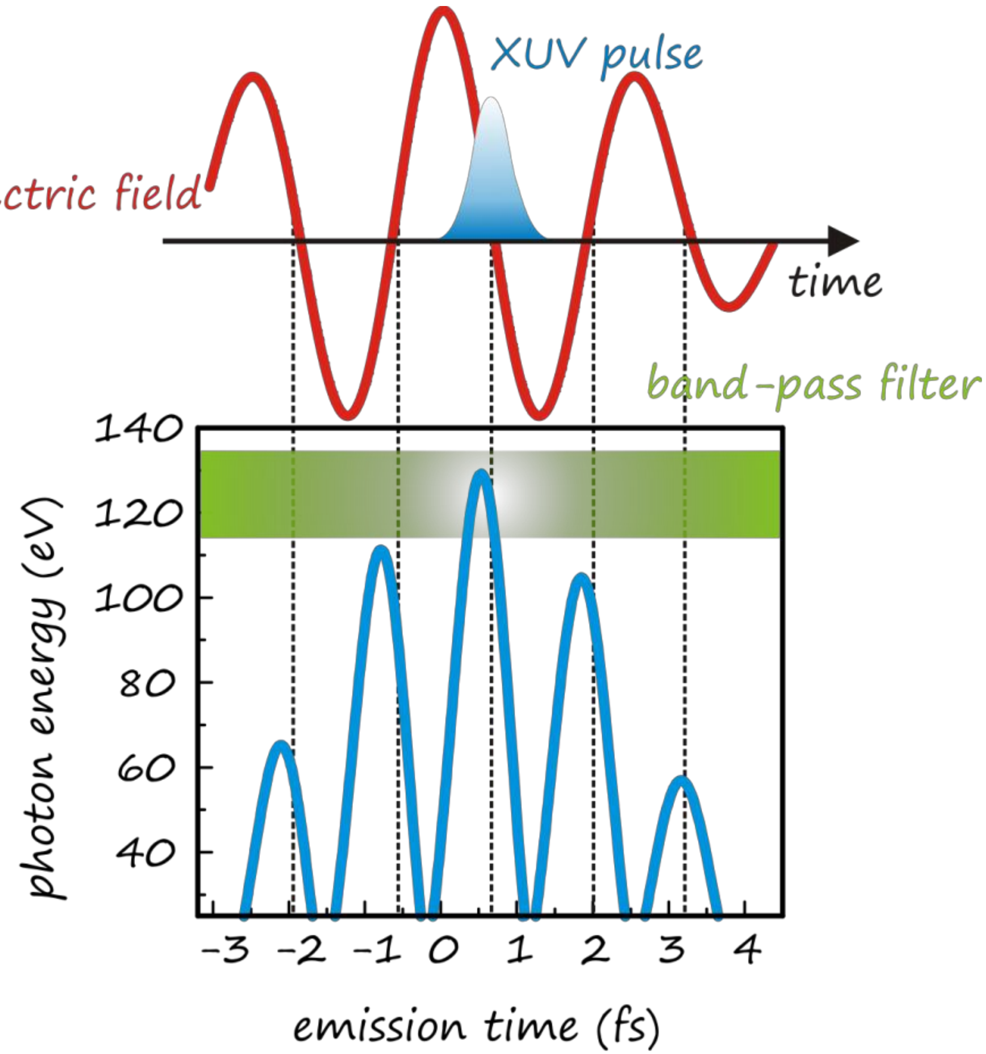
...to a single attosecond pulse

Single Attosecond Pulse recipe:

- selection of only one emission event:
  - spectral selection
  - temporal gating
- attochirp compensation
- CEP stability
- few-cycle pulses (most of the time...)



Spectral selection



spectral selection of cutoff photons leads to generation of SAP

<u>requirements</u>:

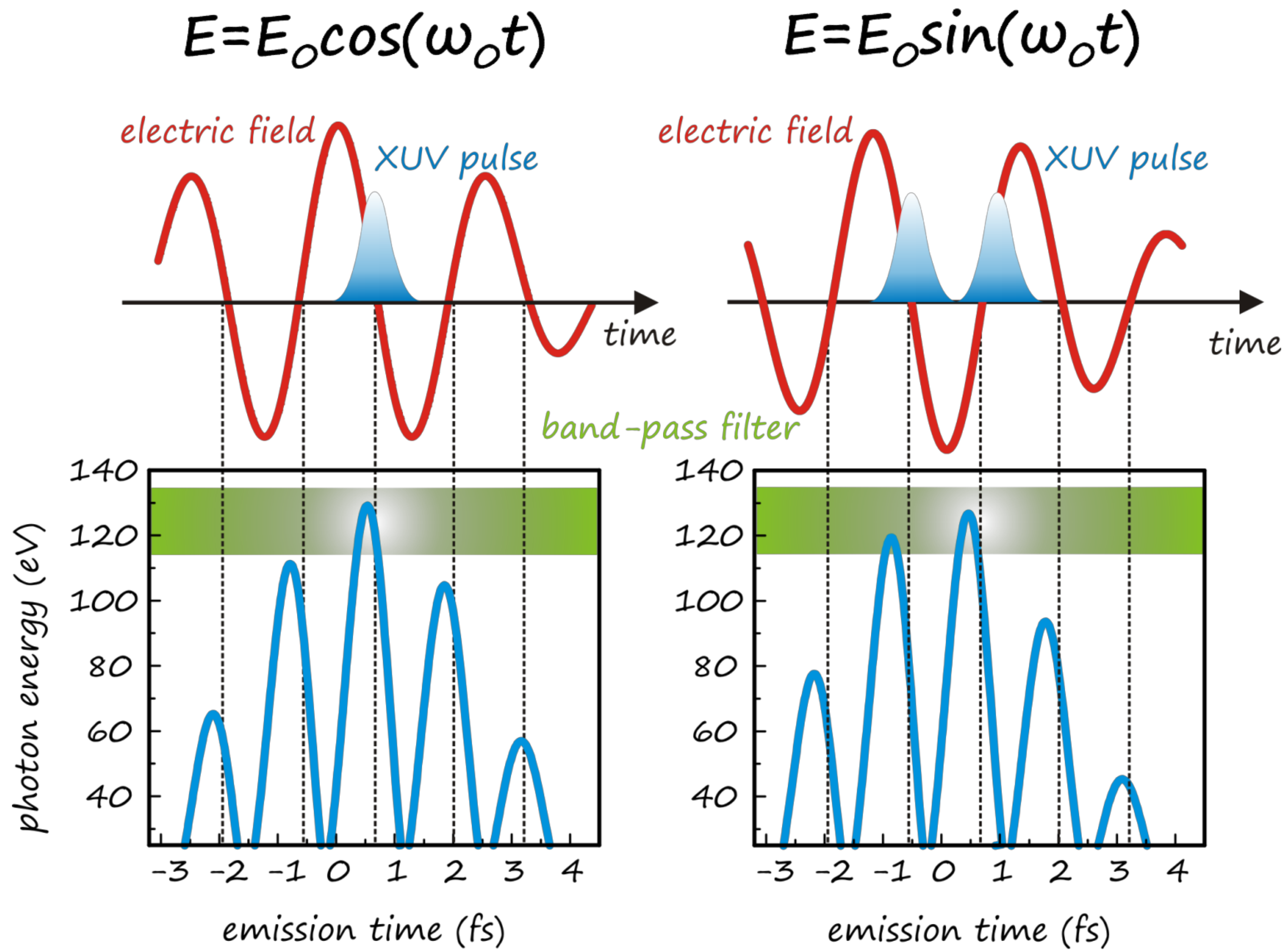
linear polarization

sub-5-fs pulses

CEP stability

I. Christov et al., Phys. Rev. Lett. 78, 1251 (1997)
A. Baltuska et al., Nature 421, 611 (2003)

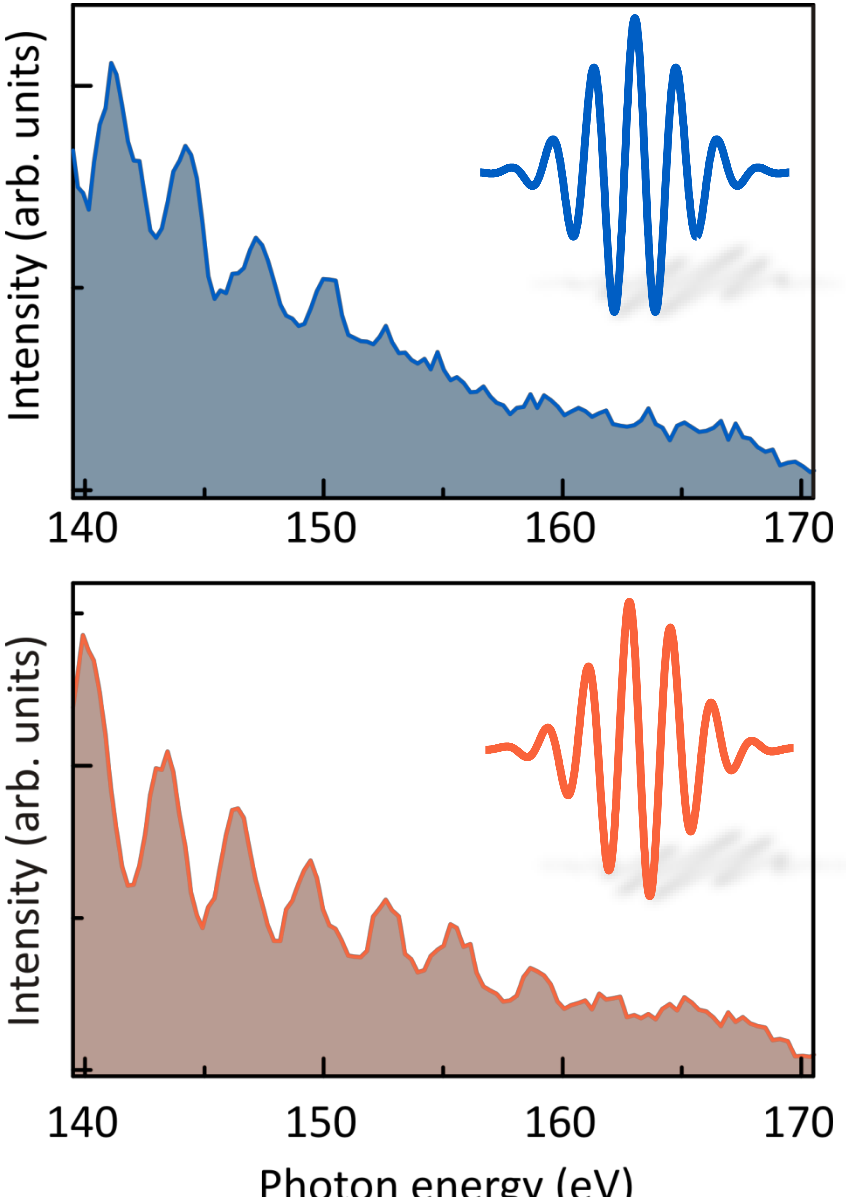
Spectral selection role of the CEP



spectral selection of cutoff photons leads to generation of one or two attosecond pulses!

Spectral selection

HHG in Neon



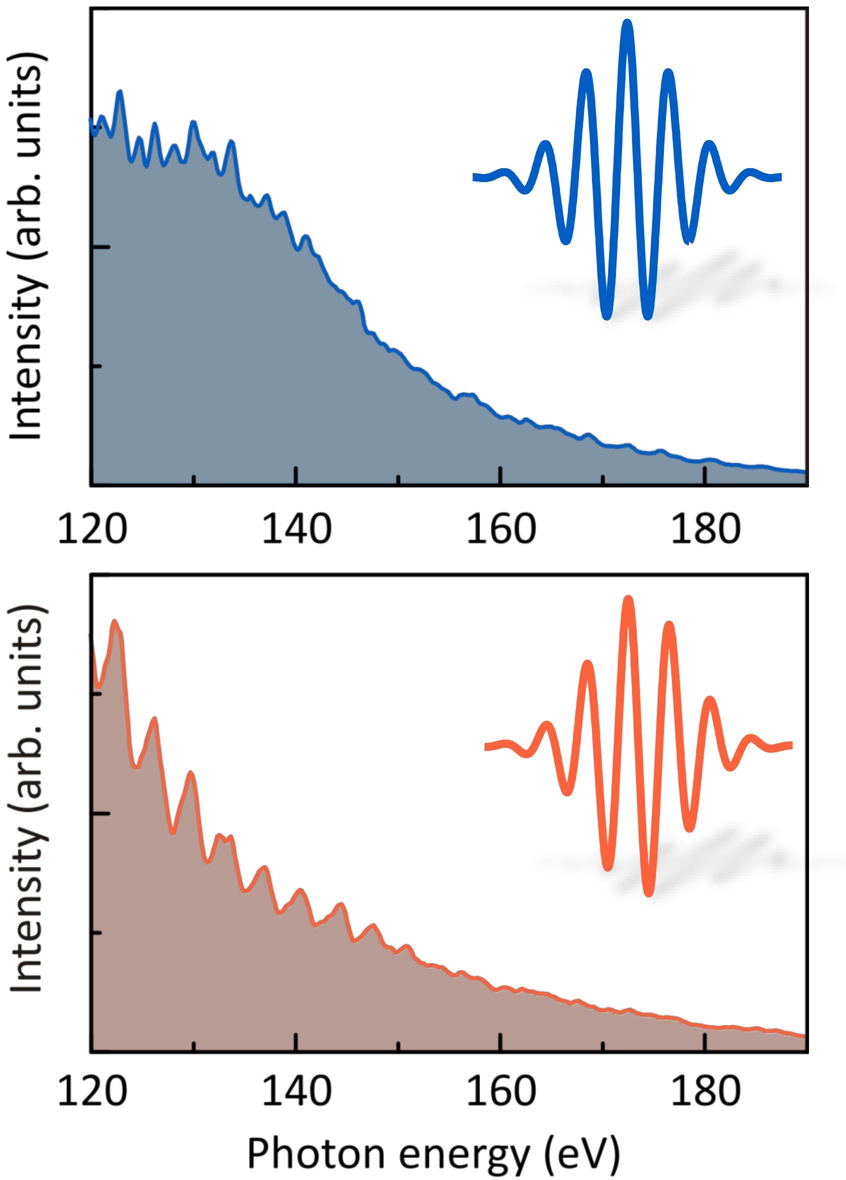
- pulse duration 5 fs
- stabilized CEP

broad continuum

only in the cut-off!

Spectral selection

HHG in Neon



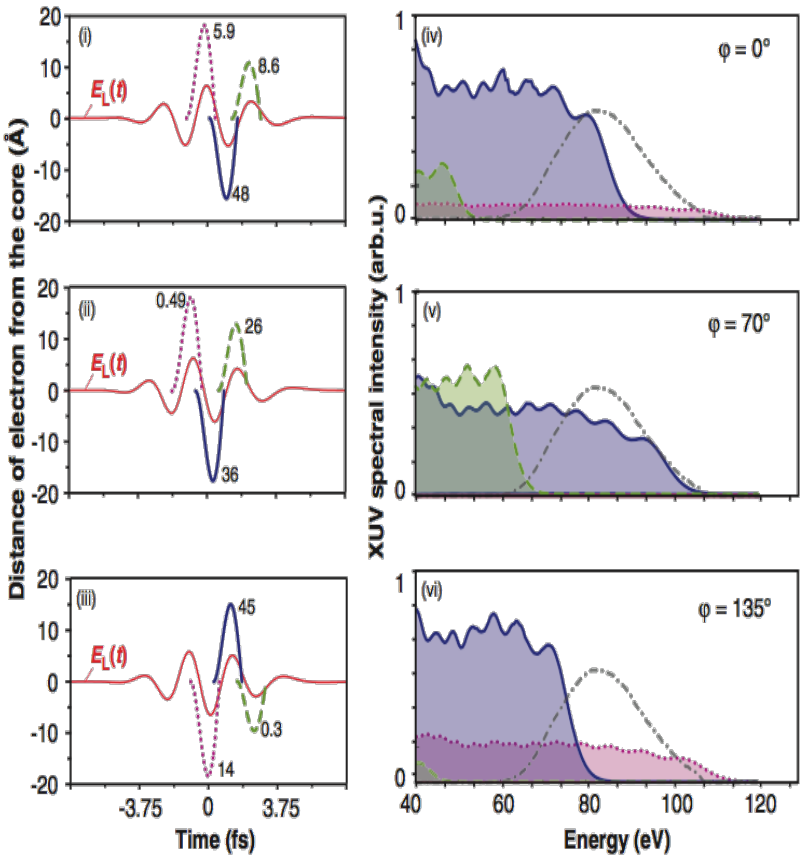
- pulse duration ~ 4 fs
- stabilized CEP

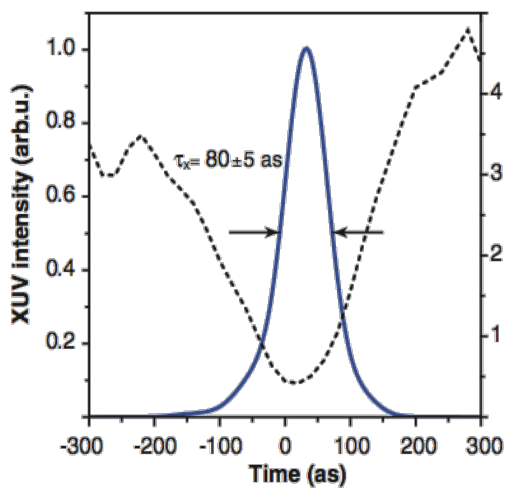
broad continuum ☺

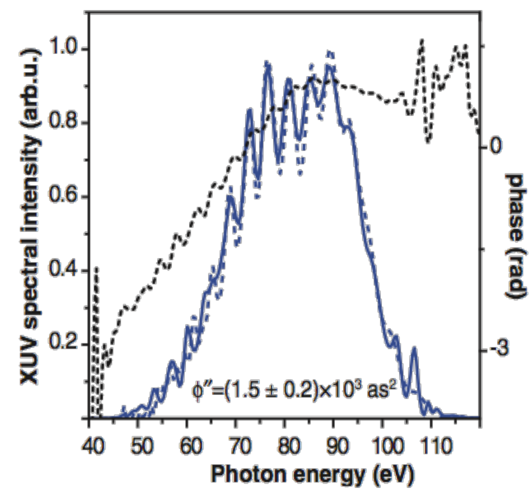
Spectral selection

state of the art

generation of a single 80 as pulse around 80 eV

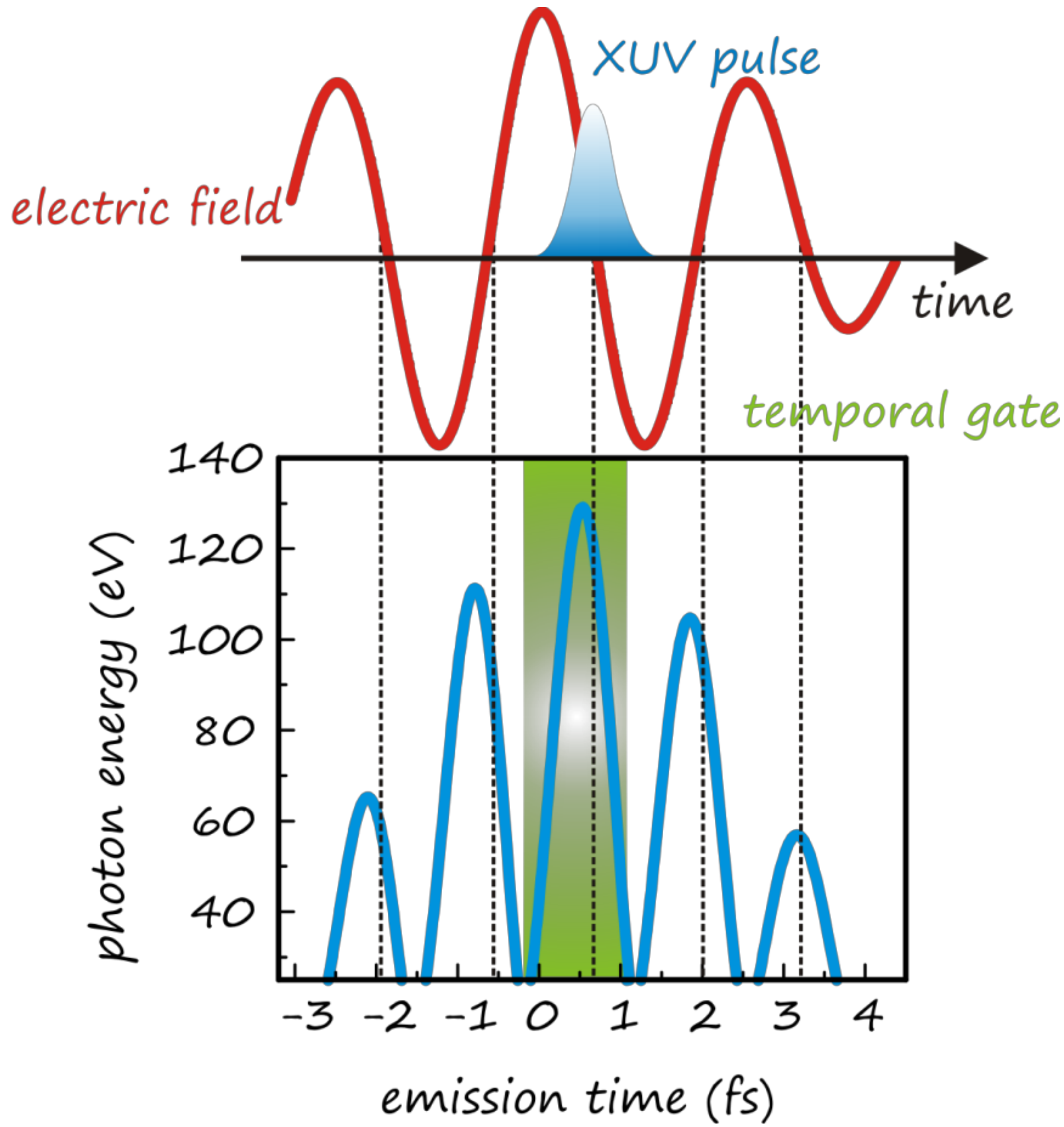






E. Goulielmakis et al., Science 320, 1614 (2008)

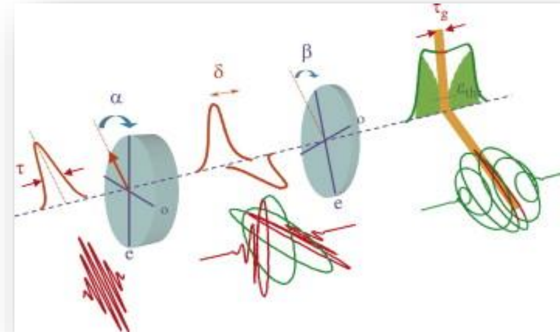
Temporal gate



Temporal gating schemes

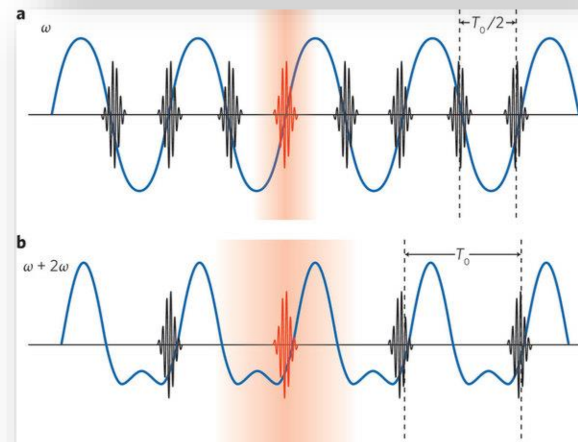
- **Polarization gating**

  one-color scheme
  (Generalized) Double Optical Gating



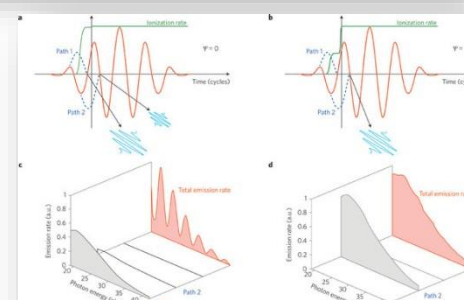
- **Two-color gating**

  intense IR pulses + intense visible few-cycle pulses



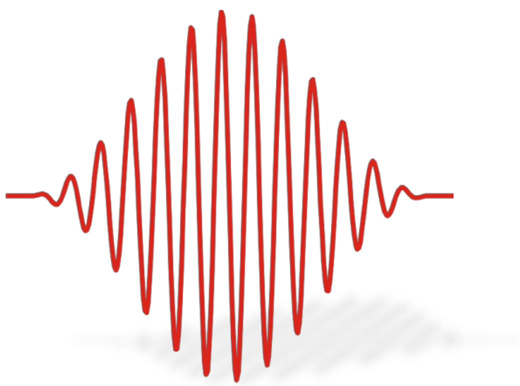
- **Ionization gating**

  few-cycle pulses with above saturation intensity and controlled electric field: high-energy isolated pulses on target

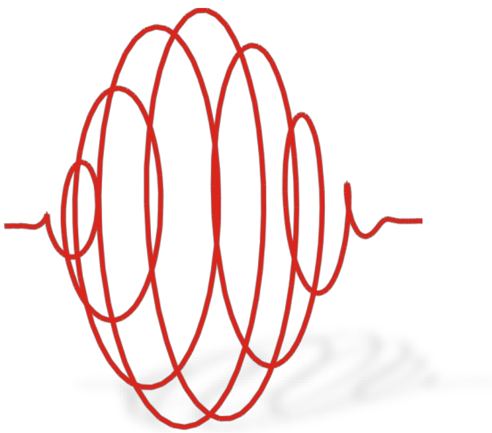


HHG polarization dependence

linear polarization



circular polarization

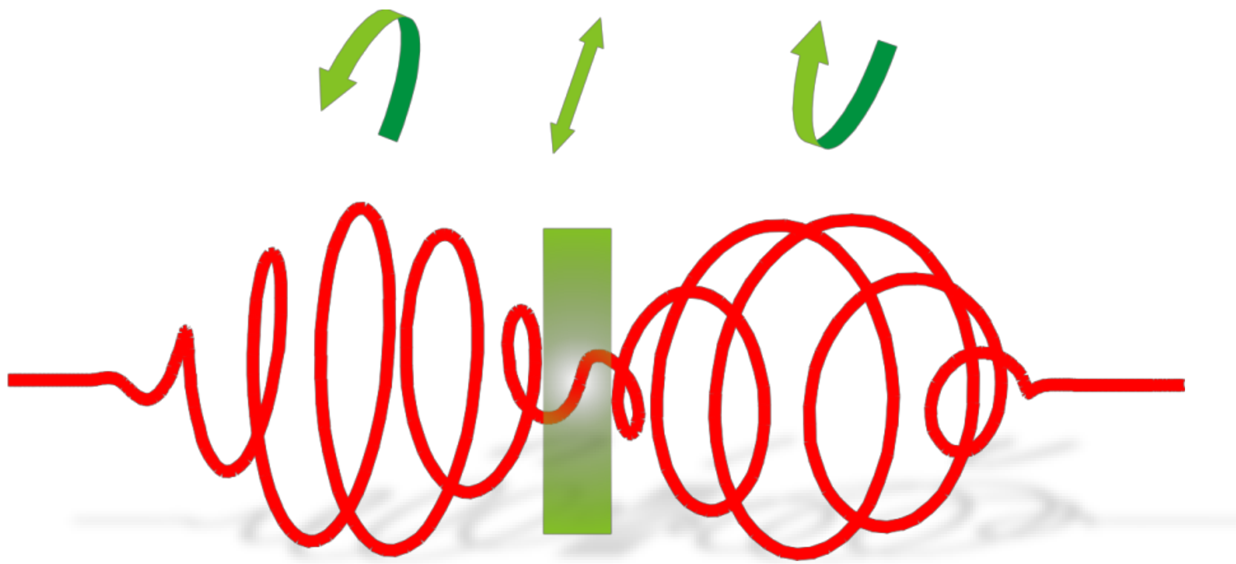


electron returns to the parent ion
HHG emission possible

electron doesn't return to the parent ion
HHG emission strongly reduced

Polarization gating

time-dependent polarization



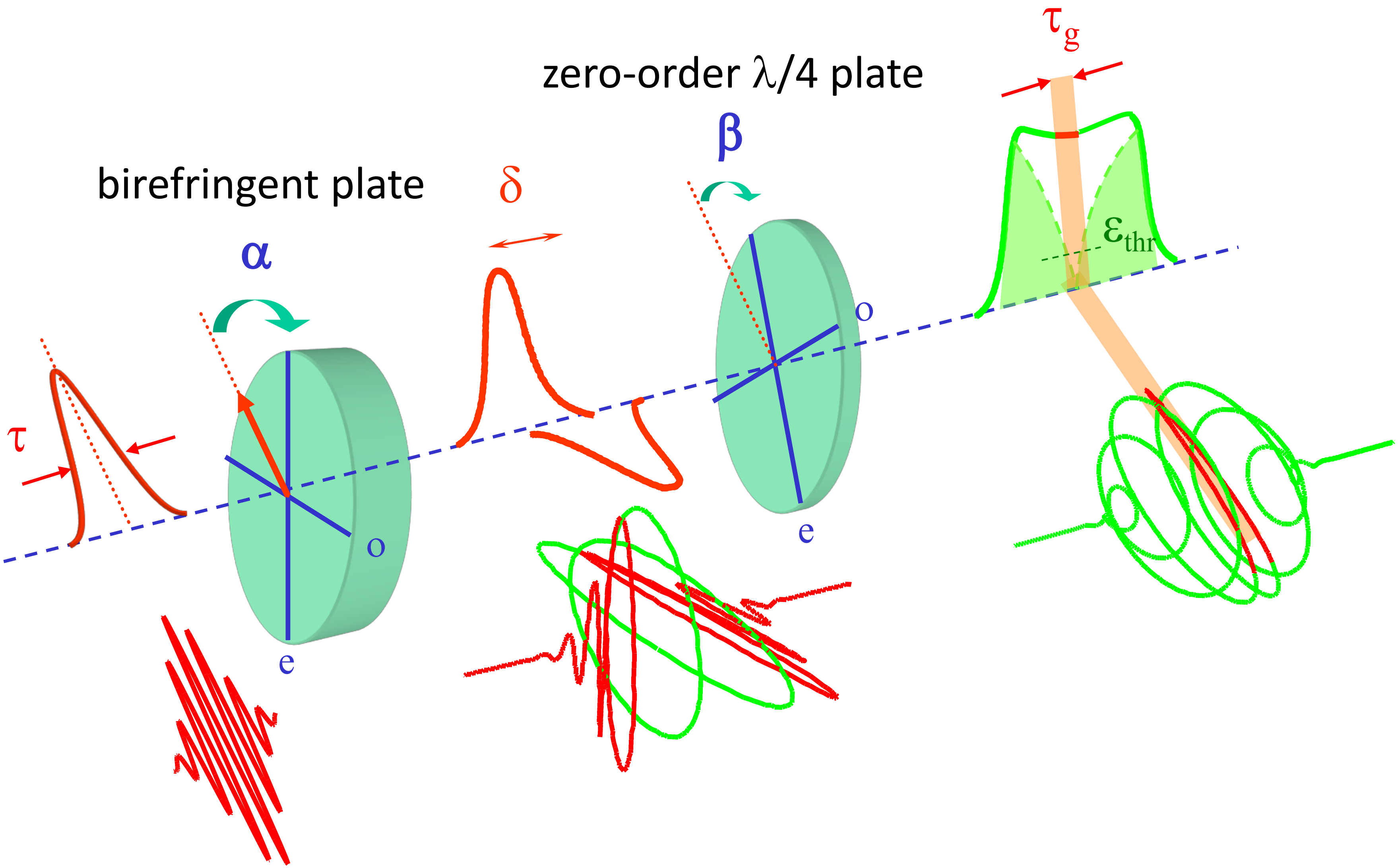
requirements:

few-cycle pulses

CEP stability

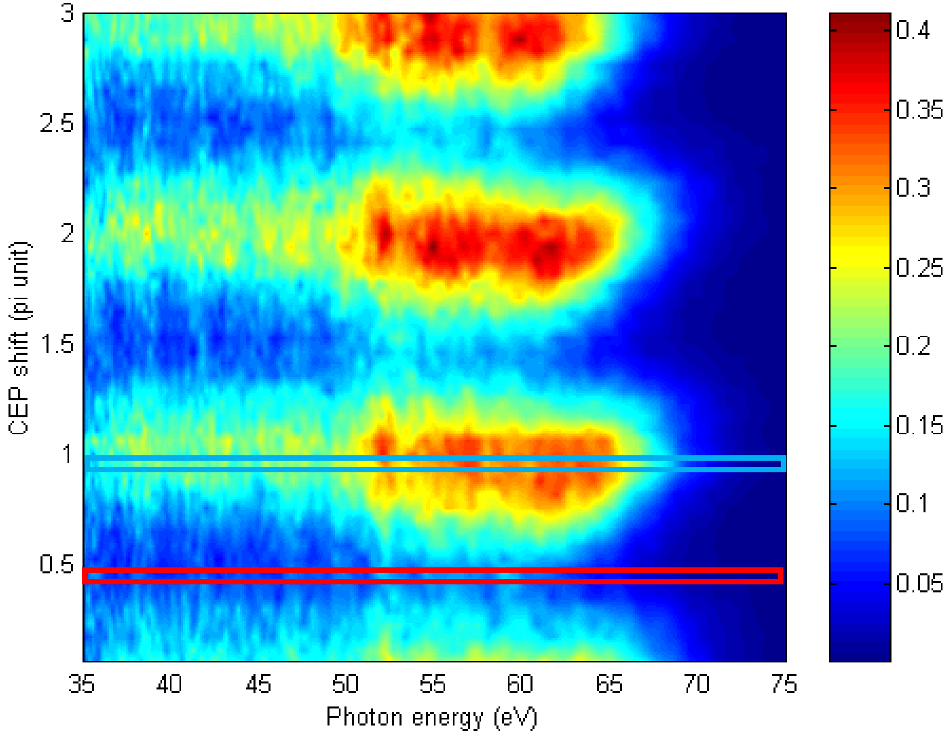
P. Corkum et al., Opt. Lett. 19, 1870 (1994)
O. Tcherbakoff et al., Phys. Rev. A 68, 043804 (2003)

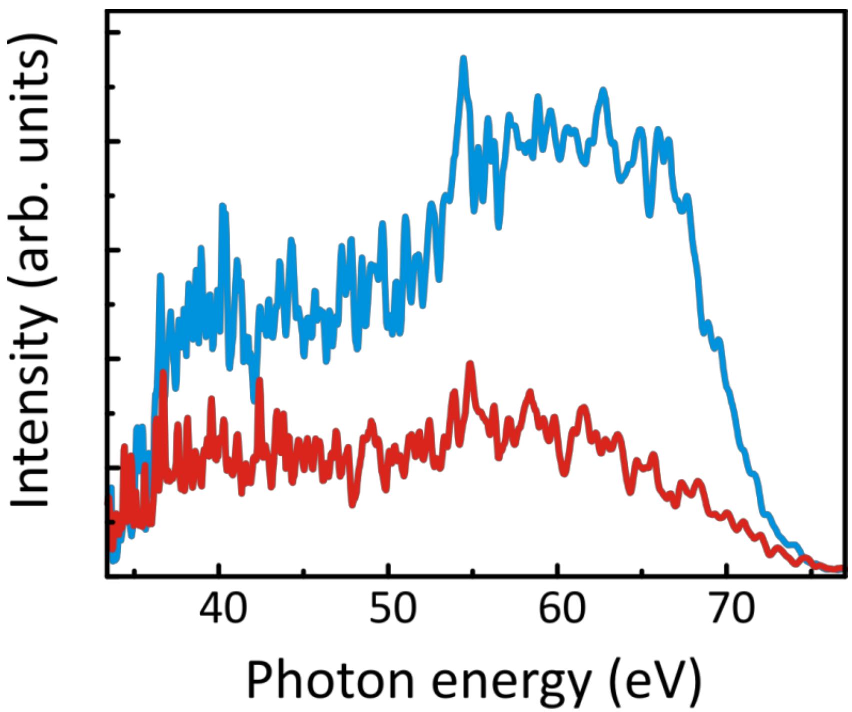
Polarization gating



Polarization gating results in Ne

pulse duration 5 fs, delay δ= 6.2 fs, $\phi_0 < \phi < \phi_0+3\pi$

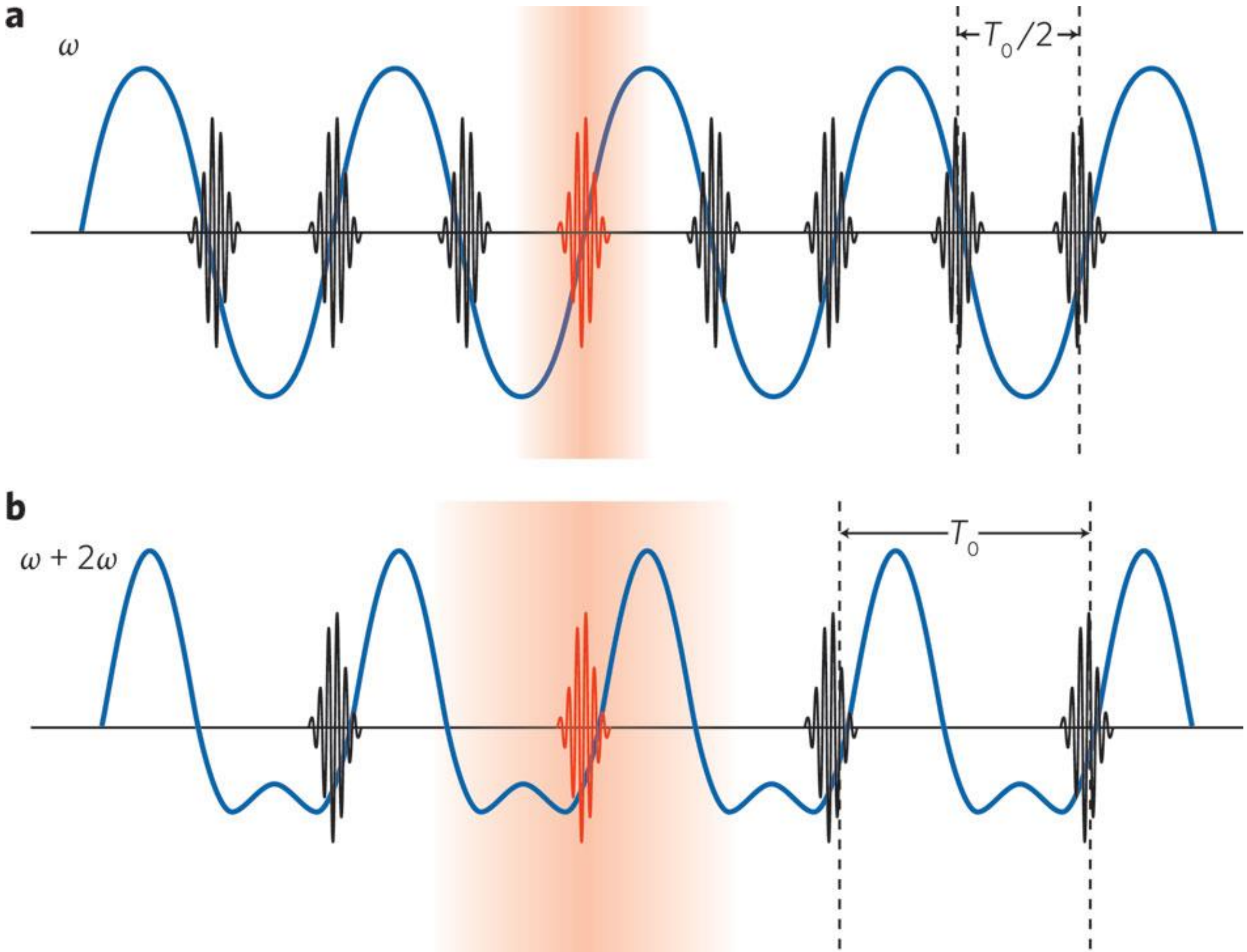




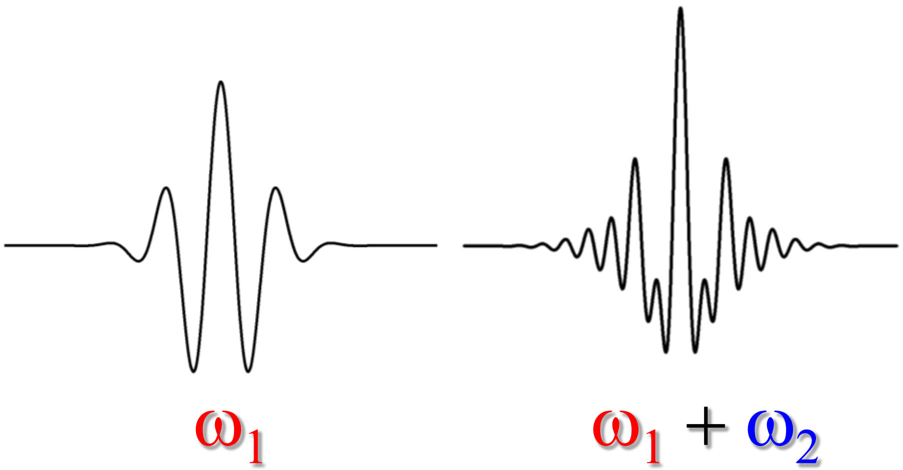
- strong periodic modulation of emission efficiency for $\Delta\phi = \pi$
- continuous spectra from 30 eV to 75 eV for all CEPs

I. Sola et al., Nature Physics 2, 319 (2006)

Temporal gating with two colors



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!

<u>key parameters</u>:

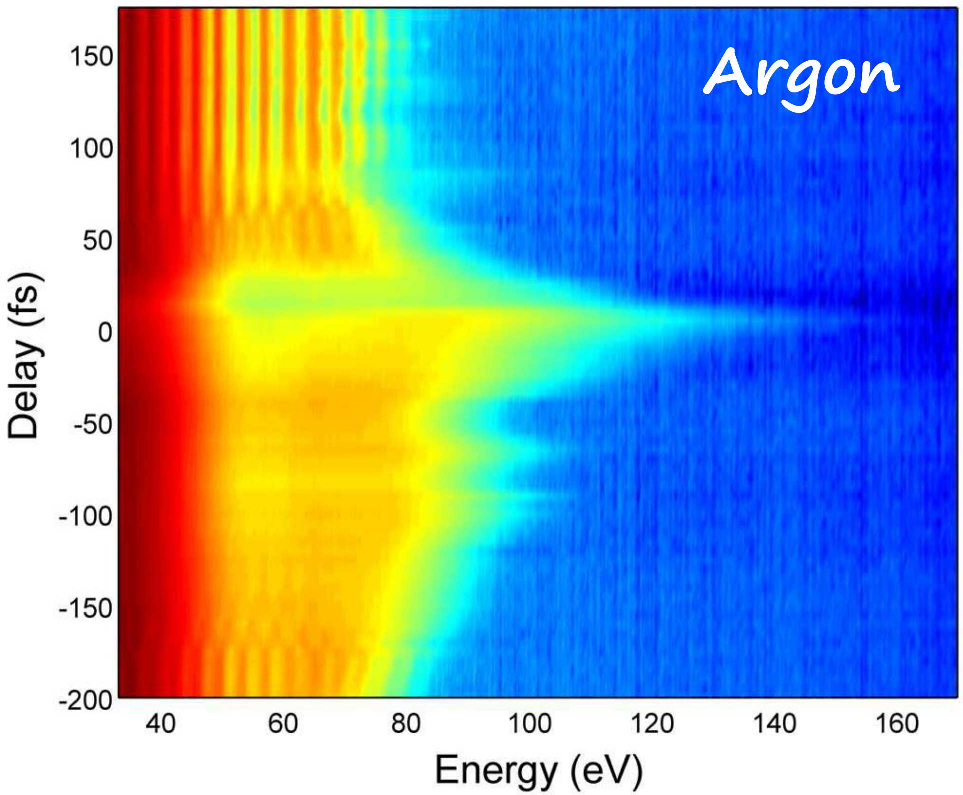
- central wavelength of the two components
- intensity of the pulses
- temporal overlap
- gas target position

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

Two-color gating

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

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



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

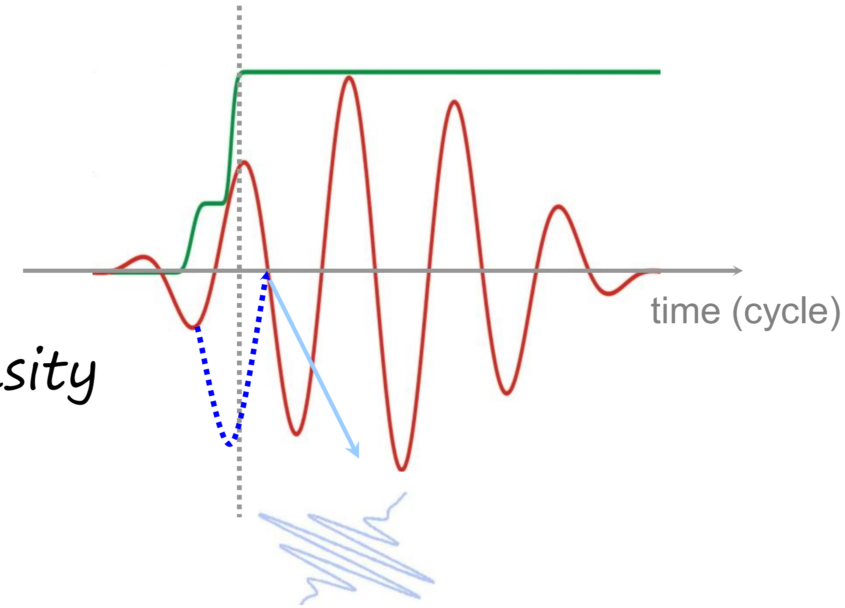
F. Calegari et al., Opt. Lett. 34, 3125 (2009)

Ionization gating

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

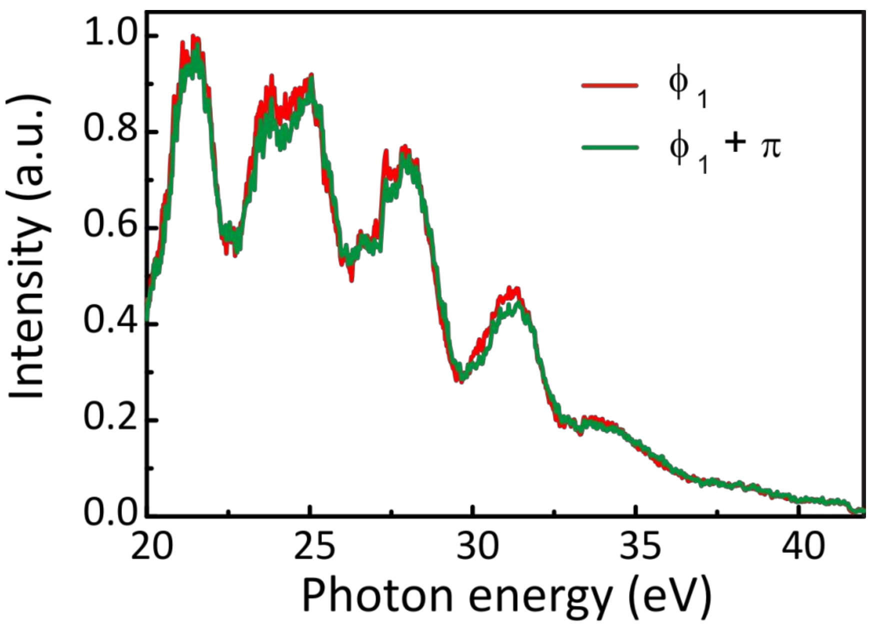


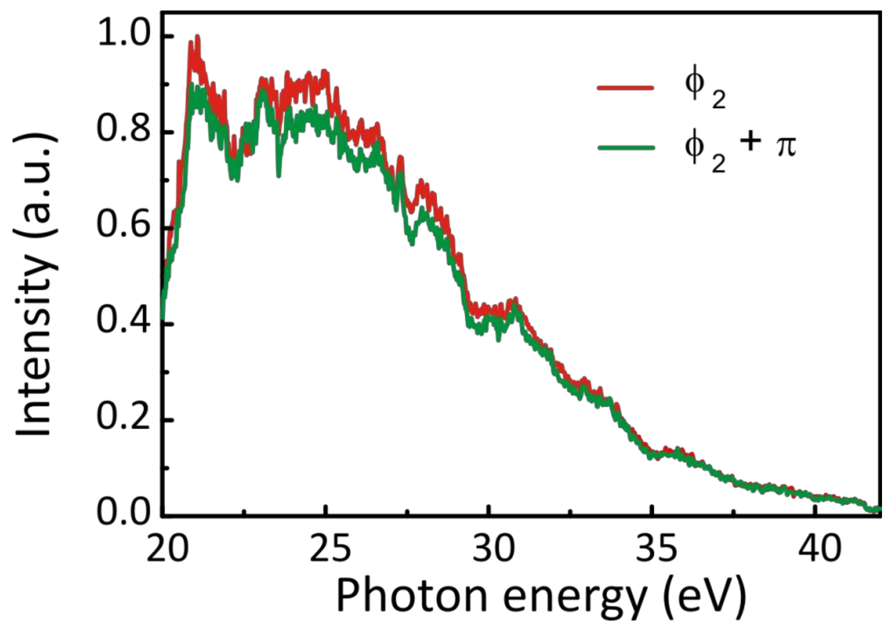
<u>requirements</u>:

- 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 5 fs, peak intensity 2.3 10^{15} W/cm^2
2.5-mm Xe cell



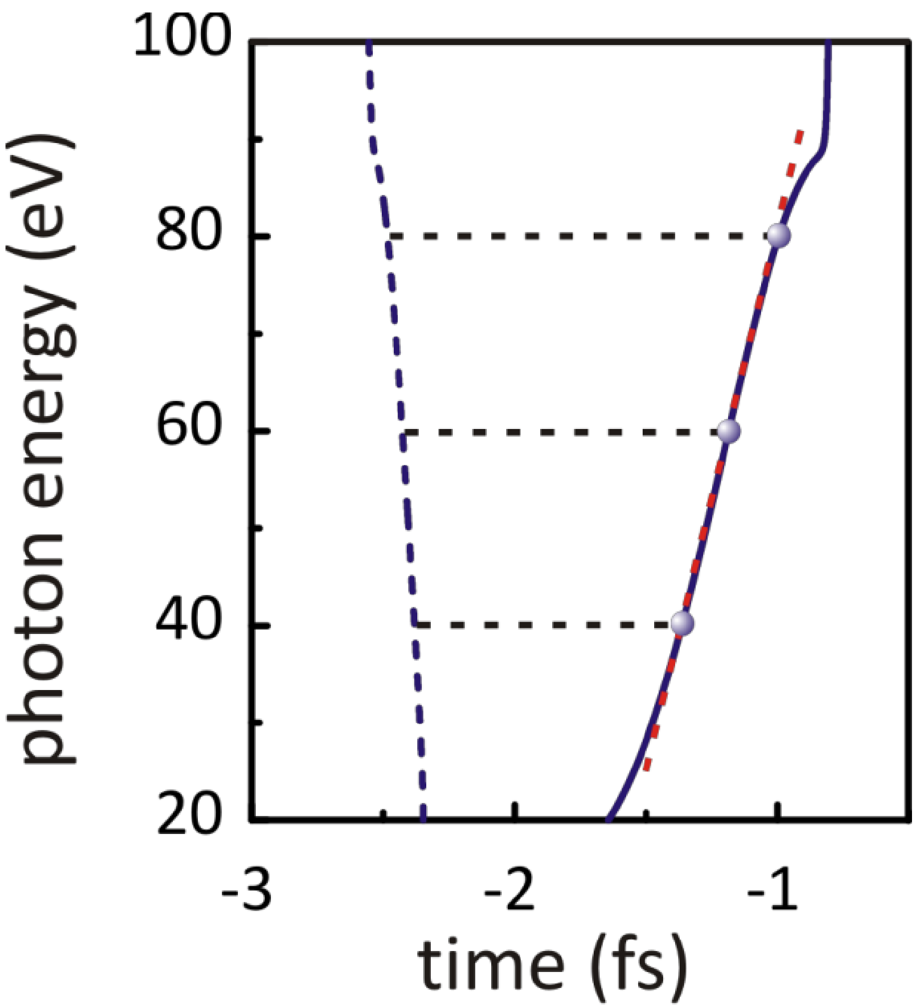


- periodic change of amplitude and shape for $\Delta\phi = \pi$
- CEP drives transition from double to single emission
- measured pulse energy on target ~ 2.1 nJ

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

Atto-chirp

selection of short trajectories

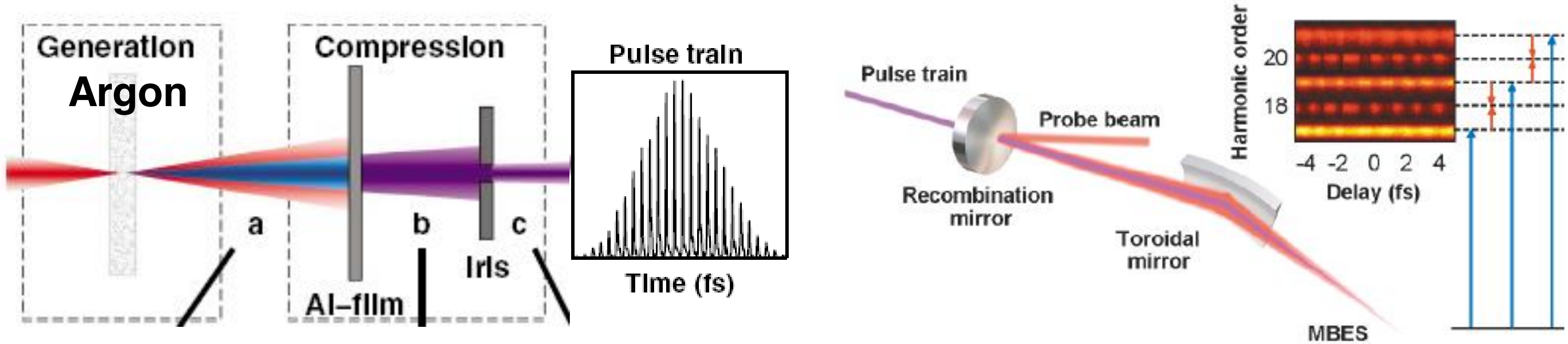


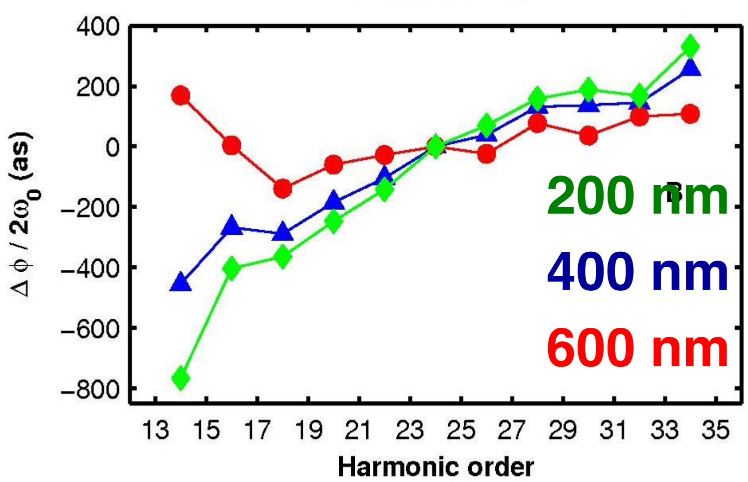
electron recombination time depends on the energy!

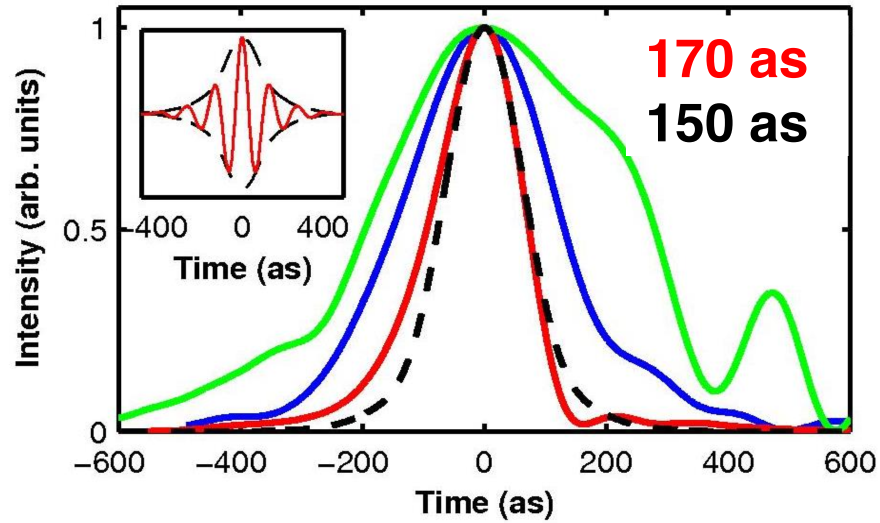
positive chirp of harmonic emission on the attosecond time-scale

and the atto-chirp??

metallic filter provides negative GDD in the XUV region!

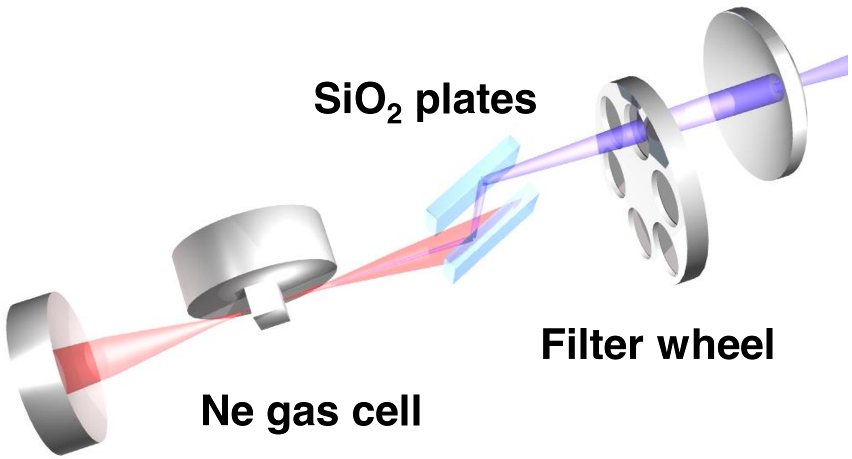


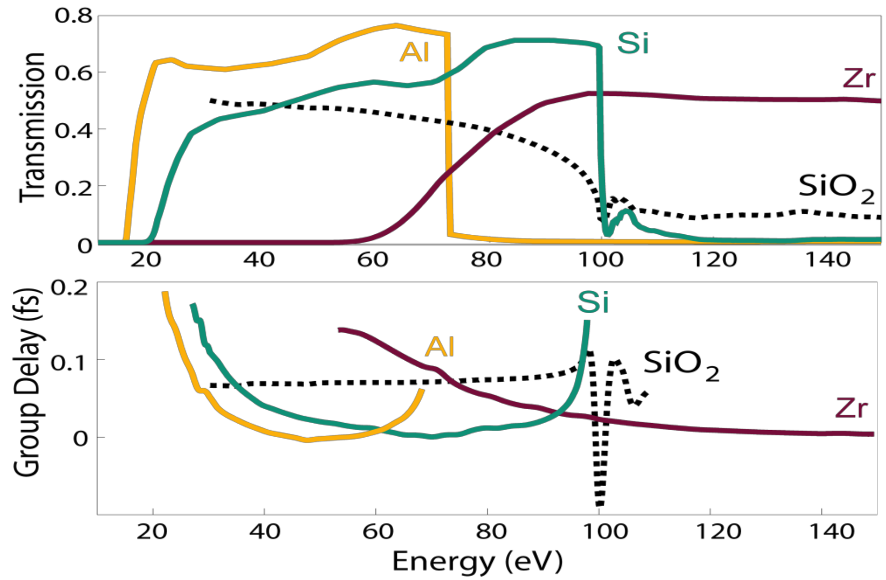


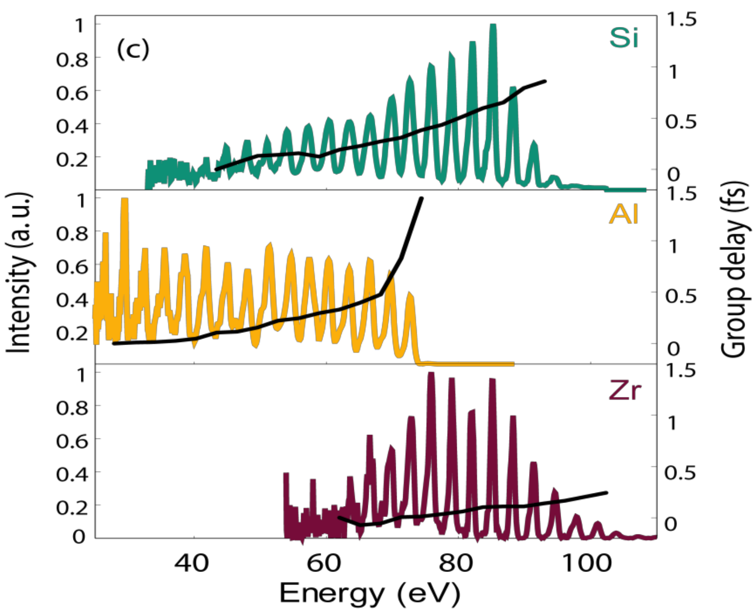


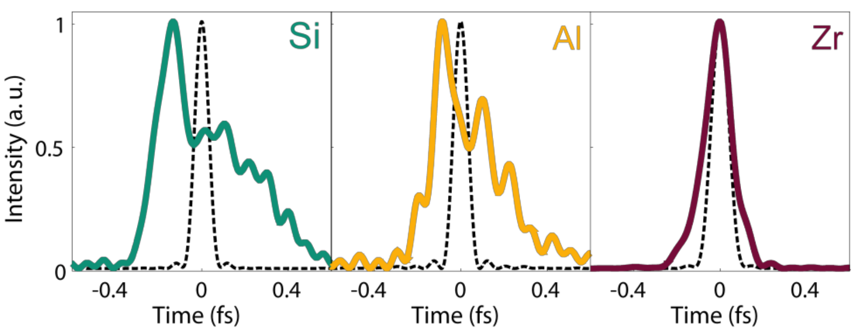
López-Martens et al. Phys. Rev. Lett., 94, 033001 (2005)

Metallic filters



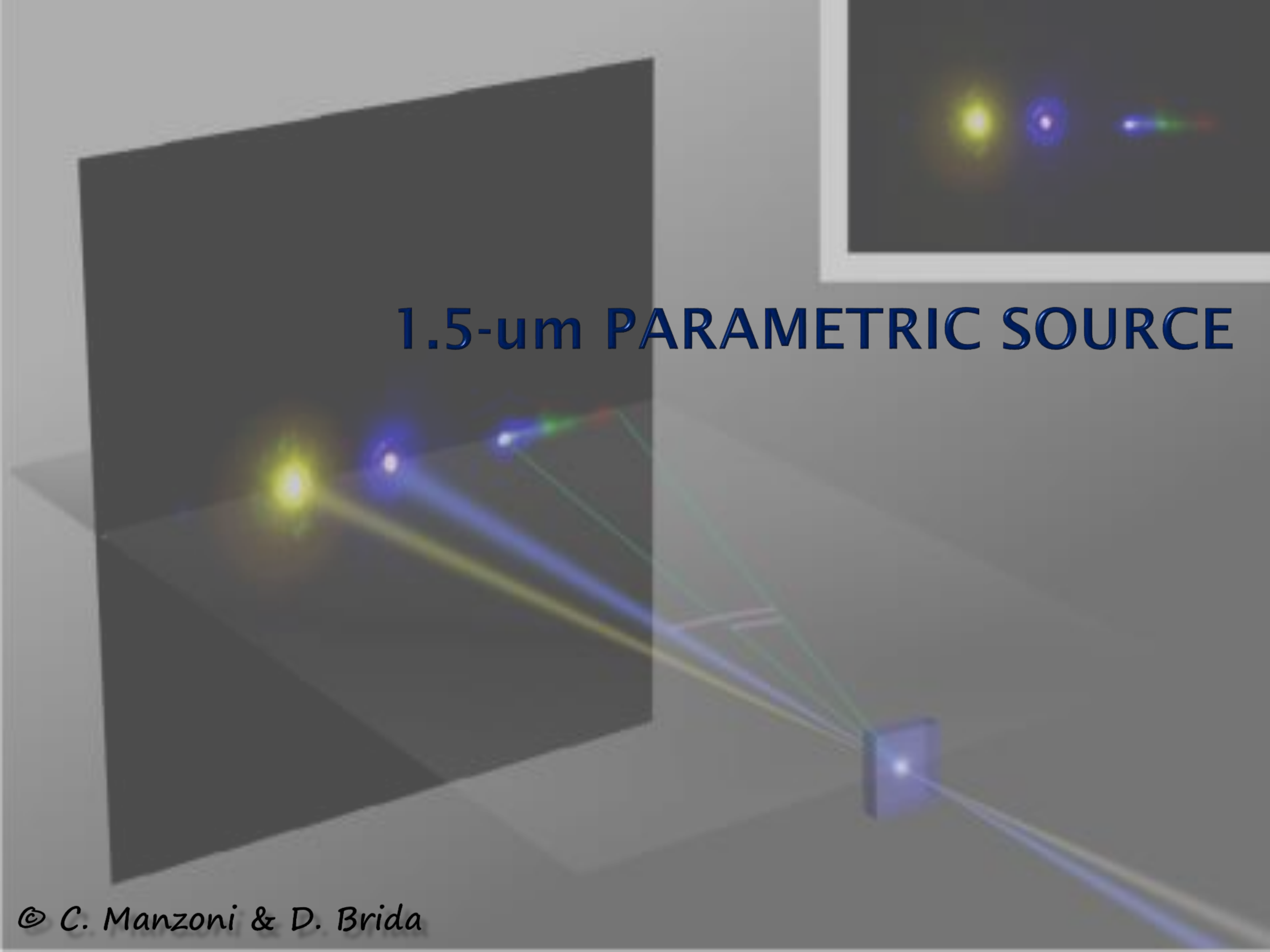






Gustafsson et al Opt. Let. 32, 1353 (2007)

1.5-um PARAMETRIC SOURCE



Looking for…

THE PERFECT DRIVING PULSE

1. Cut off extension towards the soft X ray region
   Ponderomotive electron energy $U_p \sim \lambda^2$

2. CEP stability for single attosecond pulse generation
   Attochirp scales as $\sim \lambda^{-1}$

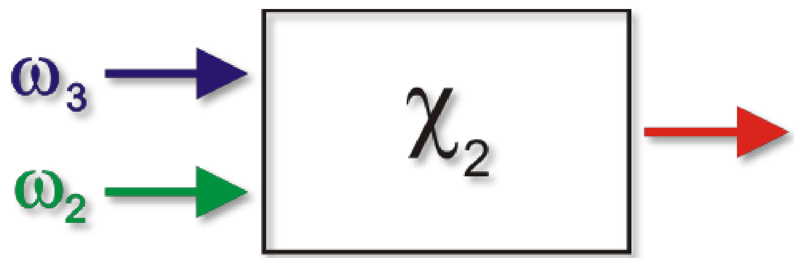
3. Tunability of discrete harmonics

☺ **DFG is the solution !!**

but… ☹ Generation yield $\sim\lambda^{-\alpha}$ with $\alpha\sim 5-6$

PASSIVE STABILIZATION OF CARRIER ENVELOPE PHASE

Difference Frequency Generation (DFG):



$$\omega_1 = \omega_3 - \omega_2$$

$$\phi_1 = \pi/2 + \phi_3 - \phi_2$$

DFG between two pulses carrying the same CEP leads to automatic phase-stabilization of the DF pulse

$$\phi_3 = \phi + c$$

$$\phi_2 = \phi + c'$$

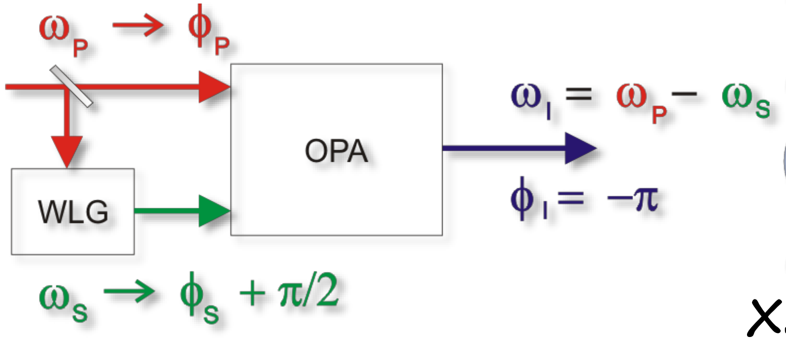
$$\longrightarrow \quad \phi_1 = cost$$

Baltuška et al., Phys. Rev. Lett. 88, 133901 (2002)

POSSIBLE IMPLEMENTATION

DFG in OPAs

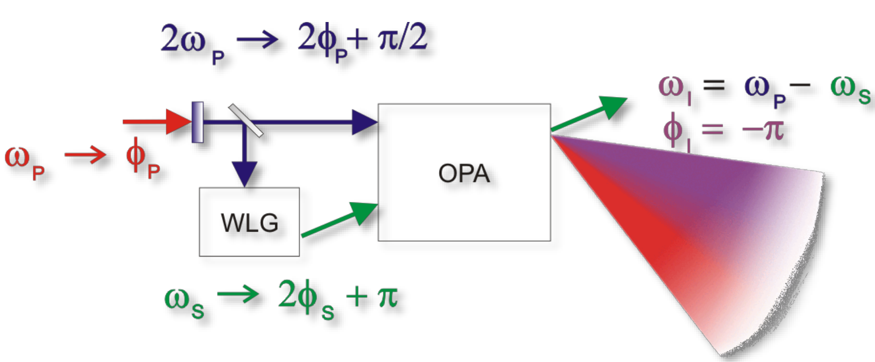
IR-pumped collinear OPA



- ☺ Idler self phase stabilization
- ☹ Narrow phase matching bandwidth

X. Fang et al., Opt. Lett. 29, 1282 (2004)

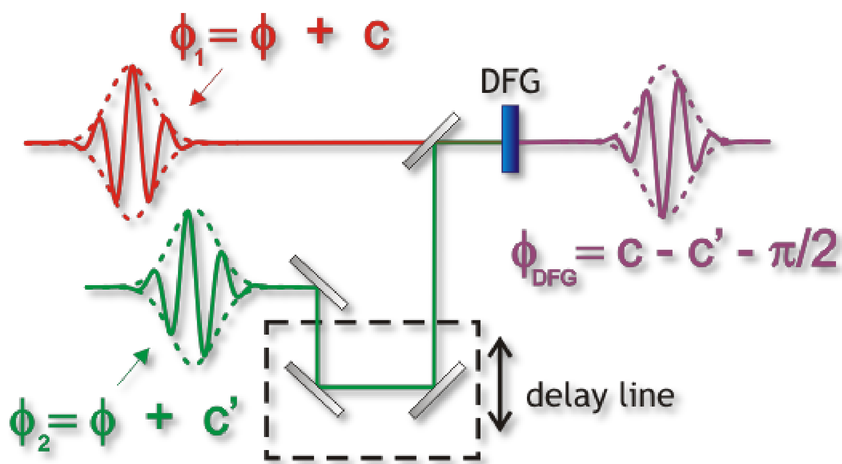
SH-pumped SH-seeded Non-collinear OPA



- ☺ Idler self phase stabilization
- ☺ Broad phase matching bandwidth
- ☹ Idler angular dispersion

S. Adachi et al., Opt. Lett. 29, 1150 (2004)

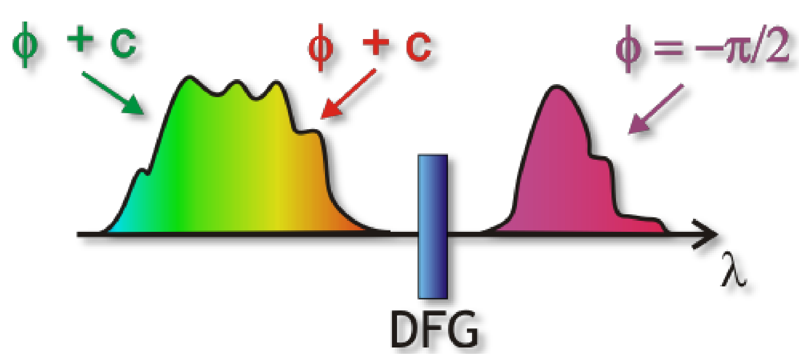
INTERPULSE AND INTRAPULSE DFG



DFG between two frequency shifted pulses

☹ Delay-induced CEP jitter

C. Manzoni et al., Opt. Lett. 29, 2668(2004)



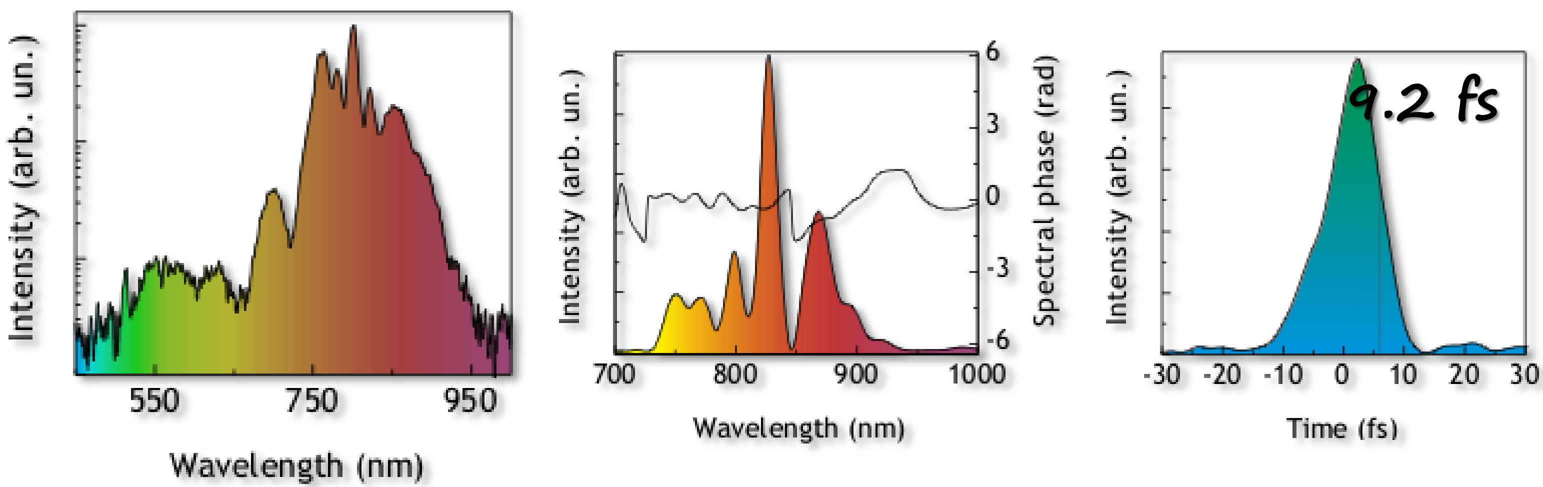
DFG between short and long wavelength components of a broadband pulse

☺ No necessity to maintain time delay between mixing pulses

T. Fuji et al., Opt. Lett. 30, 332 (2005)

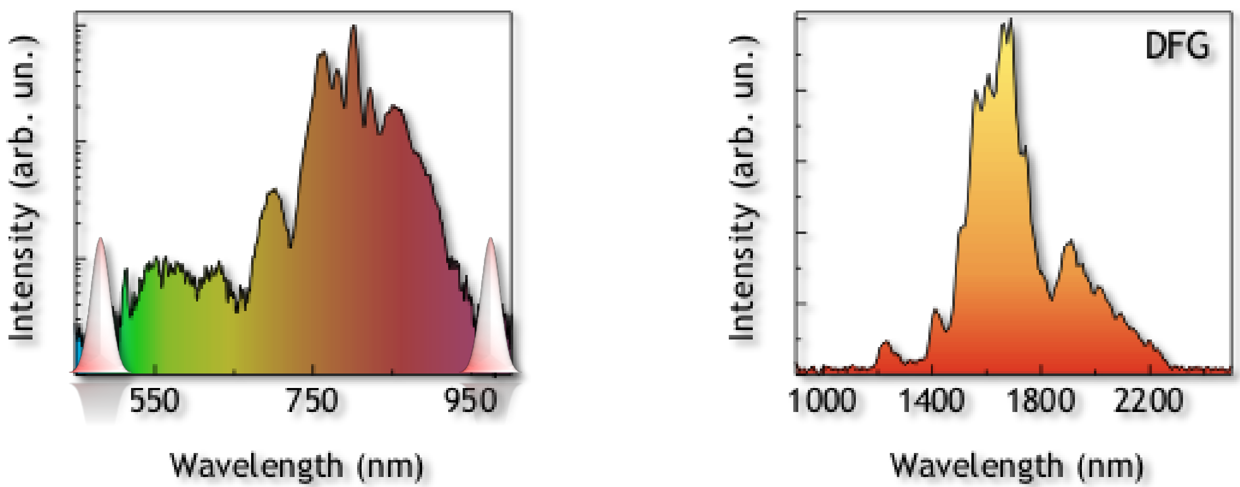
OUR PROPOSAL

1 Compression of the initial pulse by filamentation in Kr
→ high energy supercontinuum
→ pulse compression down to 10 fs



OUR PROPOSAL

1. Compression of the initial pulse by filamentation in Kr
   → high energy supercontinuum
   → pulse compression down to 10 fs

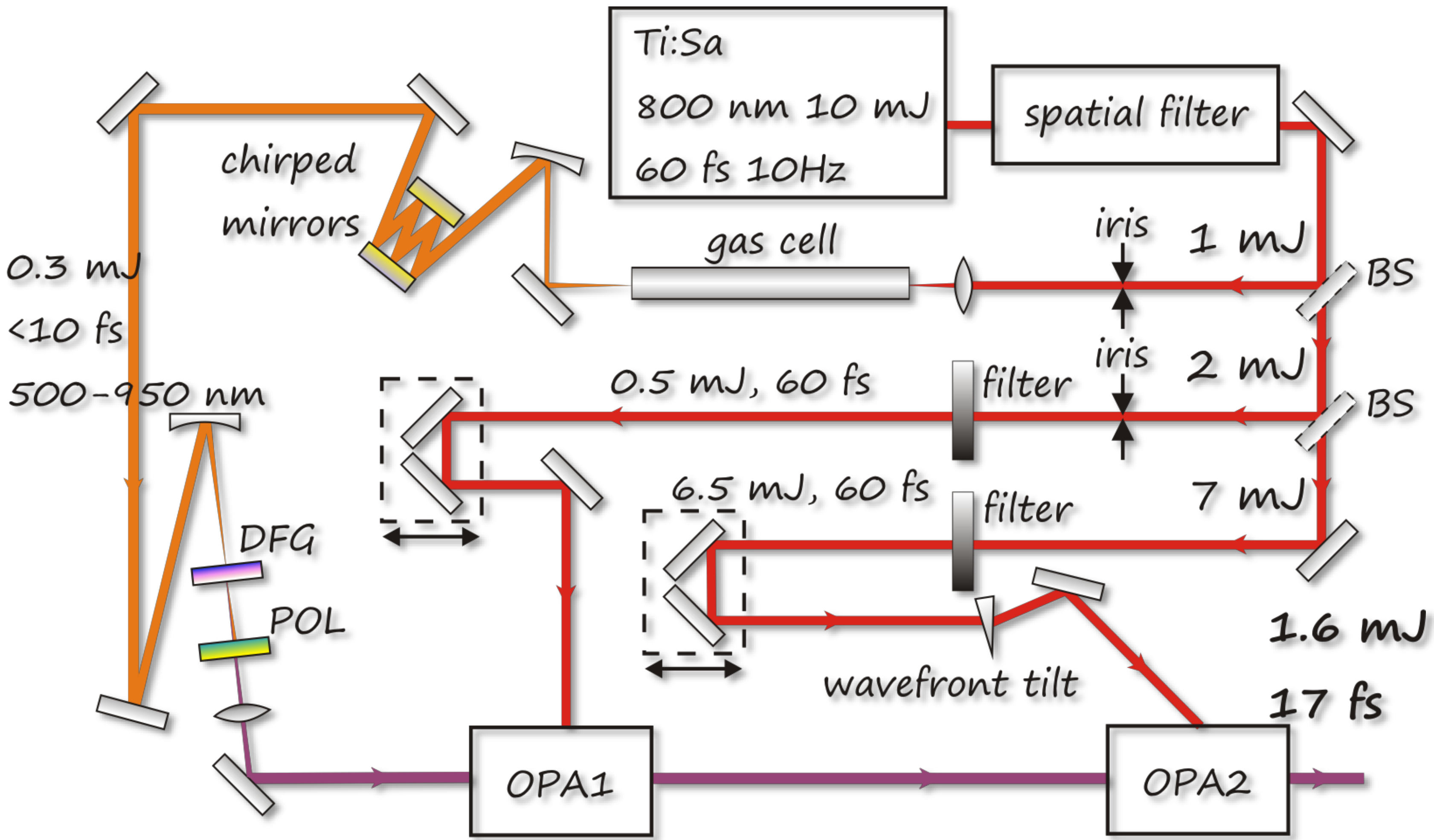


2. Difference frequency generation of the supercontinuum
   → carrier wavelength in the near IR
   → passive stabilization of the CEP

3. Amplification by Near-IR OPA at degeneracy
   → high energy, broad gain bandwidth

PARAMETRIC SOURCE

experimental setup

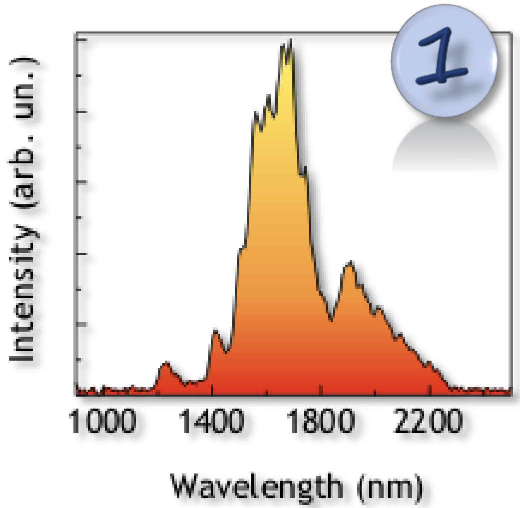


Opt. Lett. 32, 2957 (2007)

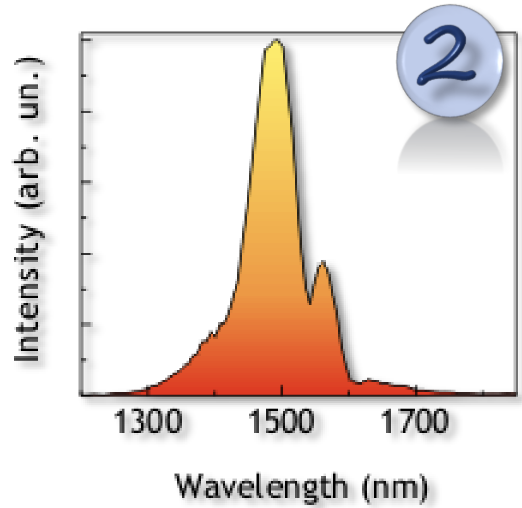
PARAMETRIC SOURCE

spectral characterization

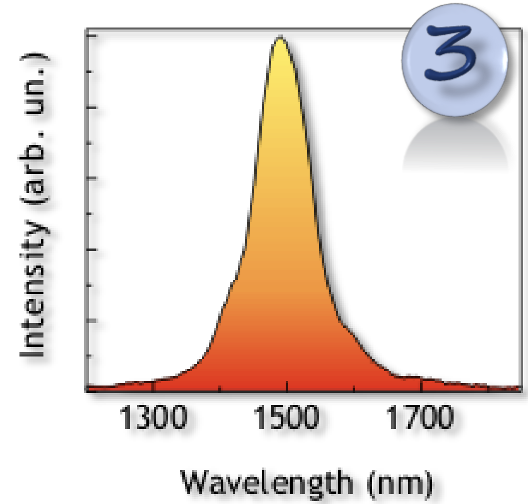
DFG
BBO type II 200 μm
transform limit 10 fs



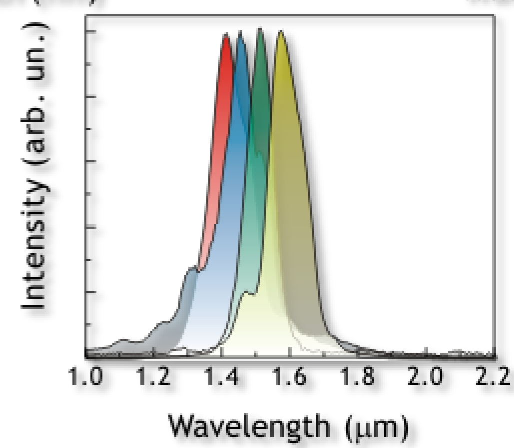
OPA1
BBO type II 2 mm
transform limit 15 fs



OPA2
BBO type II 3 mm
transform limit 17 fs



Tilt of the BBO crystals allows carrier wavelength tunability

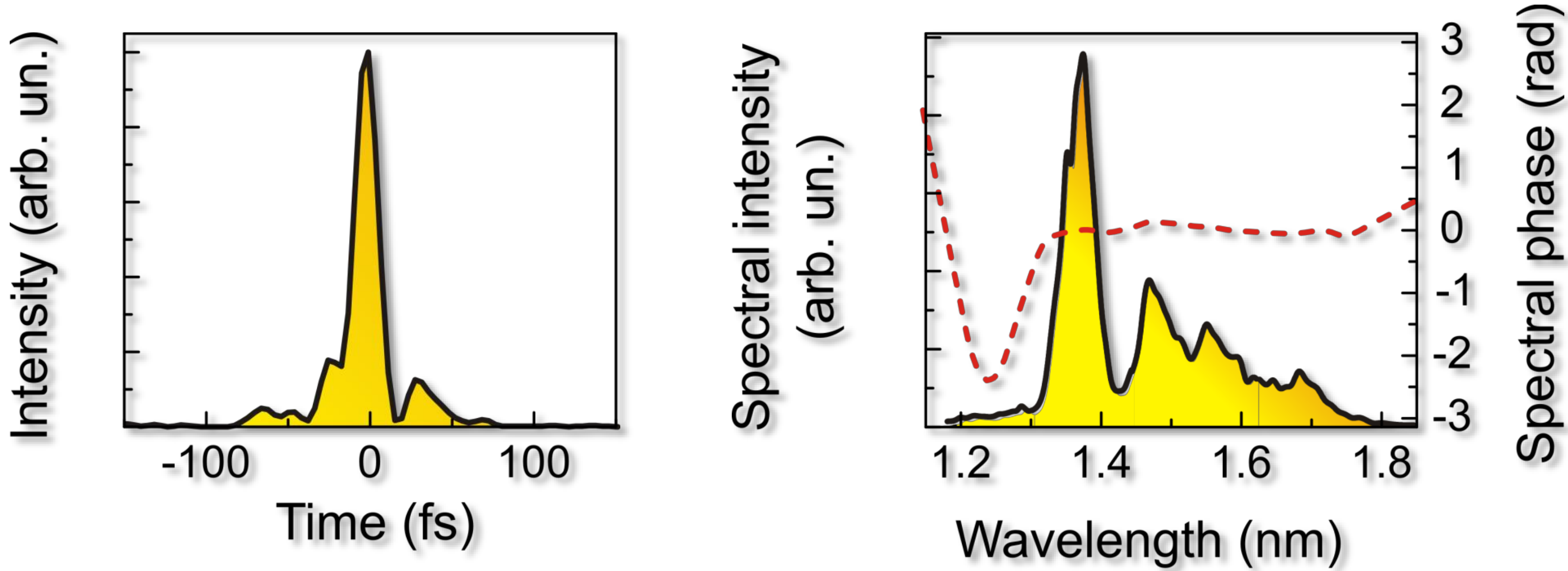


PARAMETRIC SOURCE

temporal characterization

Zero Additional Phase SPIDER

nearly transform-limited 17-fs pulse width

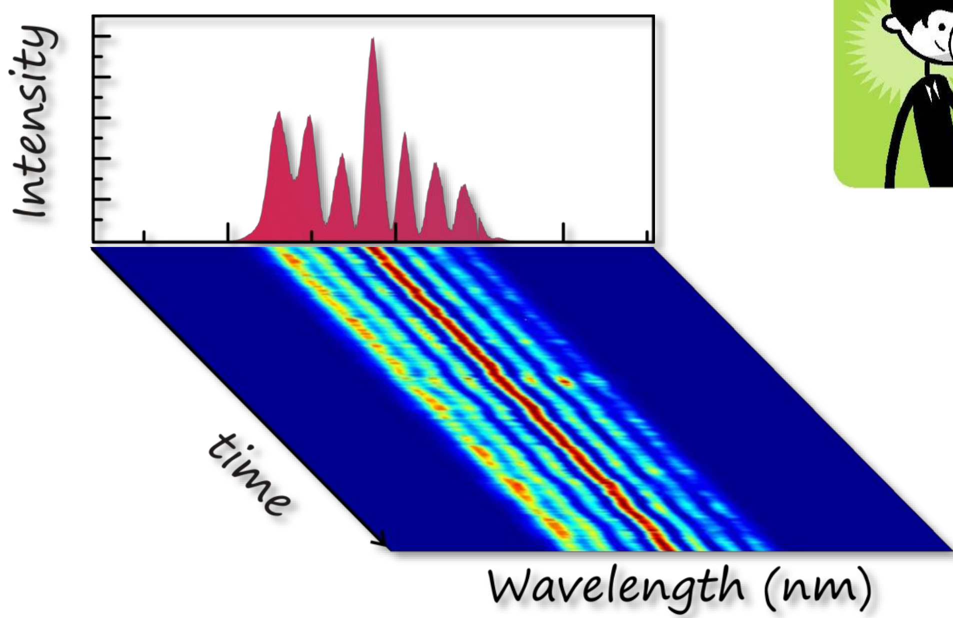


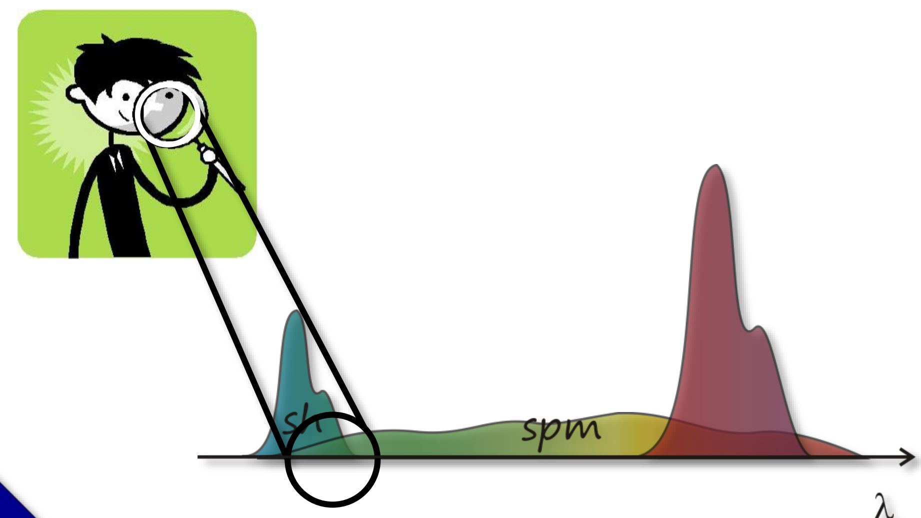
3 to 4 optical cycle pulses @ 1.5 μm

CARRIER-ENVELOPE PHASE STABILITY

how to measure it

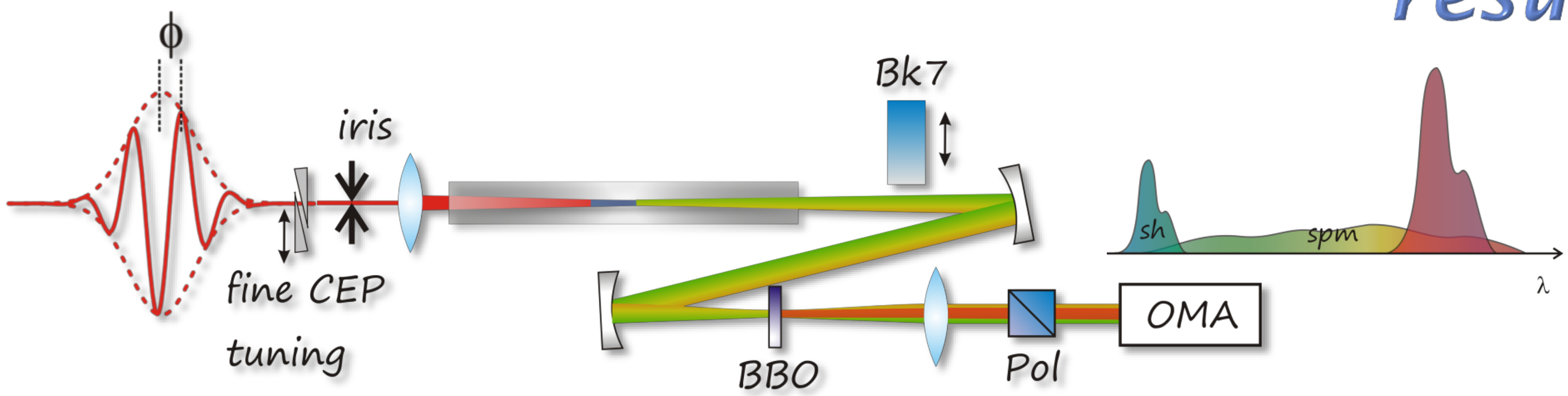
1. Generation of an octave spanning spectrum
2. Frequency doubling in a non-linear crystal
3. Observation of spectral interference between the two components



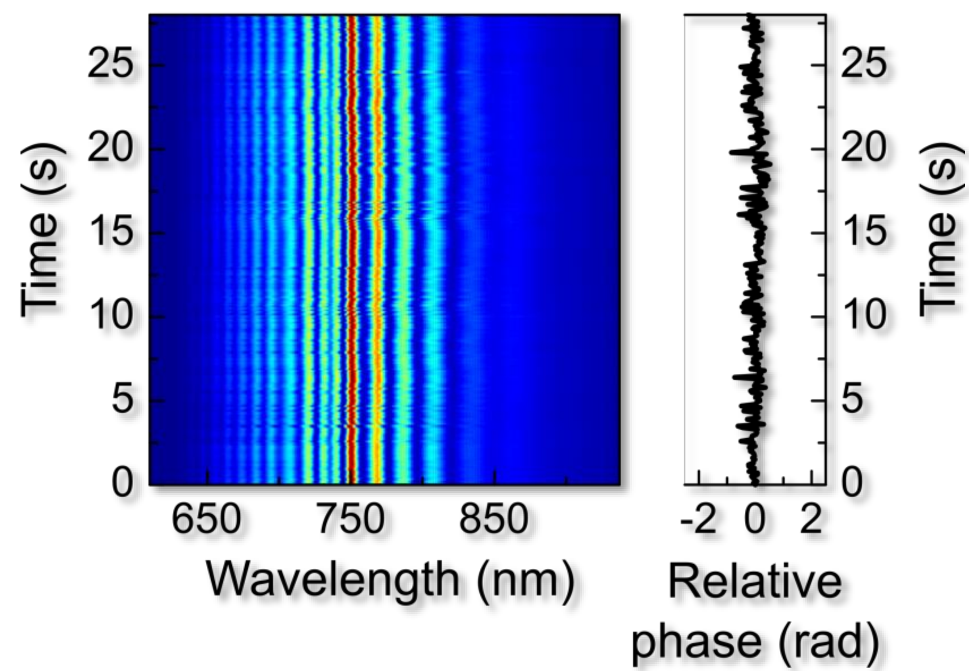


CARRIER-ENVELOPE PHASE STABILITY

results



Spectral broadening by filamentation in krypton

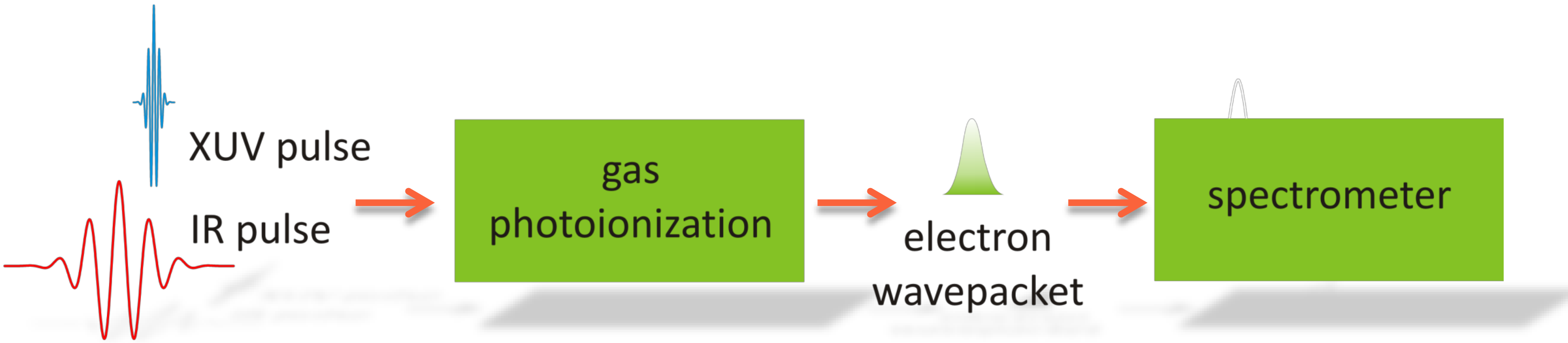


- minimizes intensity-phase coupling
- 0.19 rad RMS

J. Opt. Soc. Am. B 25, B112 (2008)

II.
Attosecond pulse characterization

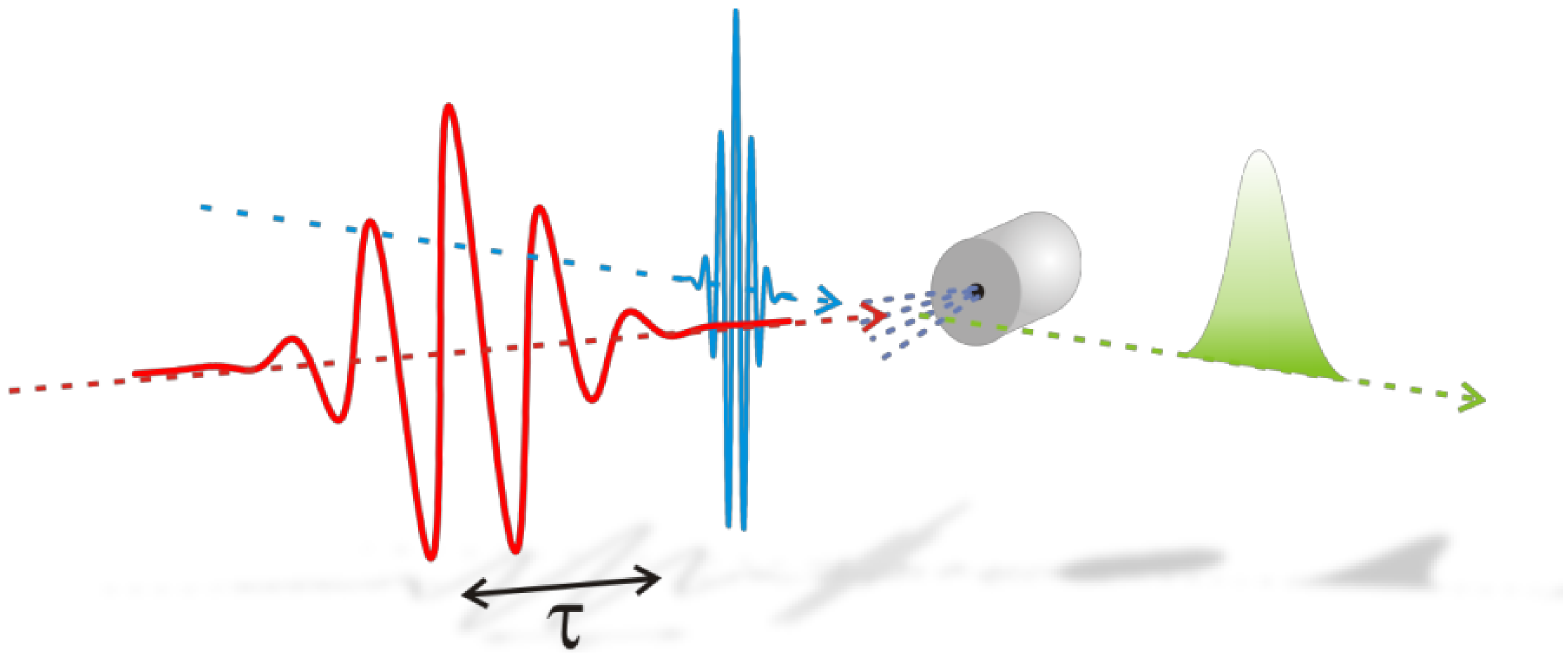
General scheme of attosecond metrology



far from resonances, attosecond electron wavepacket is a replica of the attosecond field
→ characterization of the electron wavepacket

FROG-CRAB

Frequency-Resolved Optical Gating for Complete Reconstruction of Attosecond Bursts



Y. Mairesse and F. Quéré, Phys. Rev. A 71, 011401 (R) 2005

FROG-CRAB

photoionization spectrum:

$$S(W,\tau) = \left| \int_{-\infty}^{+\infty} dt\ e^{i\phi(t)}\ \mathbf{d}\ \mathbf{E}_X(t-\tau)\ e^{i(W+I_p)t} \right|^2$$

delay between IR and XUV pulses

dipole transition element

XUV field

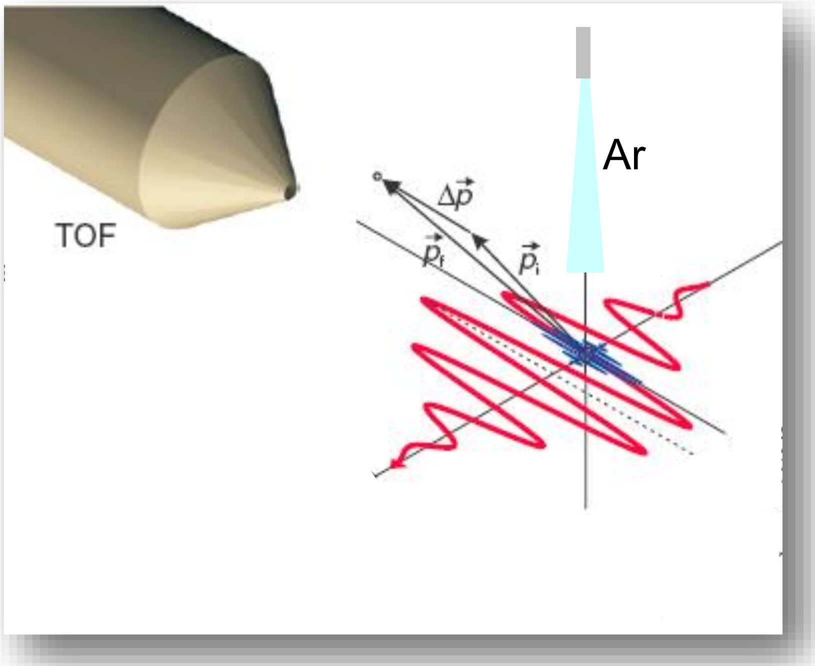
phase gate: $\phi(t) = -\int_t^{\infty} dt' [\mathbf{v}\cdot\mathbf{A}(t') + \mathbf{A}^2(t')/2]$

final electron velocity

IR vector potential

the IR laser field provides a phase gate for FROG measurements on attosecond bursts

Attosecond streak camera



initial electron momentum

$$p_i = \sqrt{2mW_o} \qquad W_o = \hbar\omega_{XUV} - I_p$$

effect of streaking pulse

$$\Delta\mathbf{p}(t) = e\int_t^{+\infty} \mathbf{E}_{IR}(t')\,dt' = e\mathbf{A}(t)$$

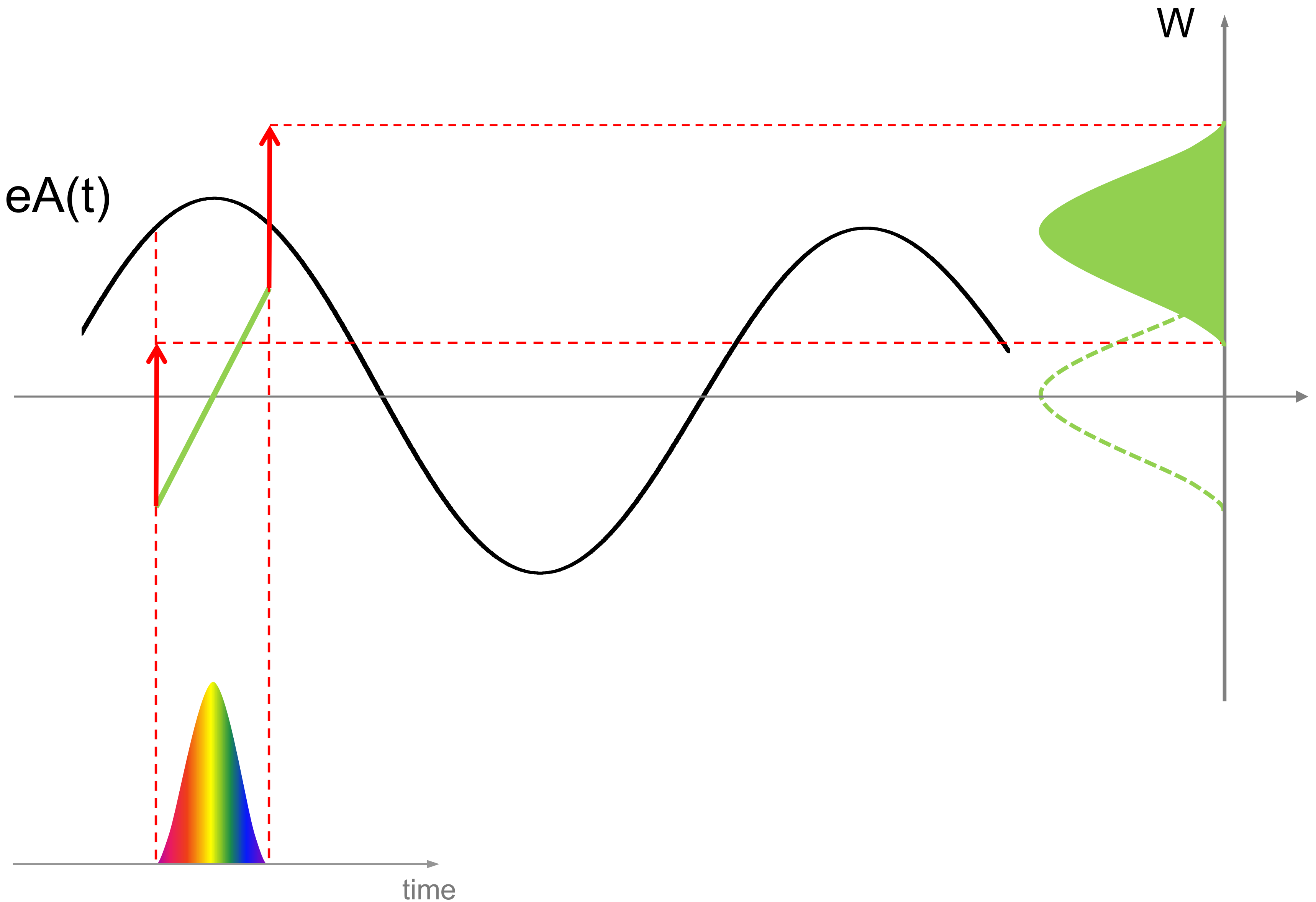
final electron momentum

$$\mathbf{p}_f(t) = \mathbf{p}_i + \Delta\mathbf{p}(t)$$

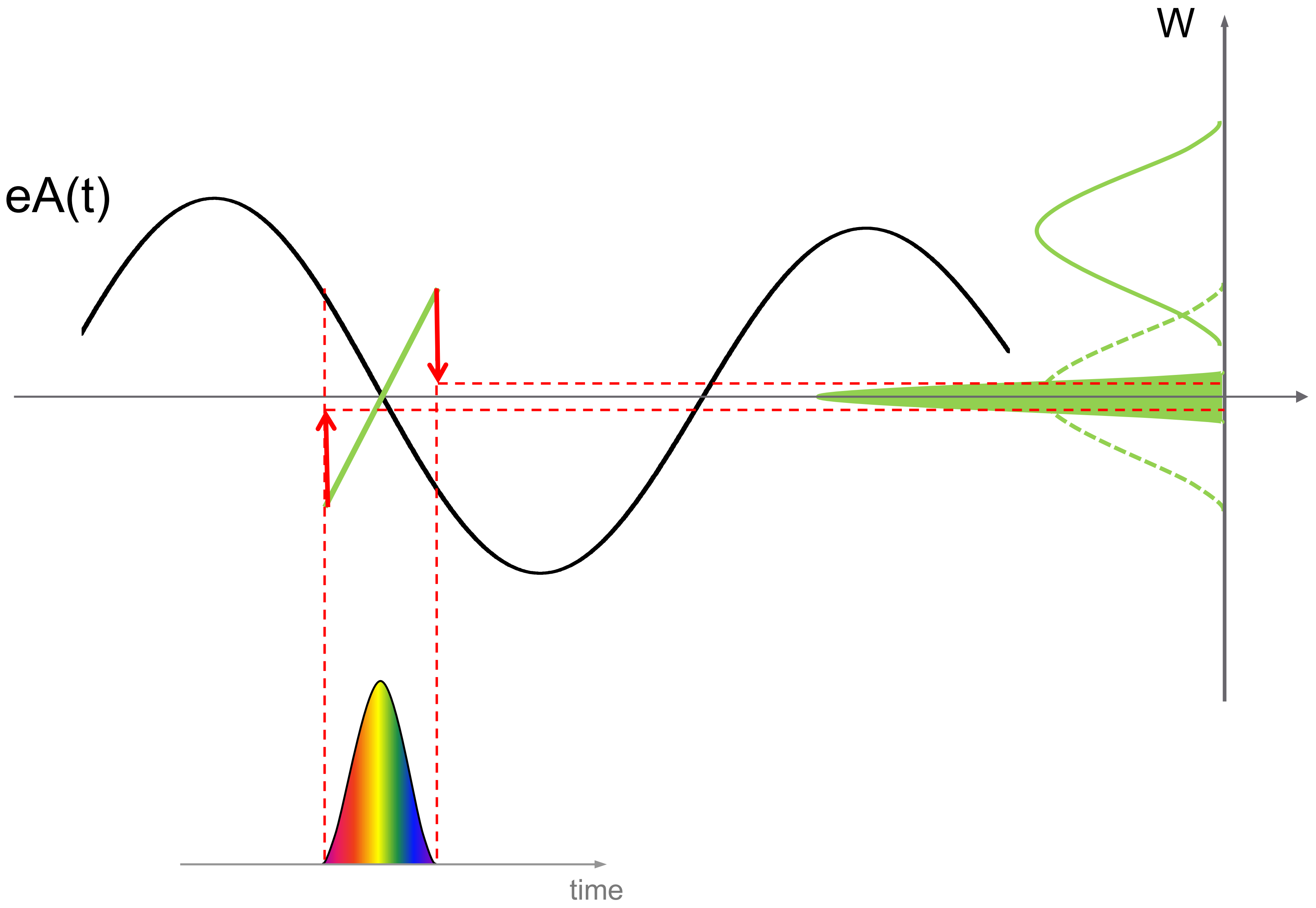
electron energy: $W(t) \approx W_o + \sqrt{8mW_o}\,eA(t)$

Kitzler et al. PRL 88, 173903 (2002)
Itatani et al. PRL 88, 173904 (2002)

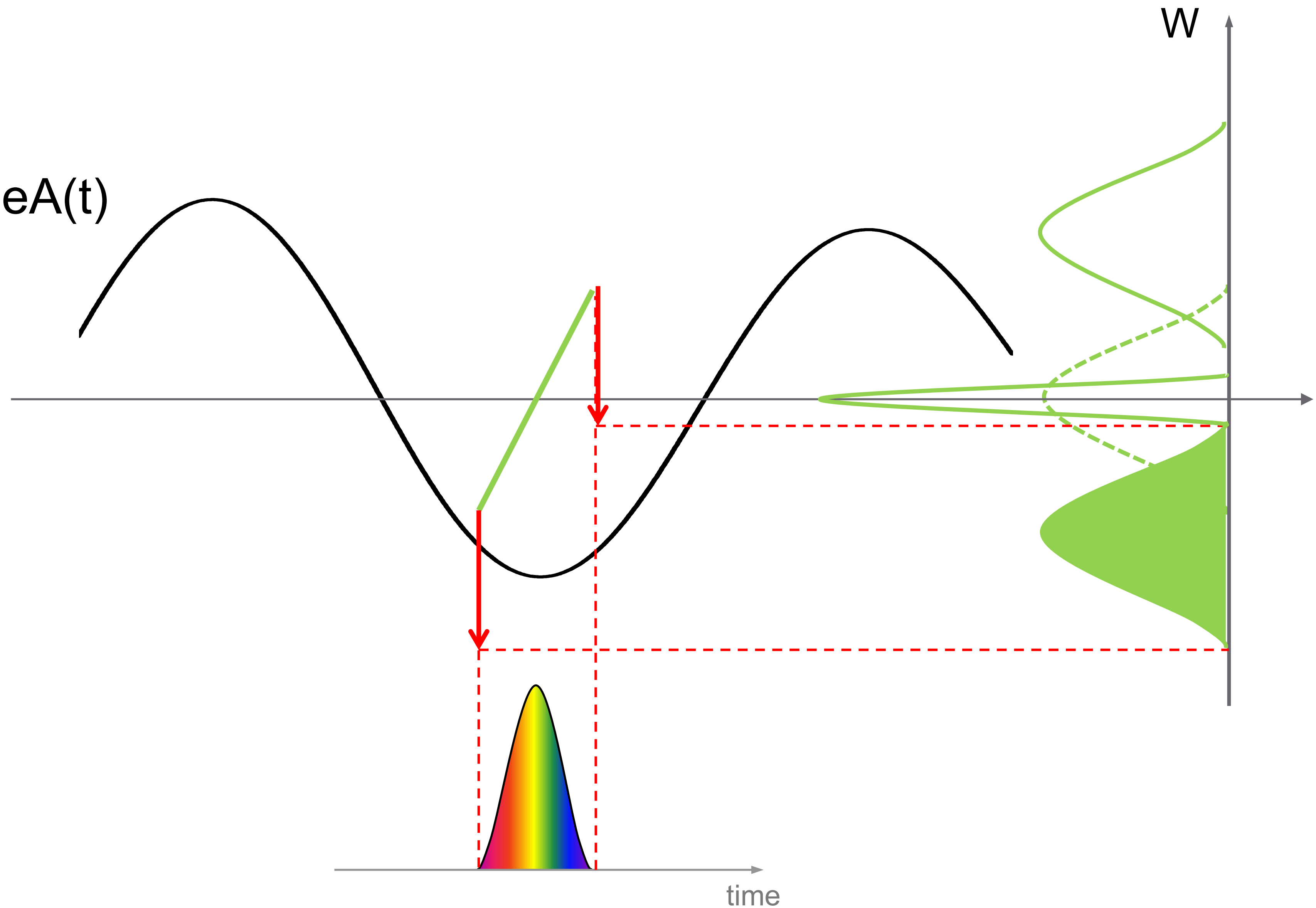
Attosecond streak camera



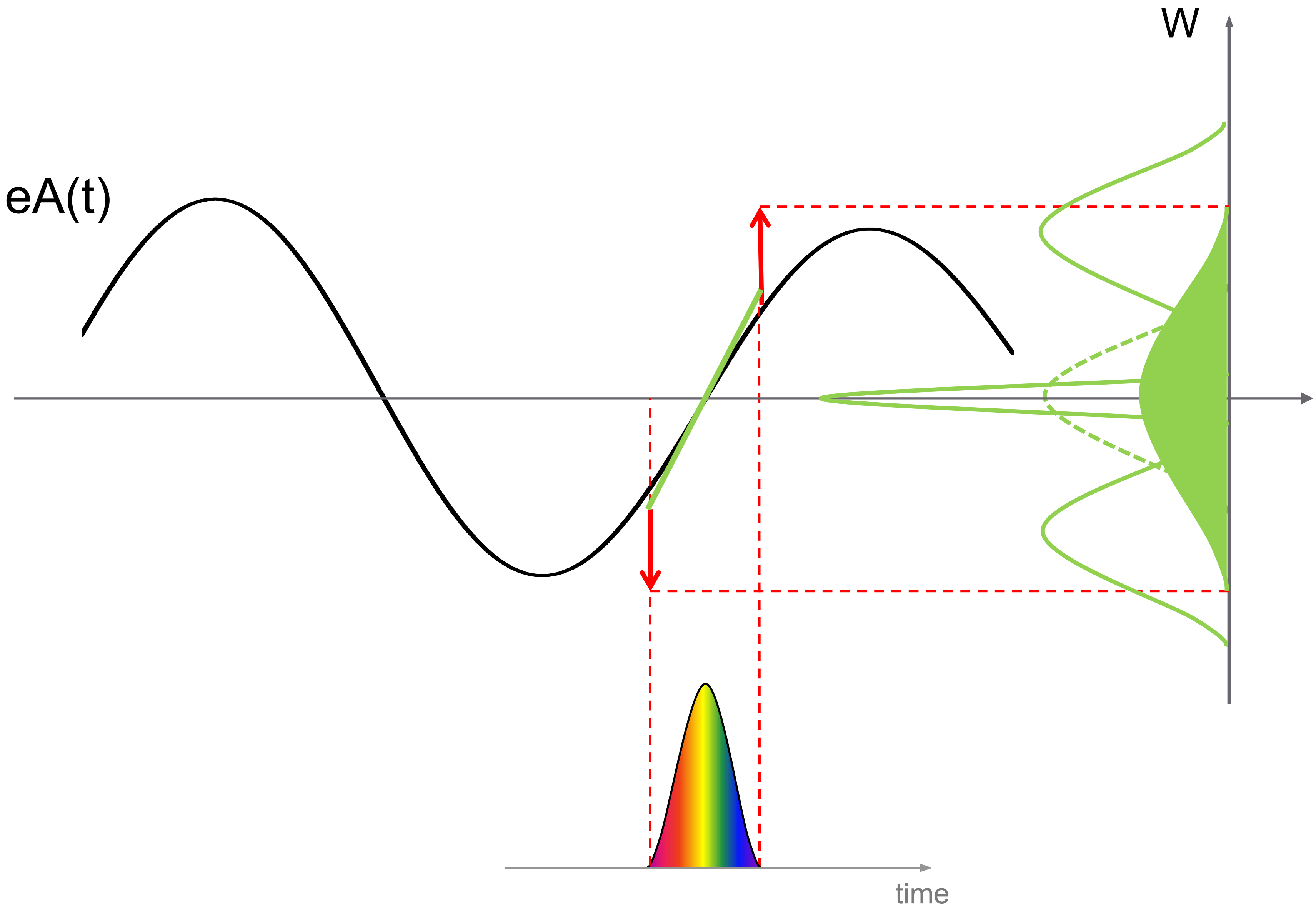
Attosecond streak camera



Attosecond streak camera

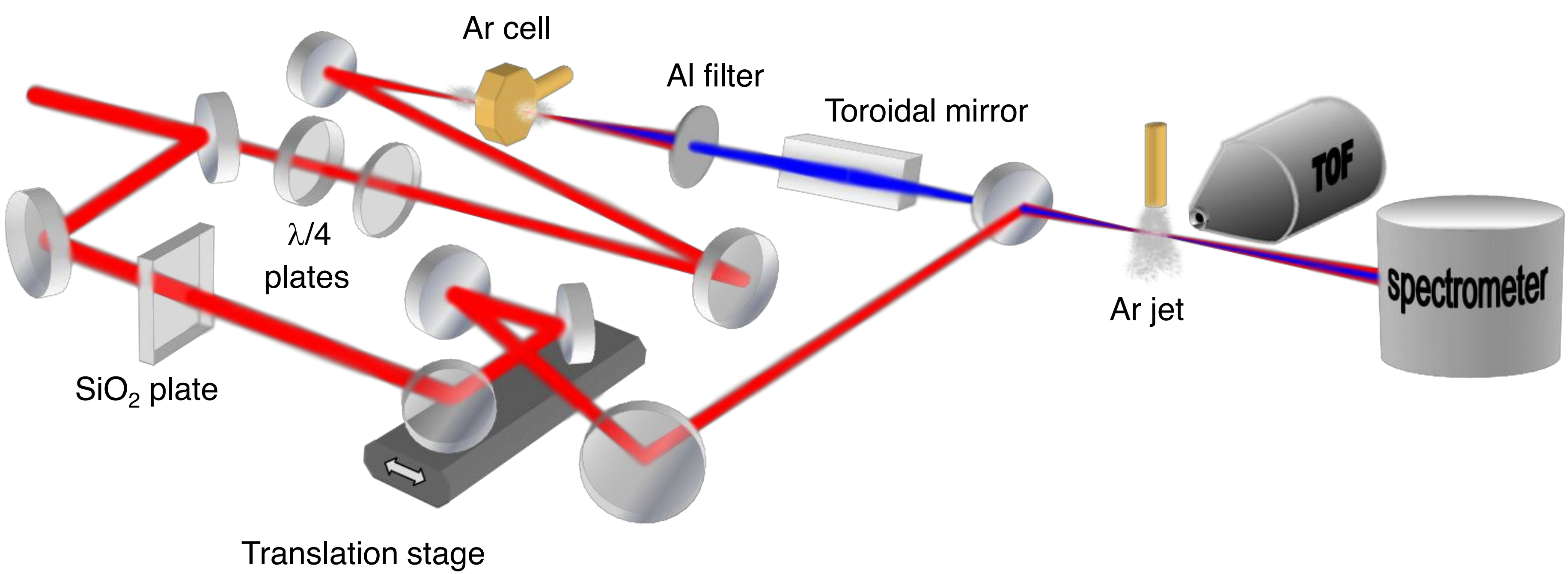


Attosecond streak camera



Attosecond Metrology

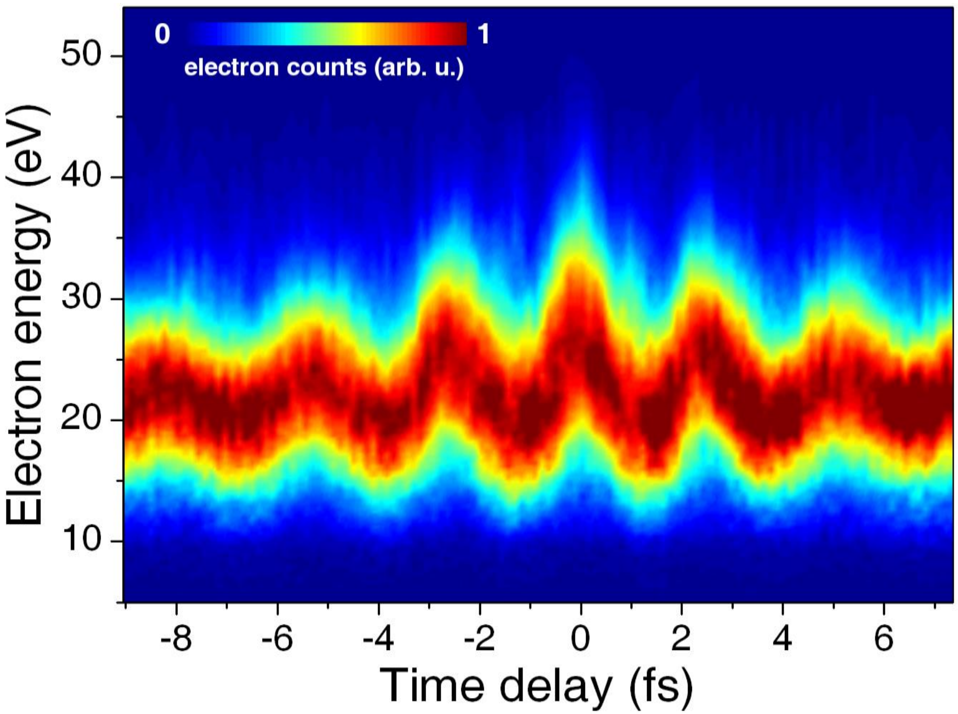
cross-correlation with driving light pulse



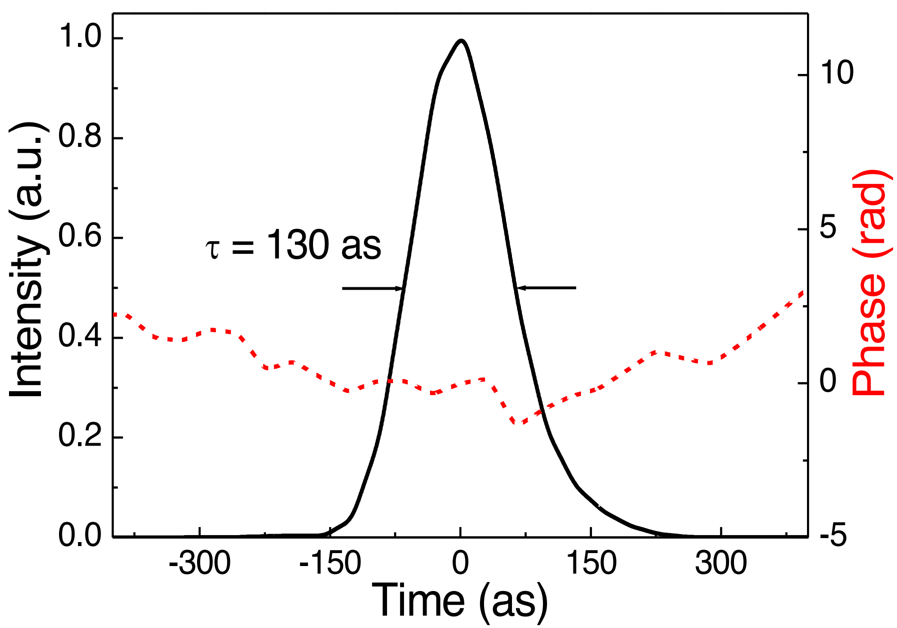
→ photoelectron spectra vs delay

Temporal characterization

dispersion compensation by 300-nm Al filter



retrieved Intensity profile and phase



good dispersion compensation
near-single cycle pulse!

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

III.
Applications

Attosecond spectroscopy

attosecond-scale electronic dynamics in molecules do affect chemical changes!

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

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

“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

Attosecond measurements

XUV ionization followed by **acceleration** of the ionized electron in a strong IR field (**streak camera approach**)

Attosecond pulse characterization

Attosecond streaking spectroscopy

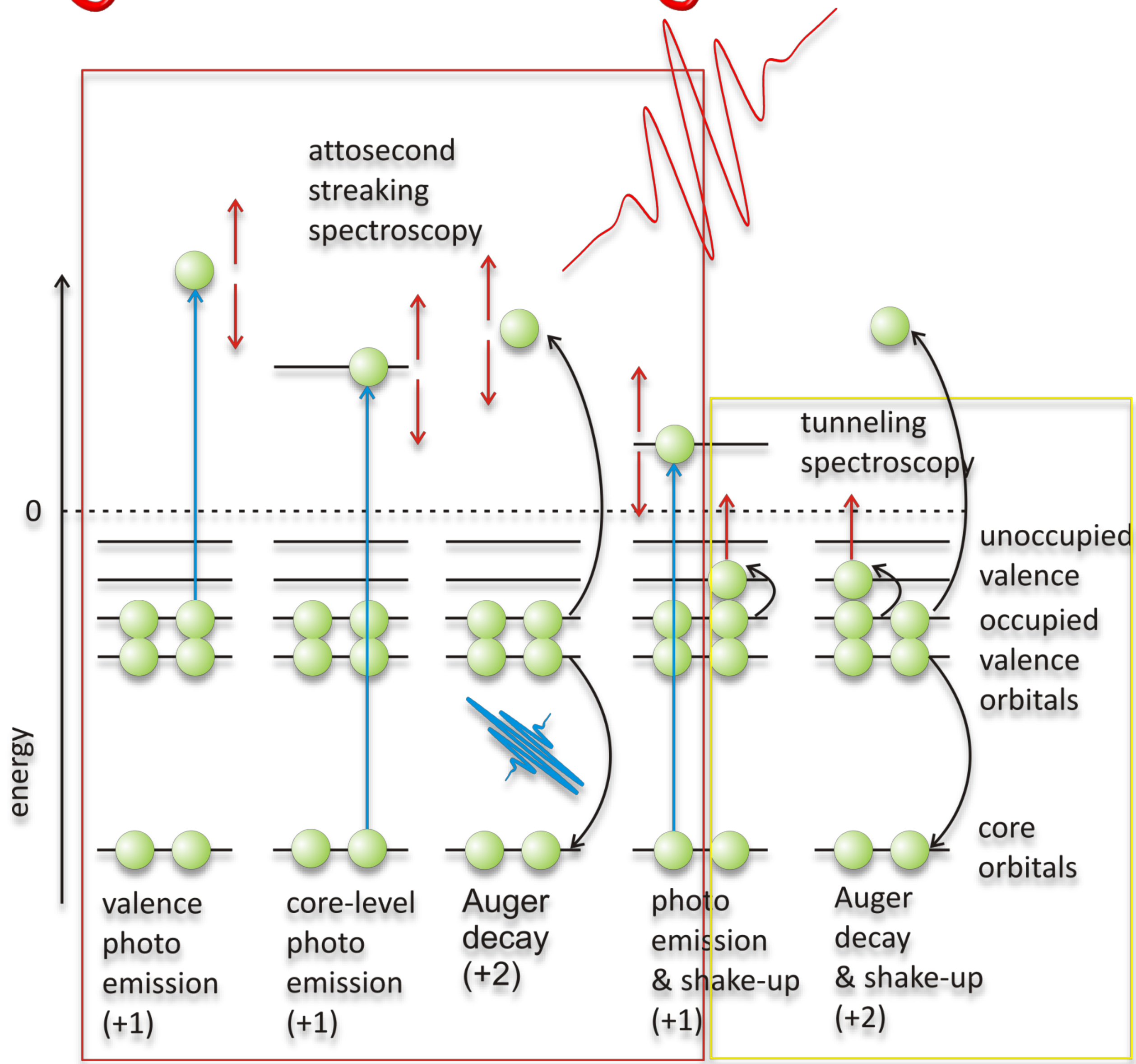
Relaxation dynamics of core-excited atoms, M. Drescher et al., Nature **419**, 803 (2002).
Attosecond spectroscopy in condensed matter, A. Cavalieri et al., Nature **449**, 1029 (2007)

XUV excitation of **bound states**, followed by ionization in a strong IR field

Real-time observation of electron tunnelling and multi-electron dynamics in atoms

M. Uiberacker *et al.*, Nature **446**, 627 (2007)

Probing electron dynamics

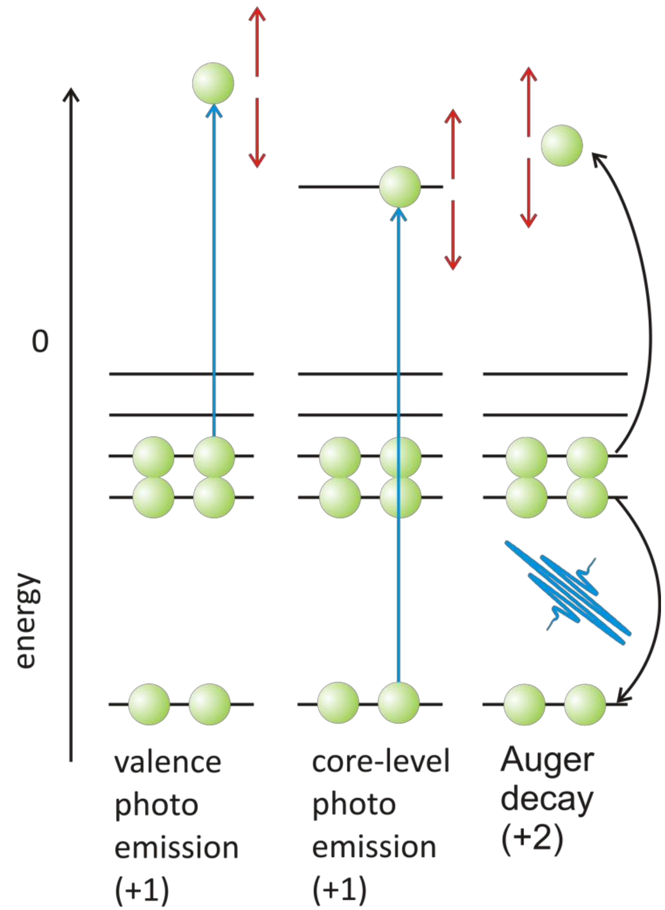


P. B. Corkum & F. Krausz, Nat. Physics 3, 381 (2007)

As streaking spectroscopy

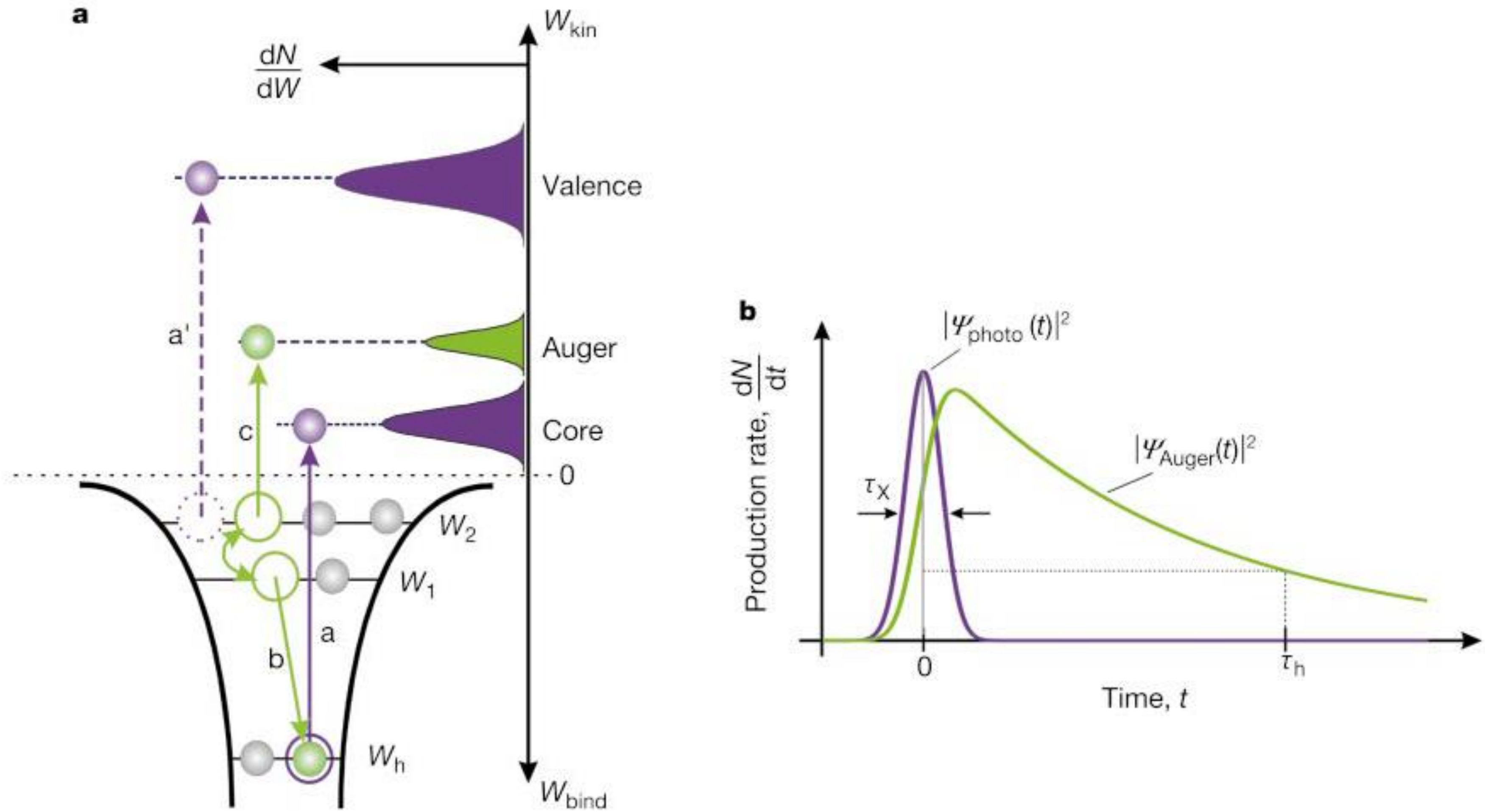
<u>as pump –as probe</u>:
- low flux of as pulses
- low two-photon transition probabilities in the XUV

<u>as streaking spectroscopy:</u>
- few-cycle IR pulse with controlled waveform + nonlinear process may replace the attosecond pulse either in probing or starting electron dynamics
- probing inner-atomic relaxation dynamics



As Streaking Spectroscopy

time-domain observation of the decay of an inner-shell vacancy via Auger relaxation in isolated atoms

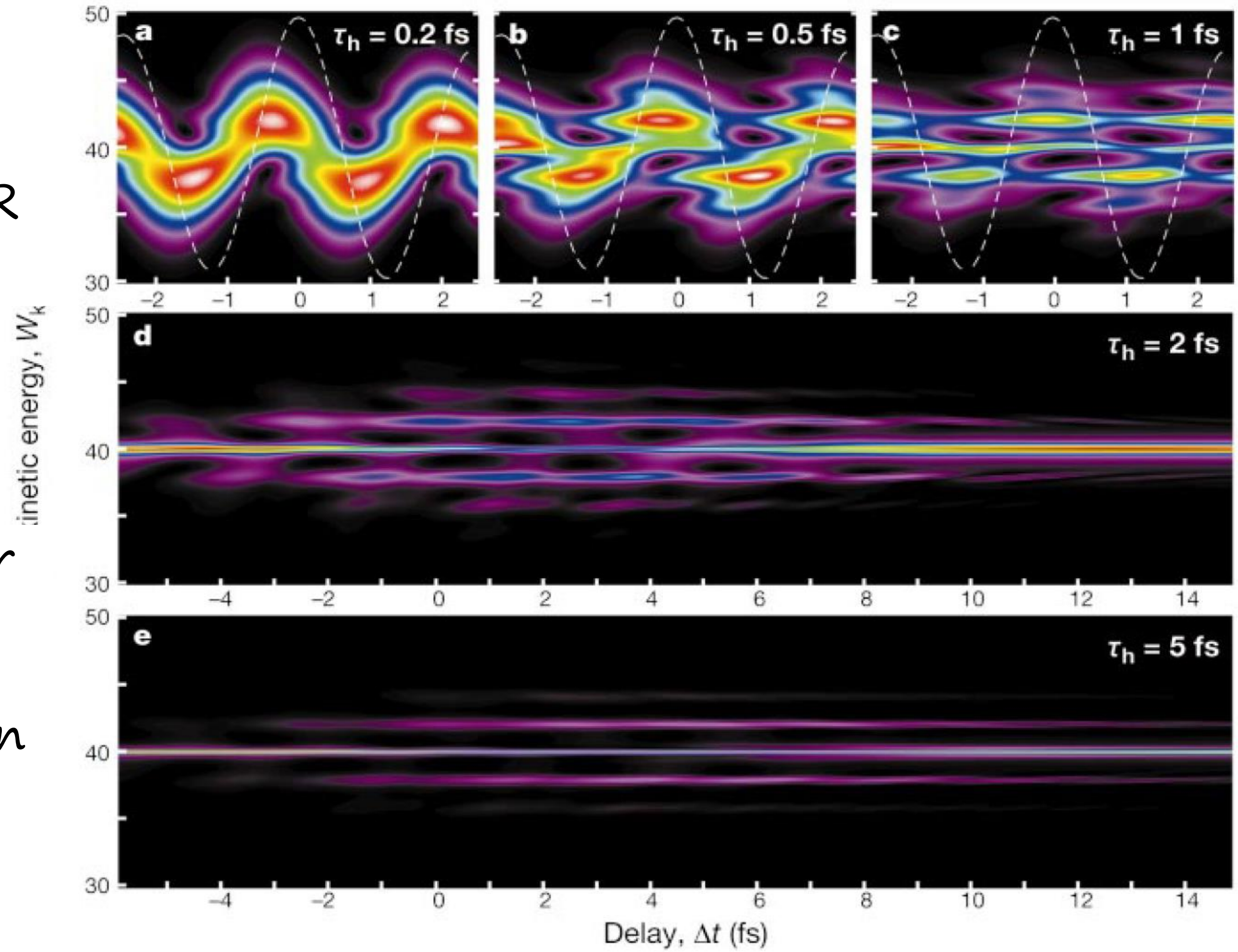


M. Drescher et al., Nature 419,803 (2002)

As Streaking Spectroscopy

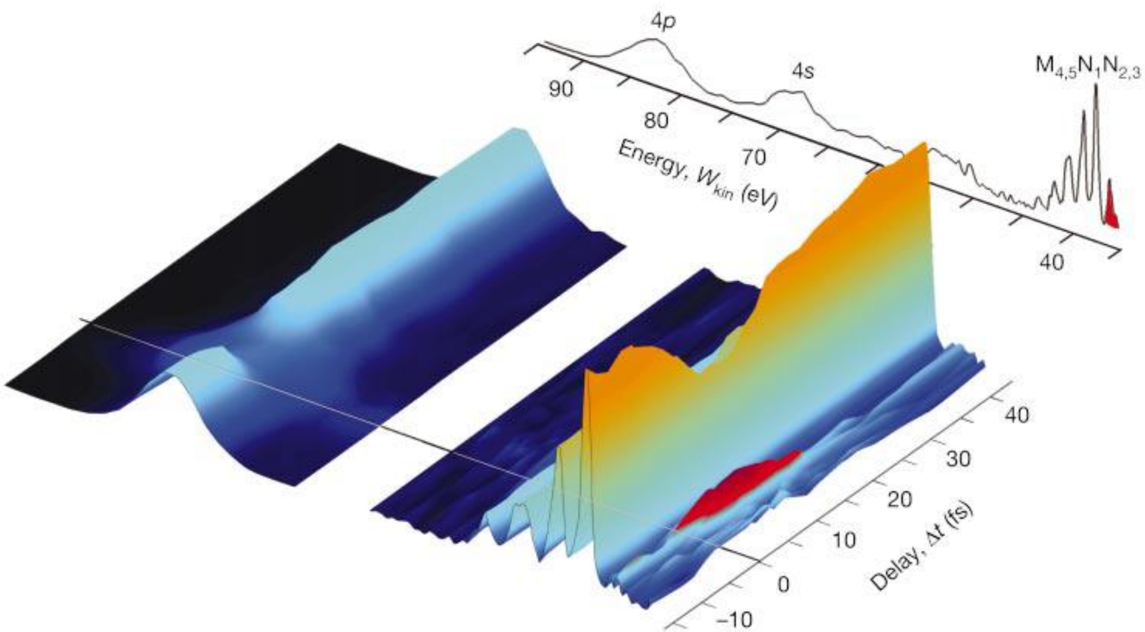
Decay much faster than period of IR: Auger electron maps out the oscillation of the IR field

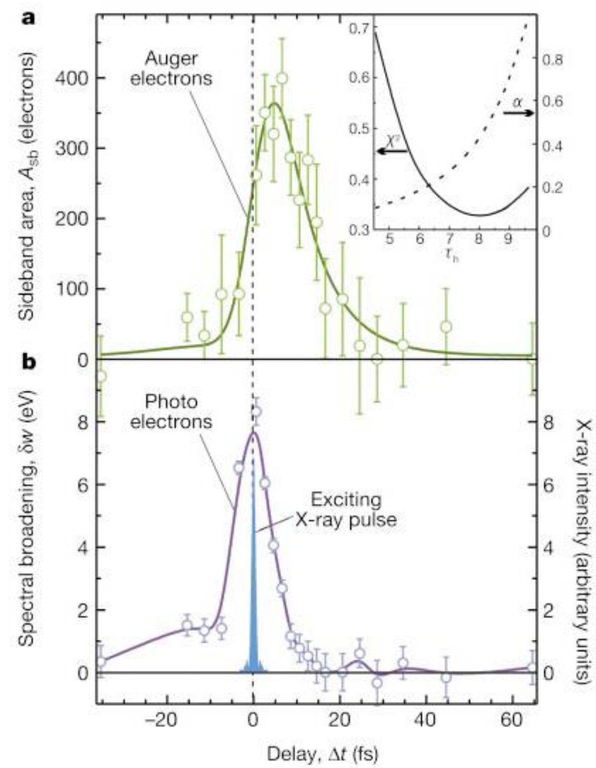
Decay much slower than period of IR: measure cross correlation between IR pulse duration and Auger decay



M. Drescher et al., Nature 419,803 (2002)

Streaked electron spectra following core-hole excitation



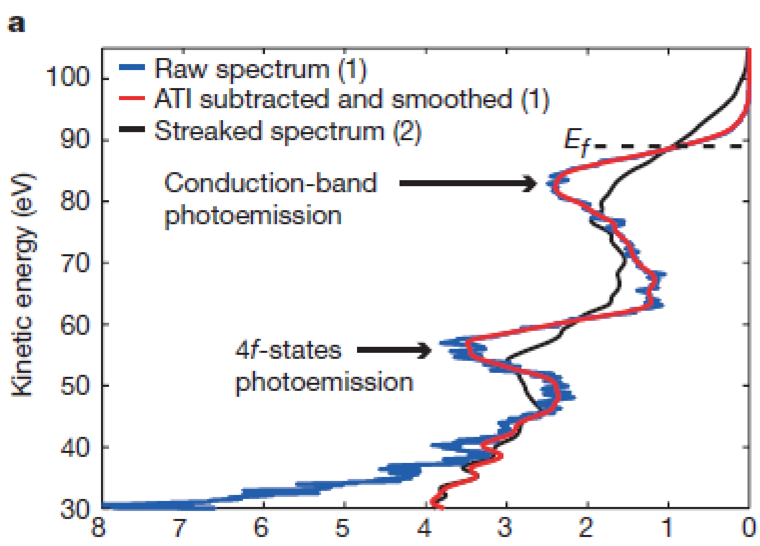


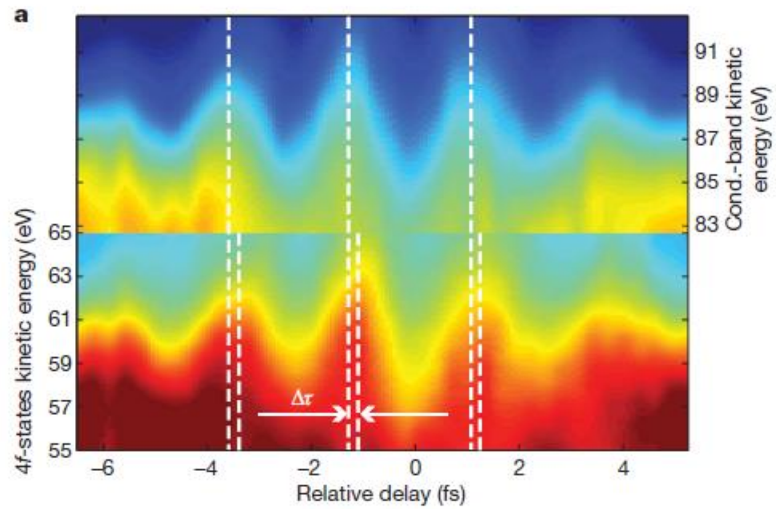
- In this experiment the Auger decay was slower than the optical period of the IR laser
- A decay time of the excitation of 7.9±1 fs could be inferred

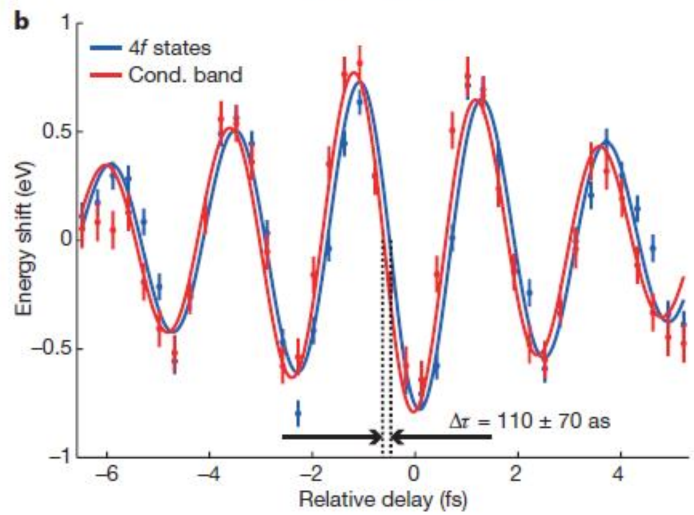
M. Drescher et al., Nature 419,803 (2002)

As spectroscopy in condensed matter

probing photoelectron emission from single-crystal tungsten



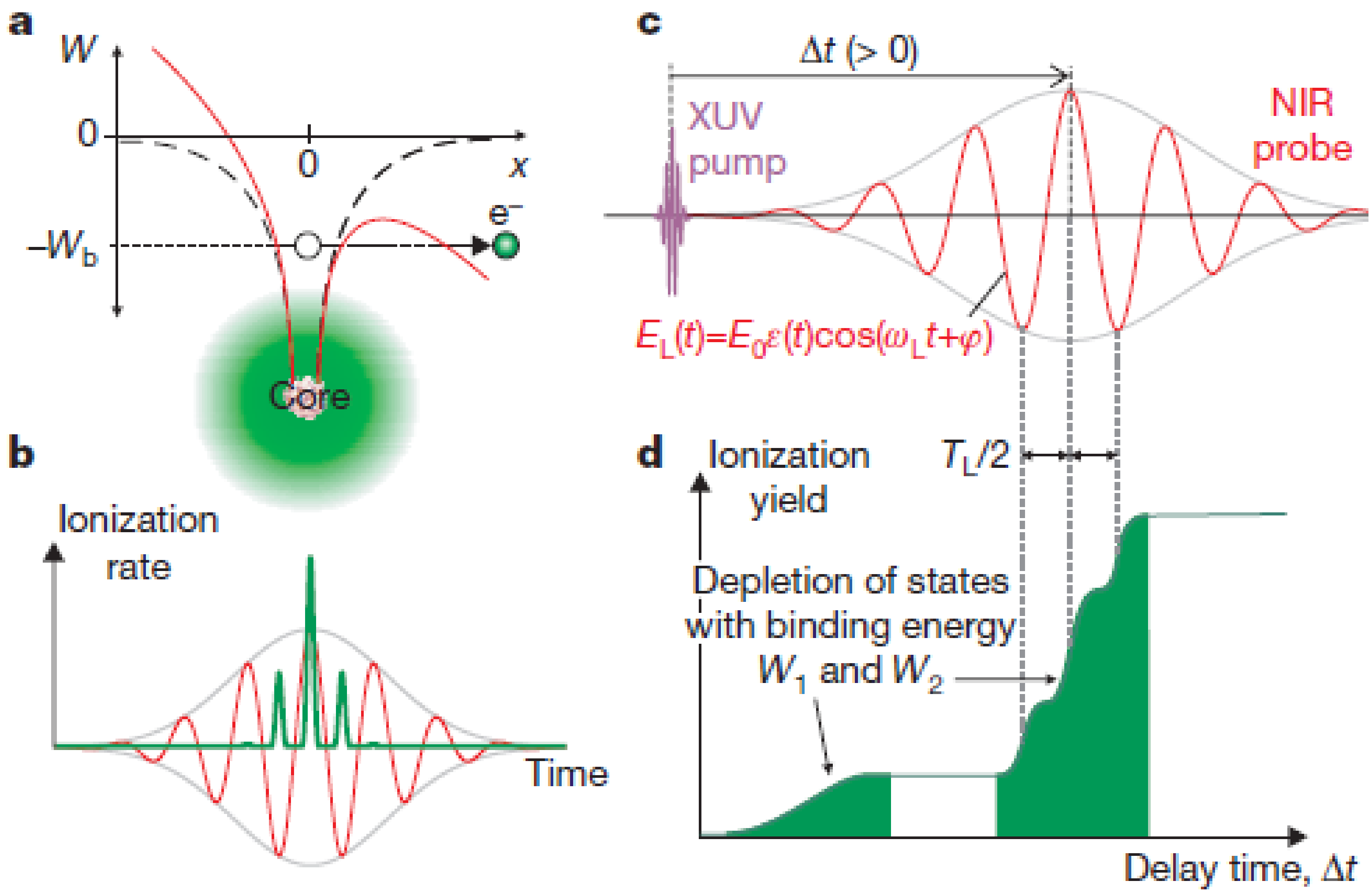




- Sub-fs photoemission from 4f core states and from conduction band
- Extension of streaking spectroscopy to condensed matter
- 100-as delay between photoelectron emission from localized core states and from delocalized conduction-band states

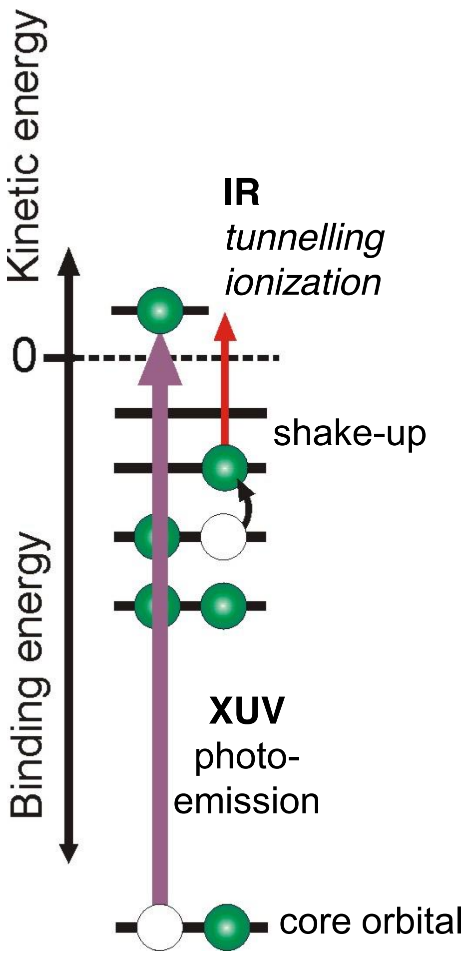
A. Cavalieri et al., Nature 449, 1029 (2007)

XUV excitation of bound states followed by ionization in a strong IR field

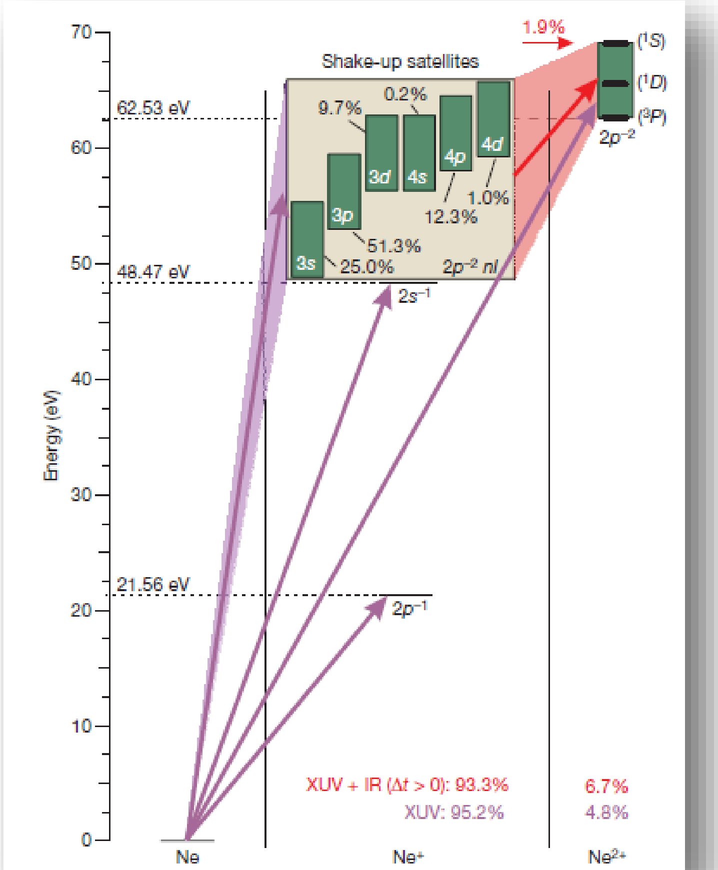


M. Uiberacker et al., Nature 446, 627 (2007)

Shake-up state in Ne

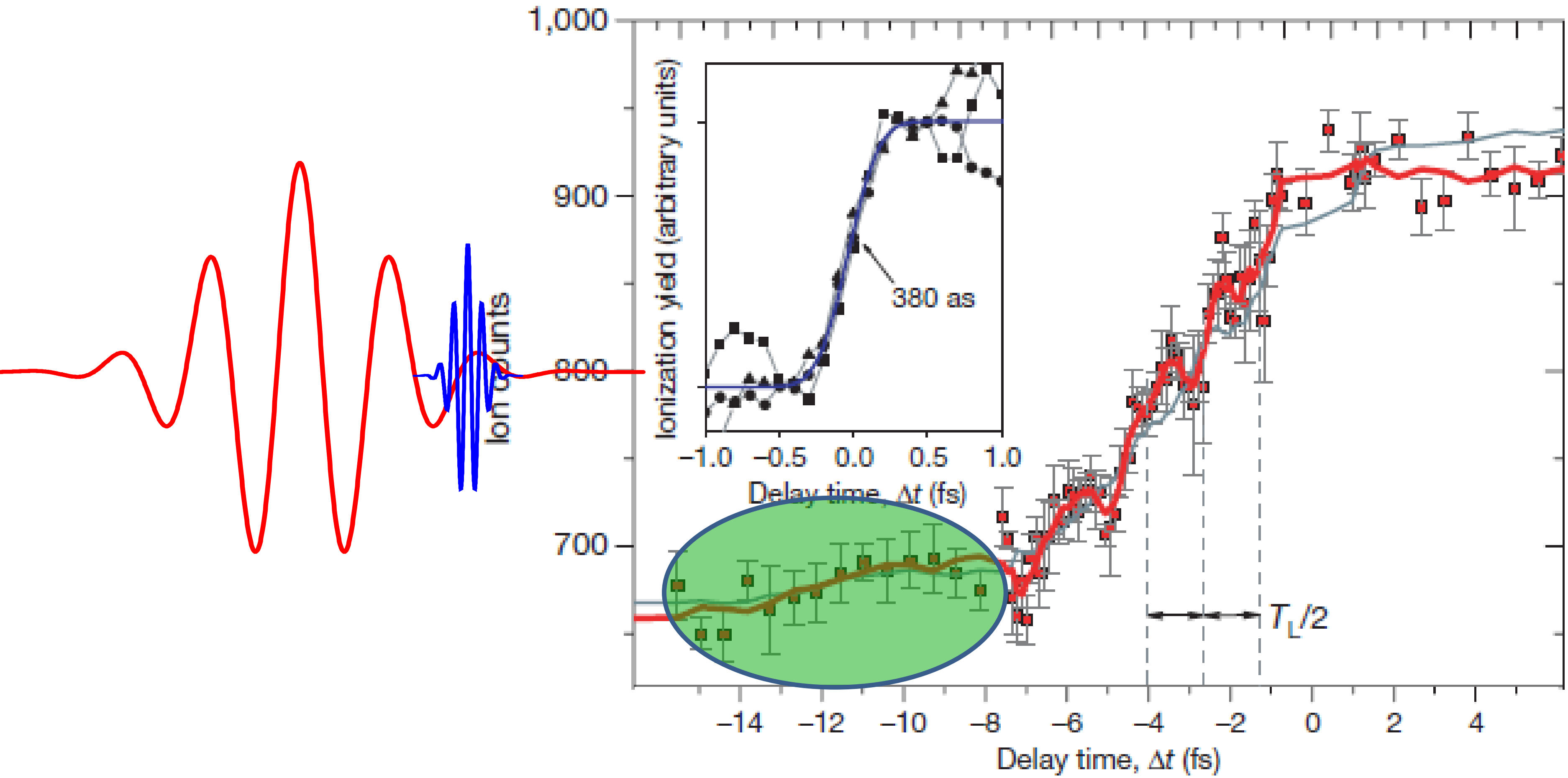


1) ionization from the 2p level
2) excitation of a second 2p level to an excited ionic states
3) ionization of excited state by an IR field
4) doubly charged ions



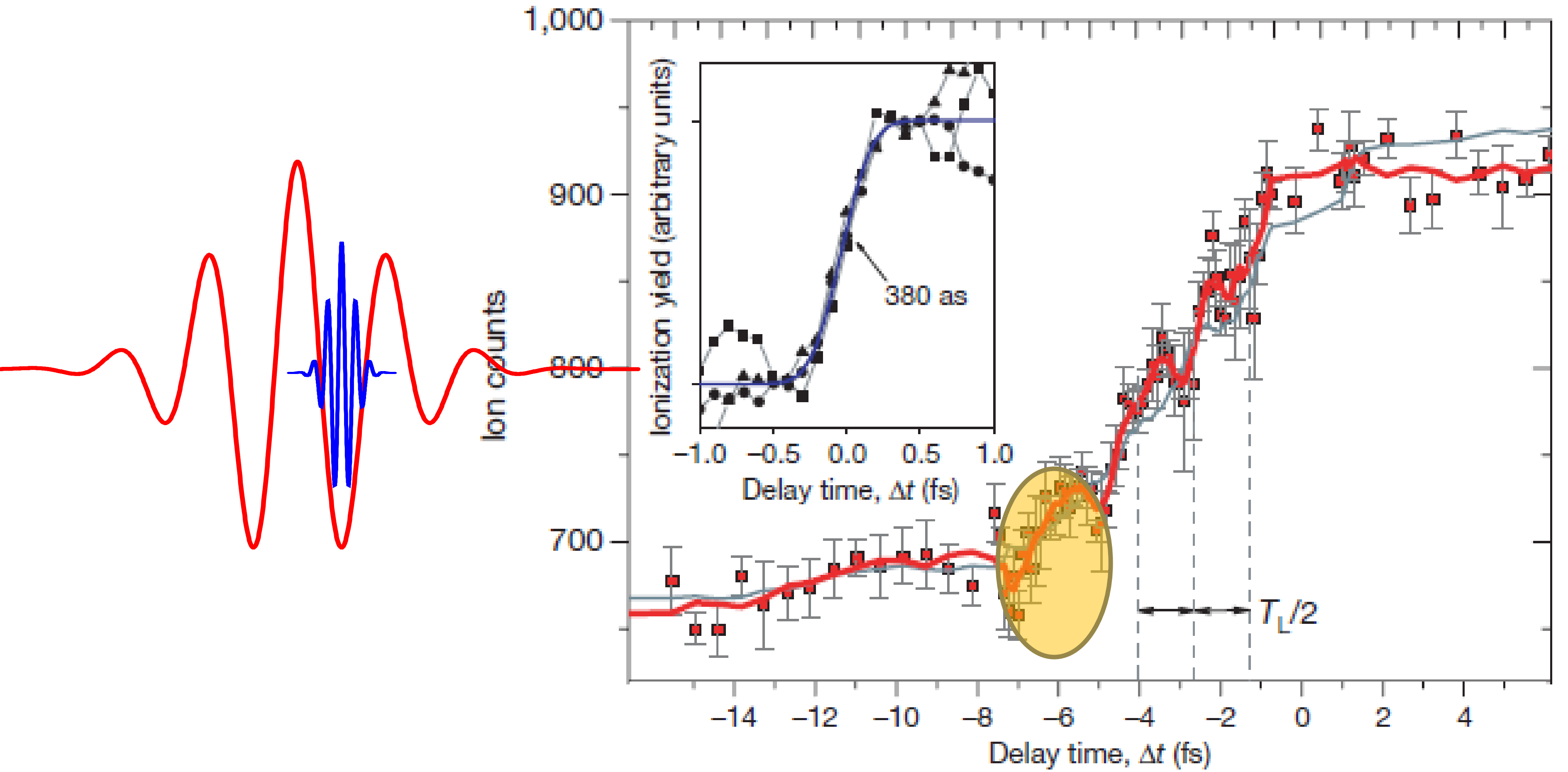
M. Uiberacker et al., Nature 446, 627 (2007)

Tunneling ionization in Ne

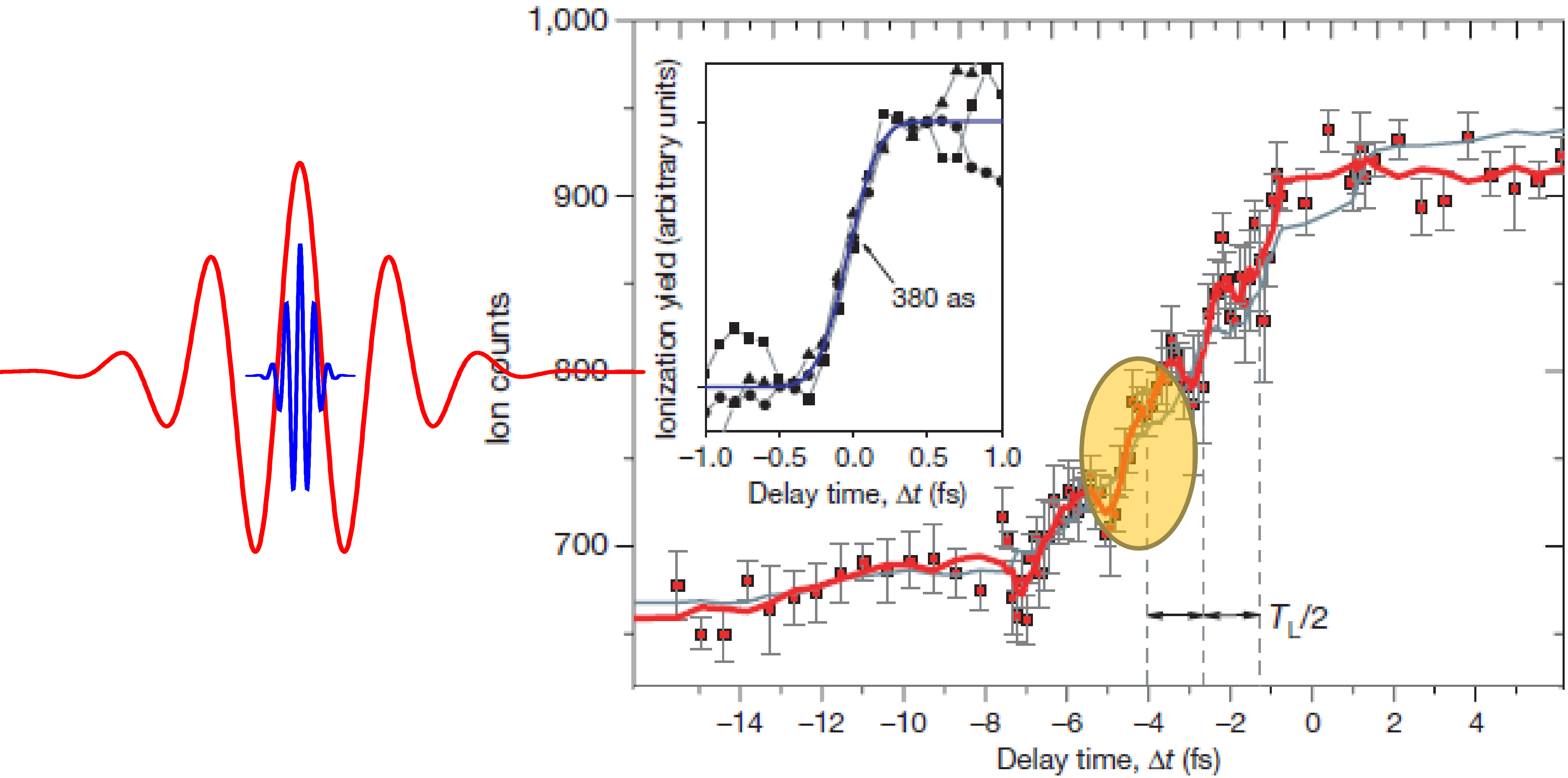


M. Uiberacker et al., Nature 446, 627 (2007)

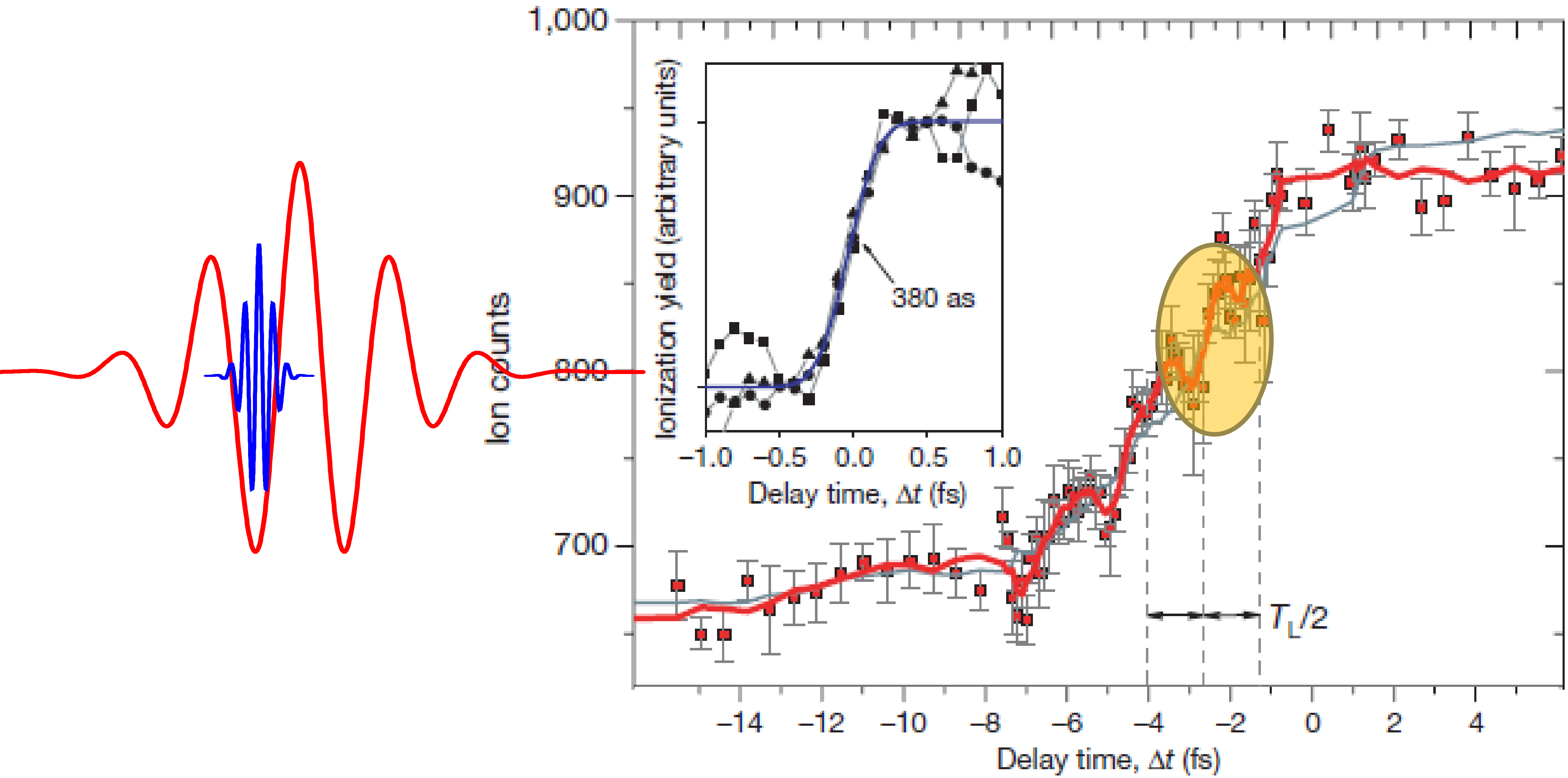
Tunneling ionization in Ne



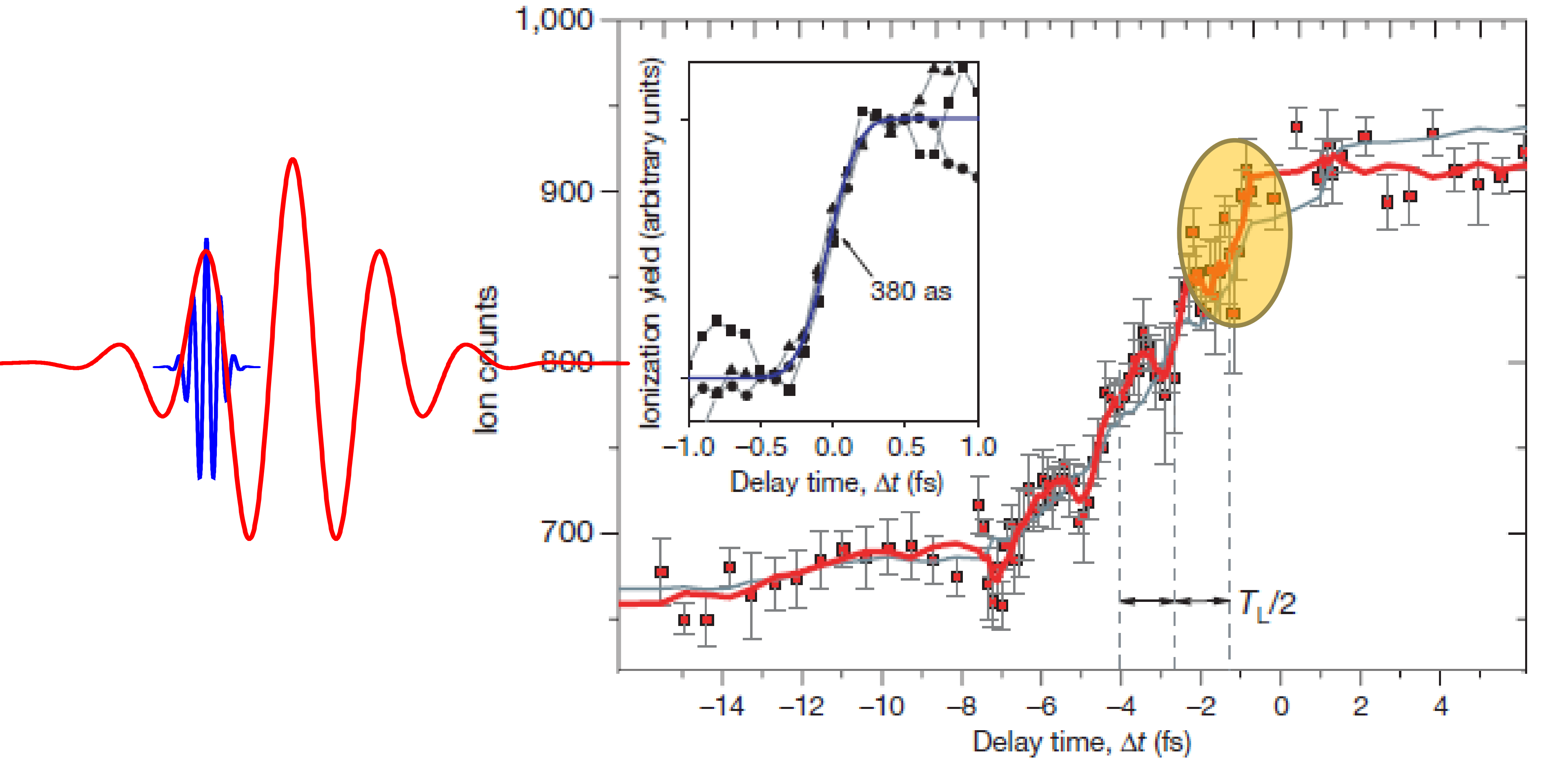
Tunneling ionization in Ne



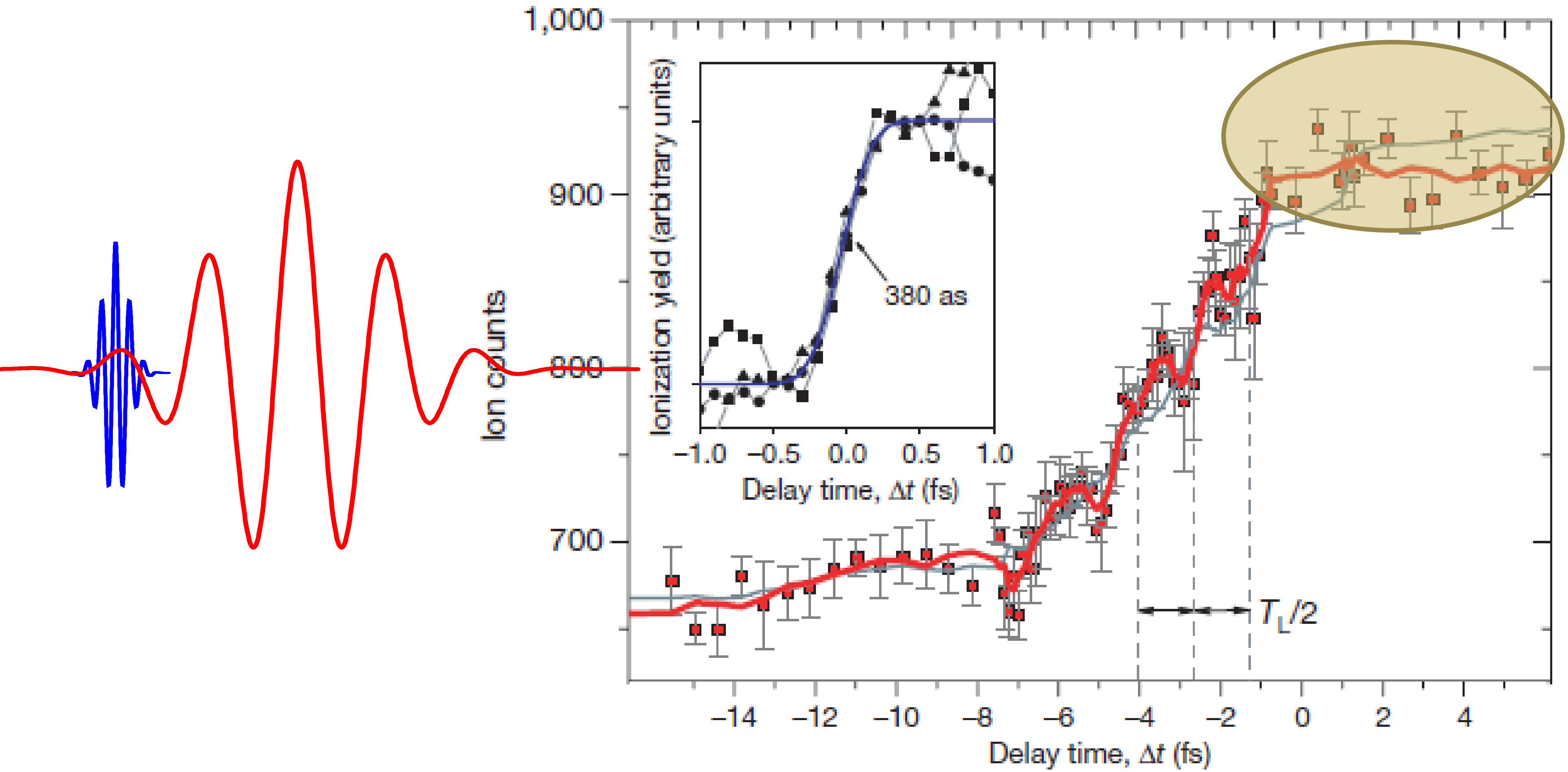
Tunneling ionization in Ne



Tunneling ionization in Ne

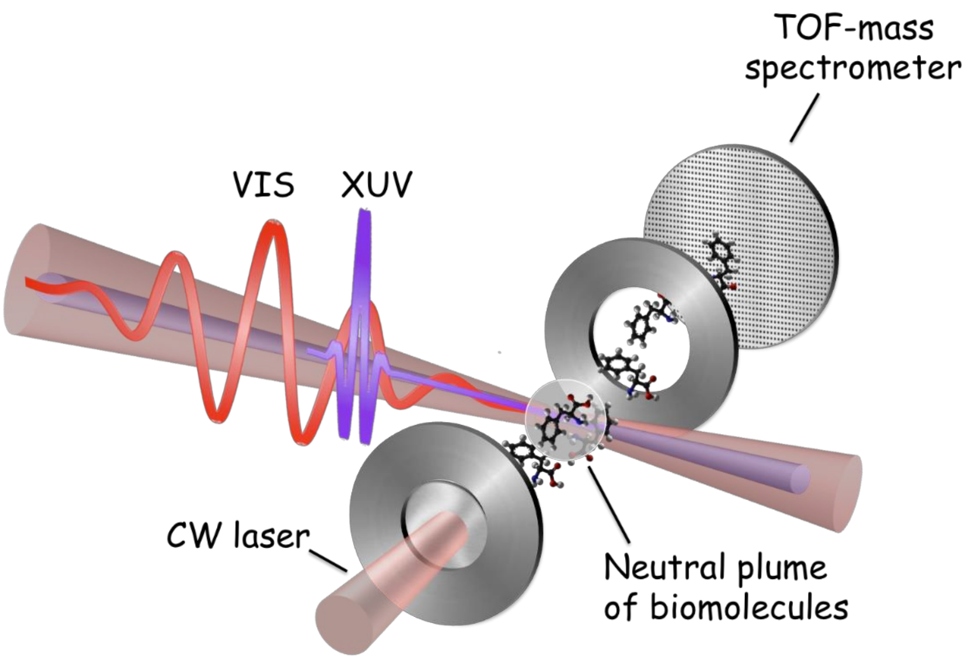


Tunneling ionization in Ne



Investigation of Ultrafast Electron Dynamics Triggered by Attosecond XUV Pulses in Amino Acids

Francesca Calegari



Application to biomolecular building blocks:

Aromatic amino acids evaporated in a TOF-mass spectrometer

Mass of fragments produced by XUV pump and VIS/NIR probe pulses measured as a function of the pump probe delay with attosecond time precision

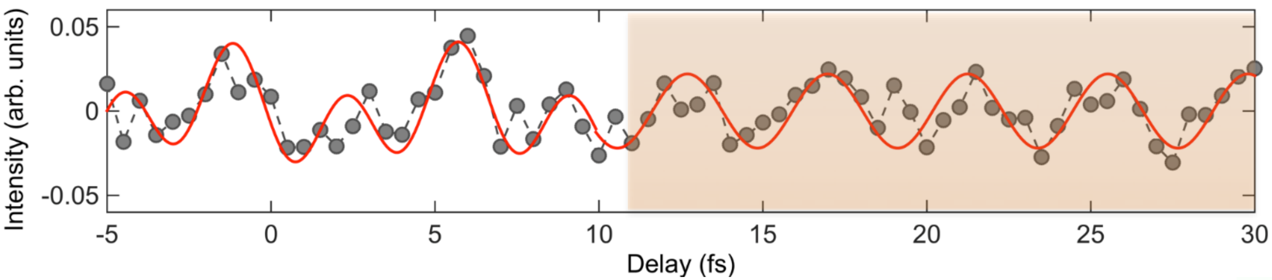
L. Belshaw et al., J. Phys. Chem. Lett. 3, 3751(2012)
F. Calegari et al., JSTQE 21, 2419218 (2015)

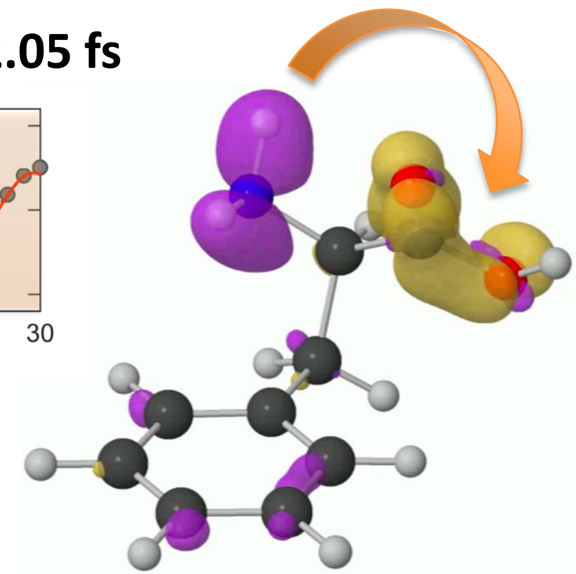
Investigation of Ultrafast Electron Dynamics Triggered by Attosecond XUV Pulses in Amino Acids

Francesca Calegari

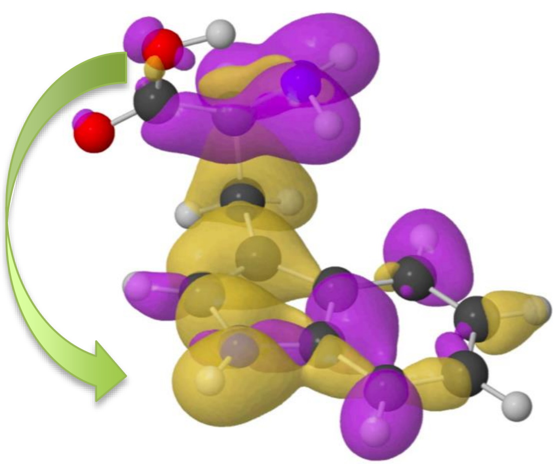
Electron migration from the amino group to the carboxylic group in **2.05 fs**

Phenylalanine – m/q= 60



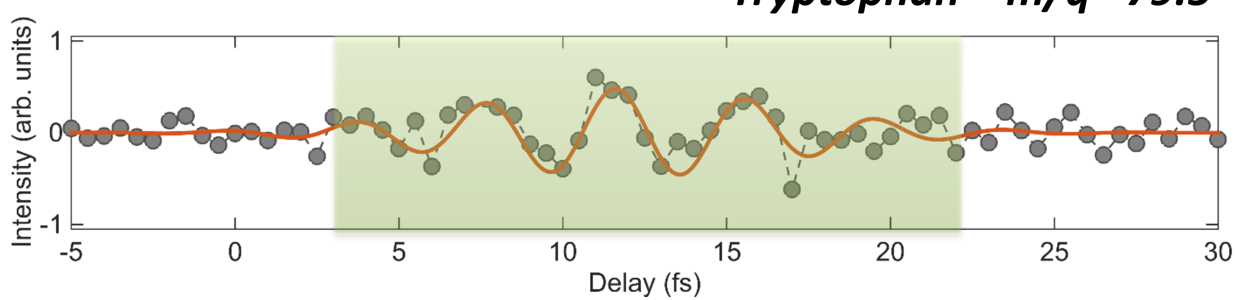


*F. Calegari et al., Science **346**, 336 (2014)*



Electron migration from the amino group to the indole group in **2.15 fs**

Tryptophan – m/q= 79.5



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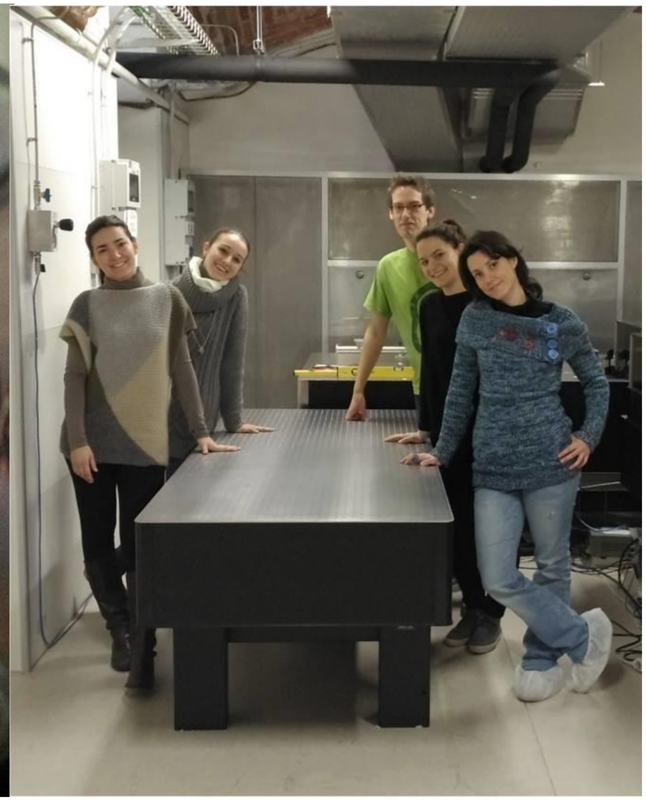


European Research Council

Established by the European Commission

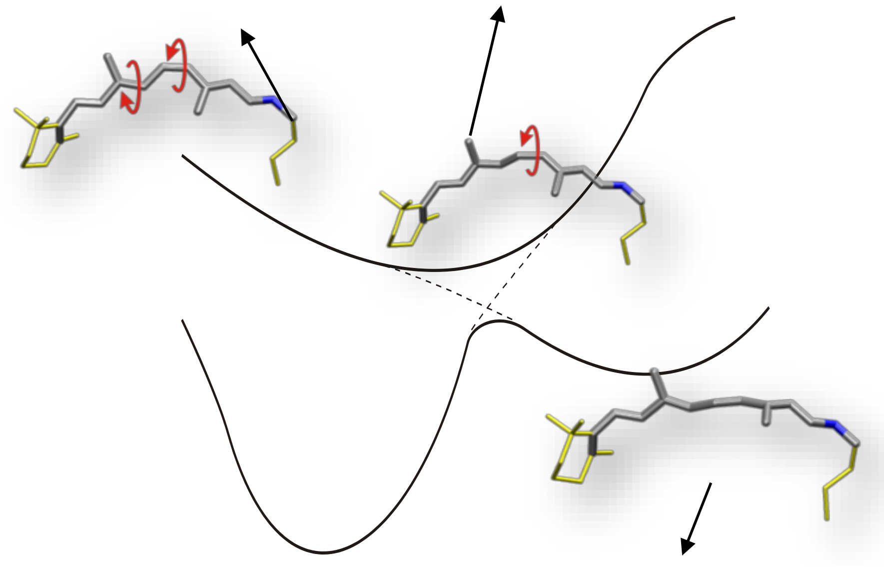
Molecular imaging

Motivation

Shooting the "molecular movie":

chemical properties of molecules are determined by the outermost electronic structure

Motivation

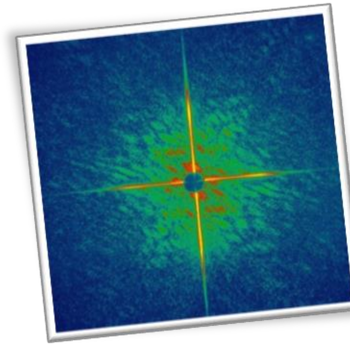


Imaging of structural changes in molecules

- direct access to excited states
- visualization of conical intersections
- precursor of coherent control

Molecular imaging

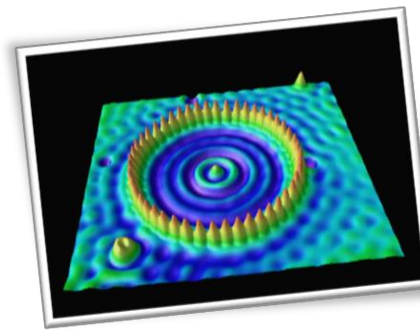
Imaging of the total electron density:



- X-ray diffraction
- electron scattering

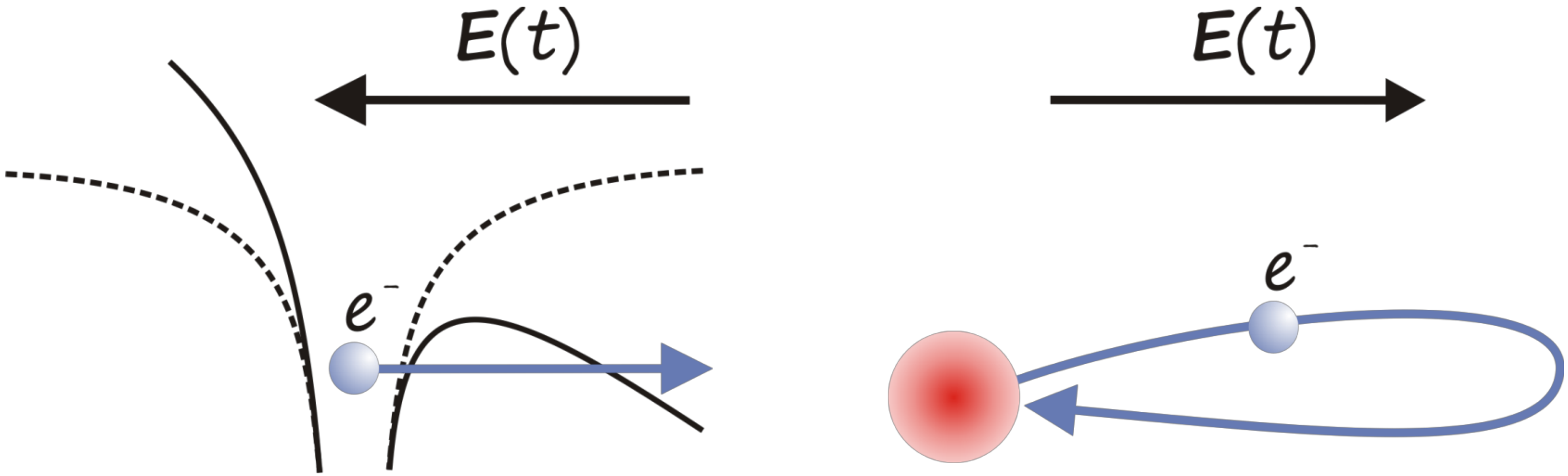
Imaging of the outermost electronic structure:

- electron momentum spectroscopy
- scanning tunneling microscopy
- high order harmonic generation



→ simple experimental technique
→ table top setup
→ temporal resolution (as to tens of fs)

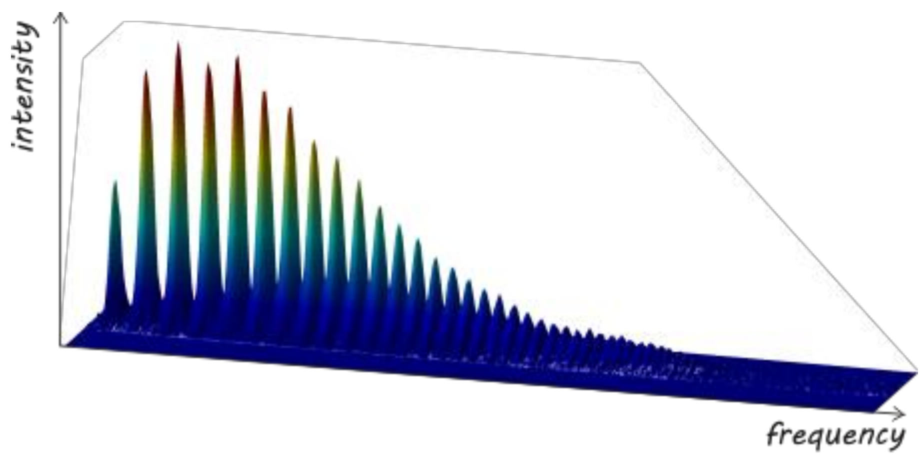
HHG as an interferometer

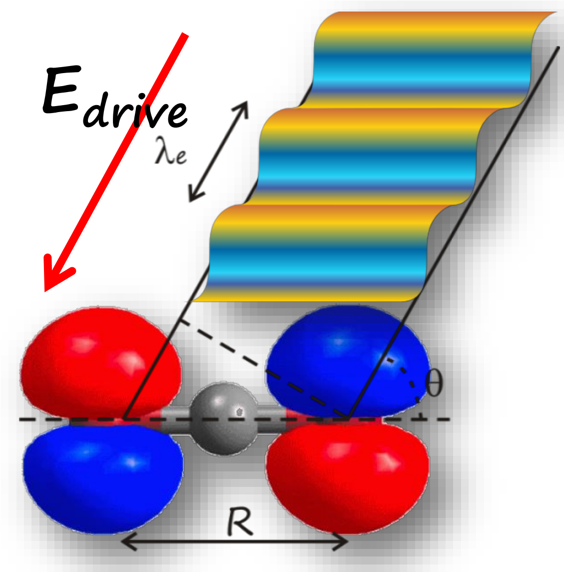


tunnel ionization → beam splitter
electron wave-packet motion → delay line
re-collision → interference

HHG for molecular imaging

orbital structure and symmetry are encoded in the harmonic spectrum





interatomic separation in matter: ~1 Å

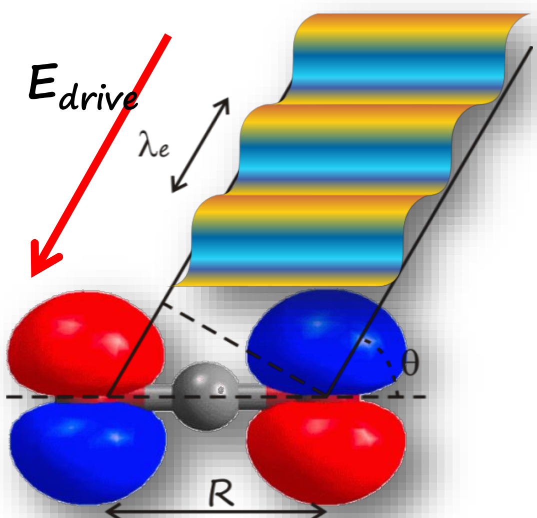
typical structure size of valence electron orbitals: ~1 Å

wavelength of a typical re-collision electron: ~1-2 Å

HHG tomography recipe

the transition dipole moment is the spatial Fourier transform of $r\Psi(r)$

$$\langle \Psi(r)|r|\exp[ik(\omega)\cdot r]\rangle$$

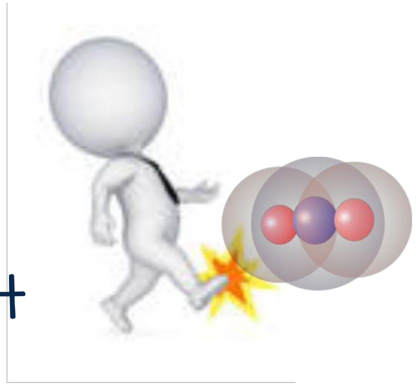


tomographic reconstruction of $\Psi(r)$:
the optical frequencies ω
map the spatial frequencies k

1 - align the molecule
2 - drive HHG for different angles

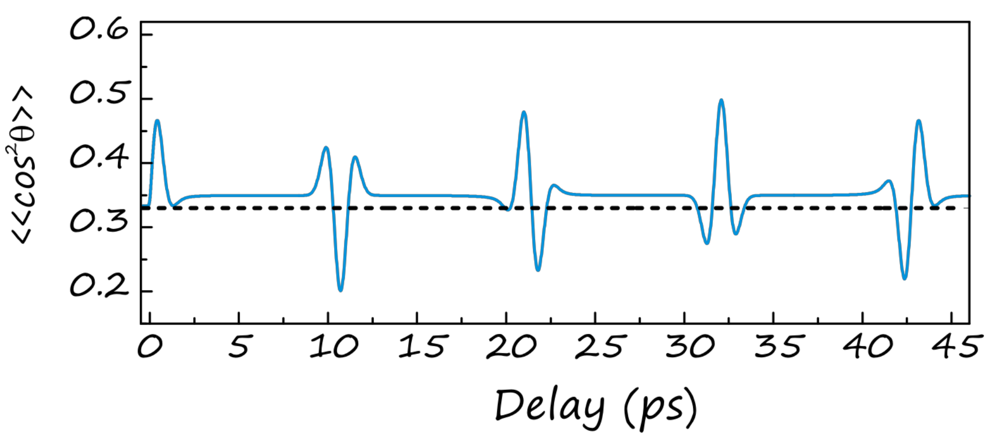
Impulsive alignment of molecules

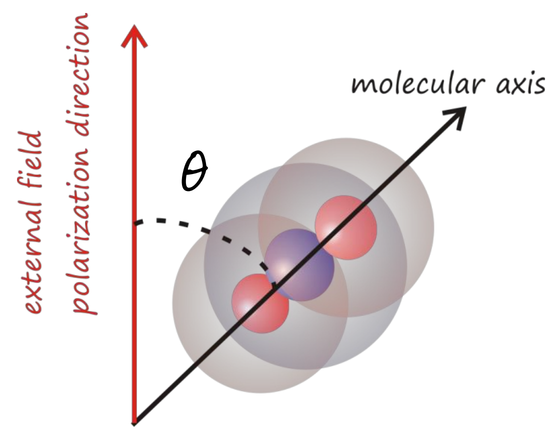
molecule in intense ultrashort laser field:
pulse duration $\tau < T_R$
intensity $I > 10^{12}$ W/cm^2



coherent excitation of rotational wave-packet
→ rotational revivals

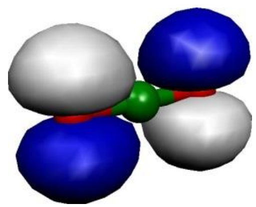
→ field-free alignment of the molecular sample for certain delays!

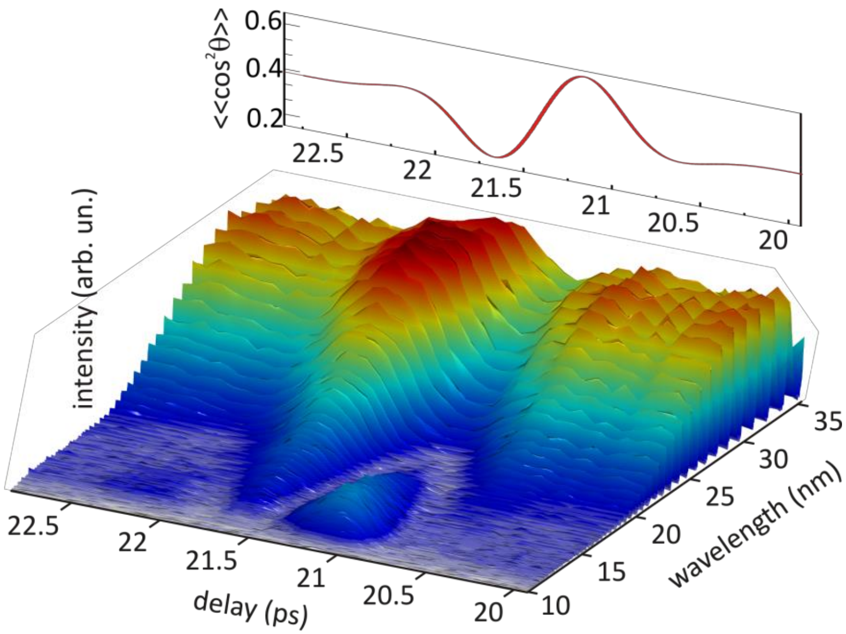




J. Ortigoso *et al.*, J. Chem. Phys. 110, 3870 (1999)

CO_2 orbital tomography

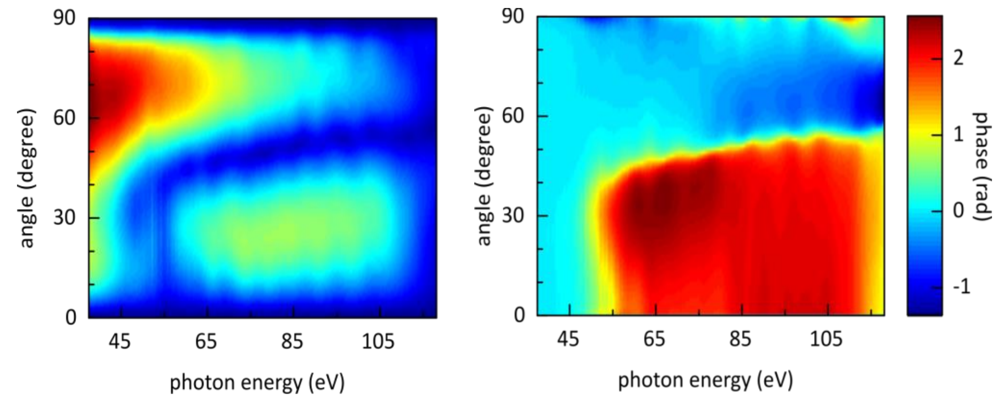




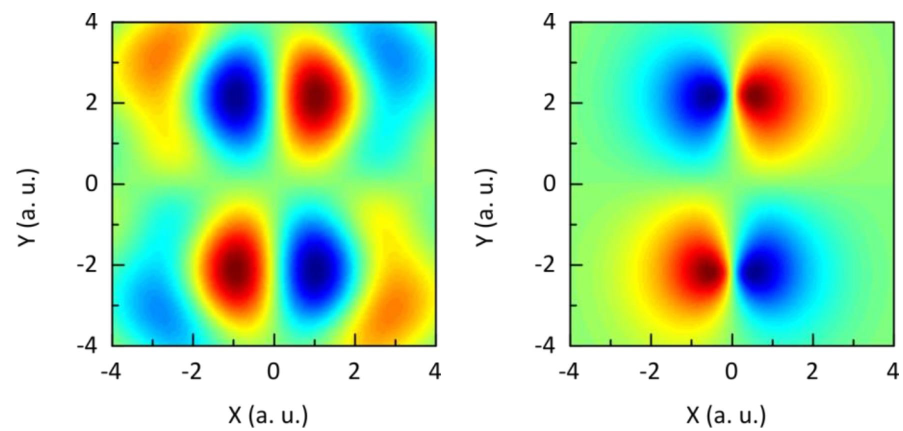
Nat. Phys. 7, 822 (2011)

amplitude

phase

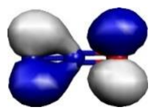


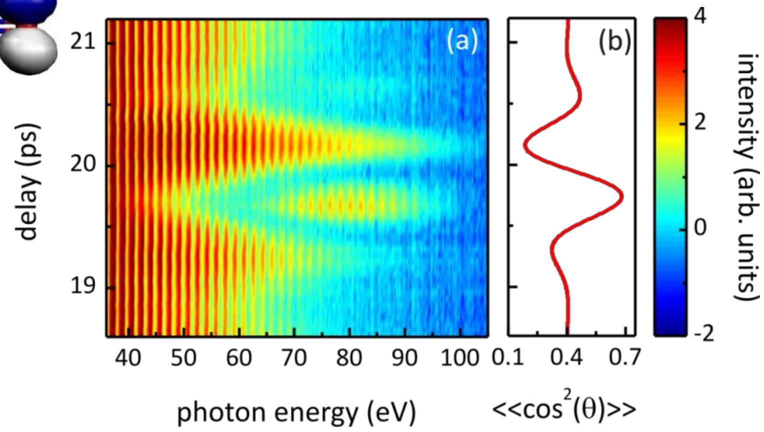
2D projection of CO_2 HOMO

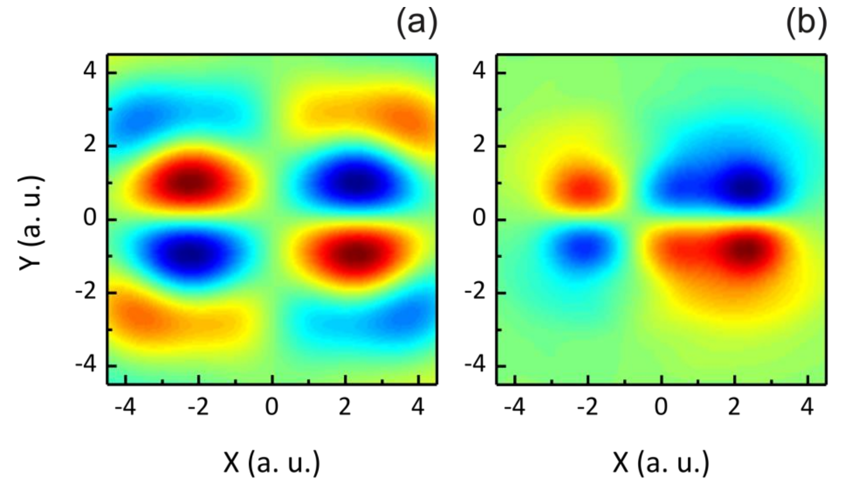


«more complex» molecules

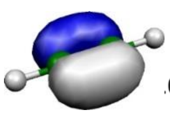
N_2O

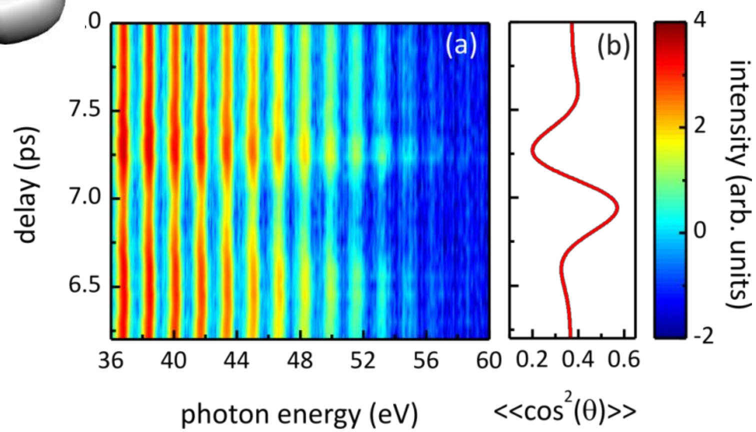


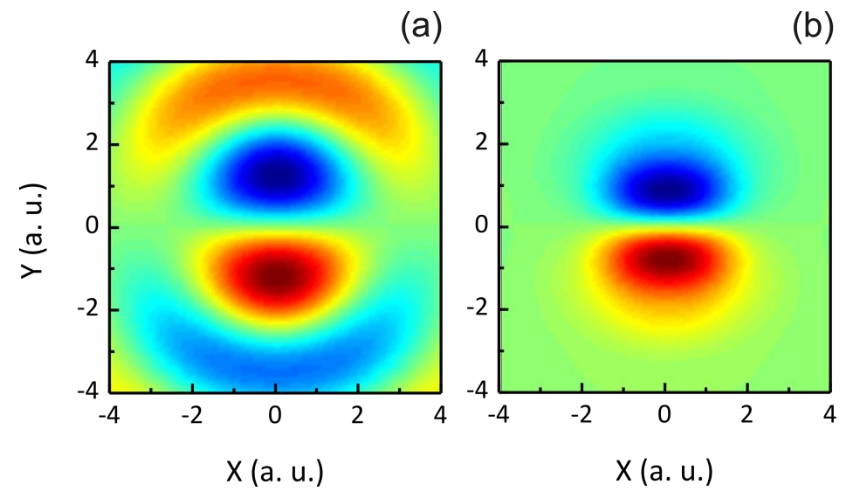




C_2H_2







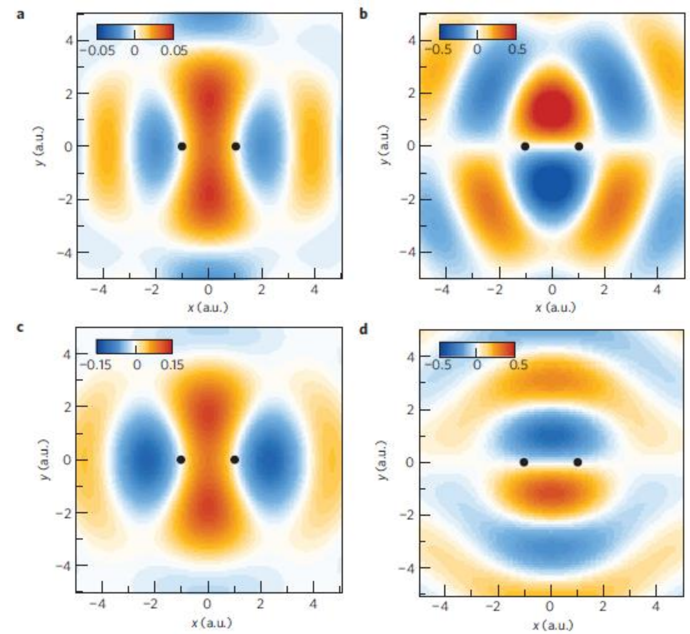
M. Negro et al., Faraday Disc 171., 2014

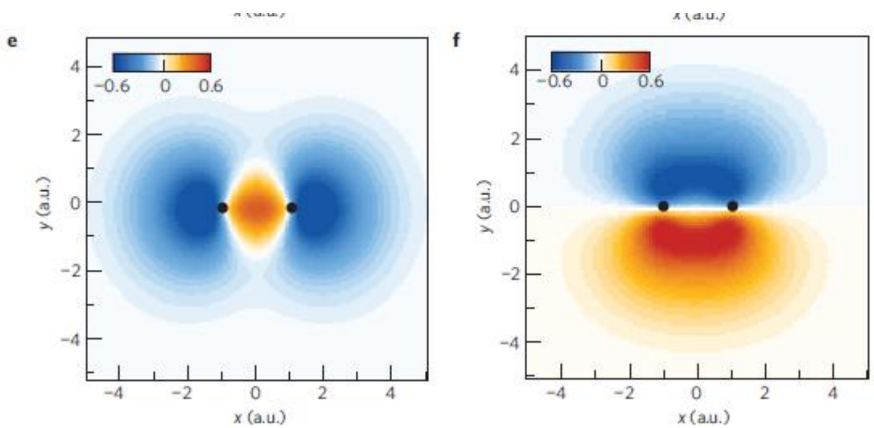
CNR IFN Istituto di Fotonica e Nanotecnologie UDynI

Multielectron dynamics

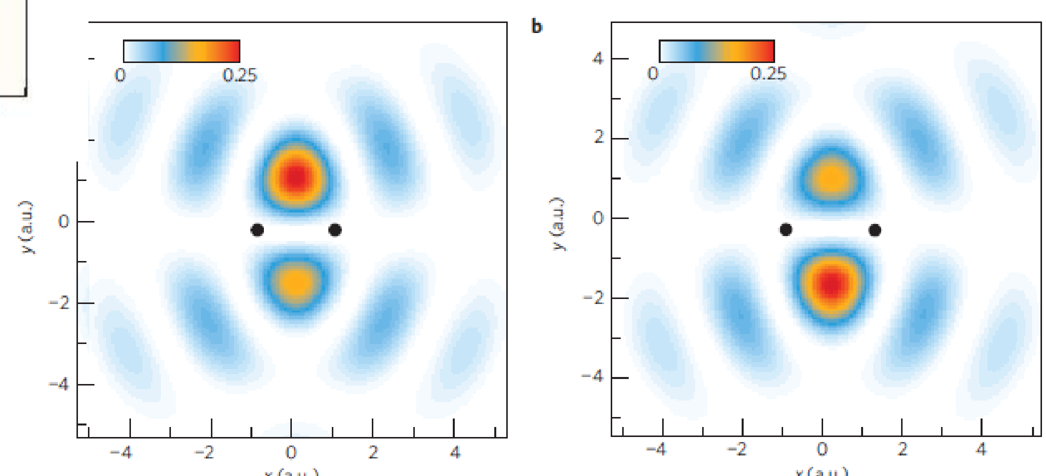
tunnel ionization from several valence orbital:

N_2 HOMO π-symmetry
HOMO-1 σ-symmetry
$\Delta E = -1.4$ eV





Reconstruction of the hole dynamics!



S. Haessler et al., Nat. Phys. 6, 200 (2010)

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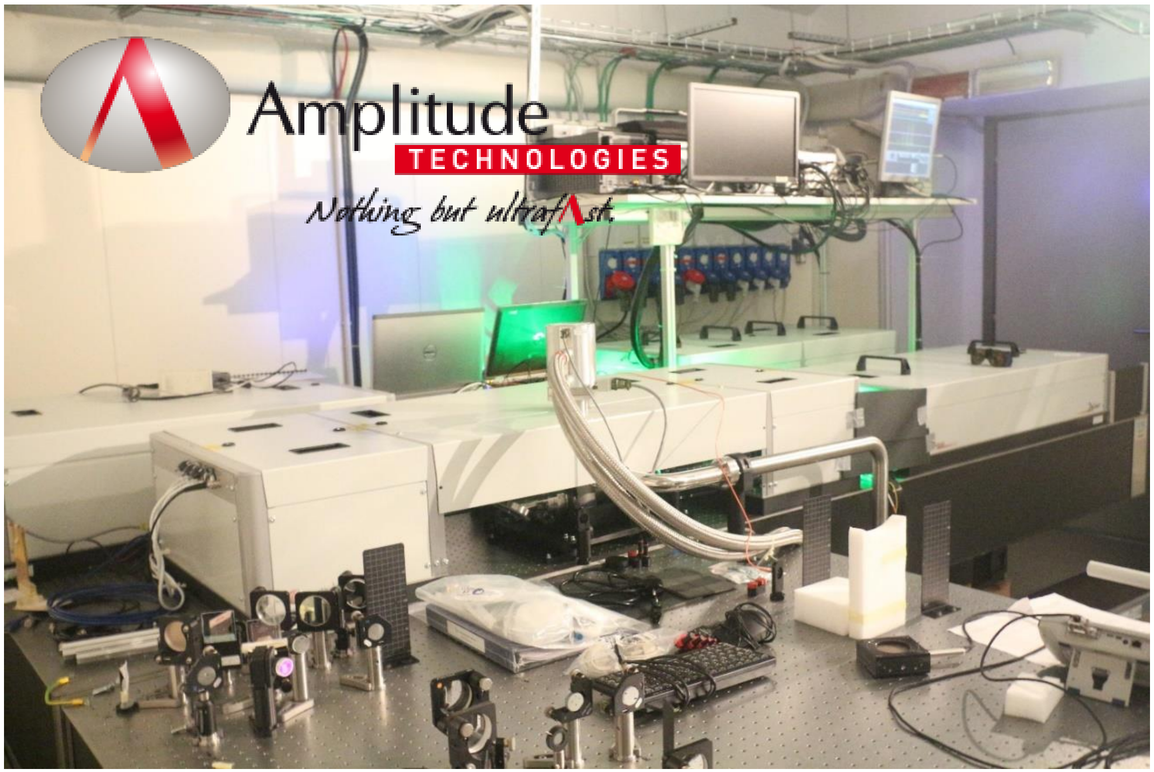
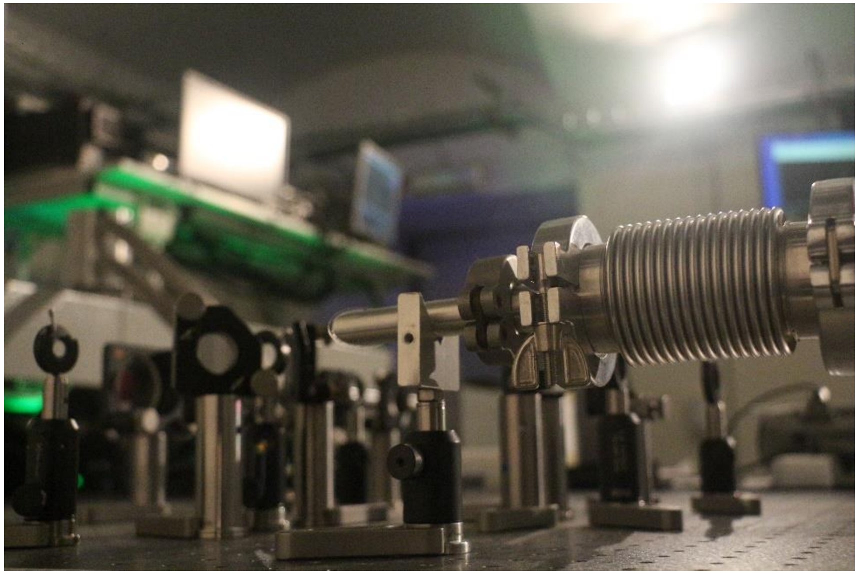
A new lab @ CNR-IFN

the laser source

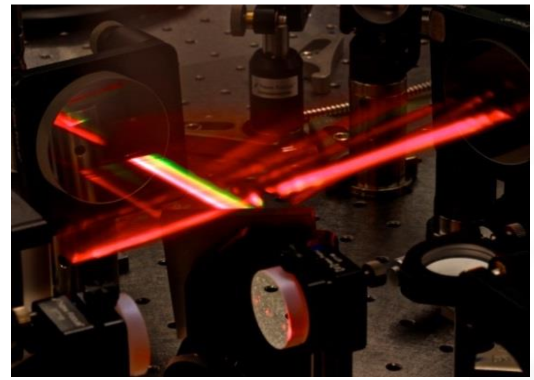


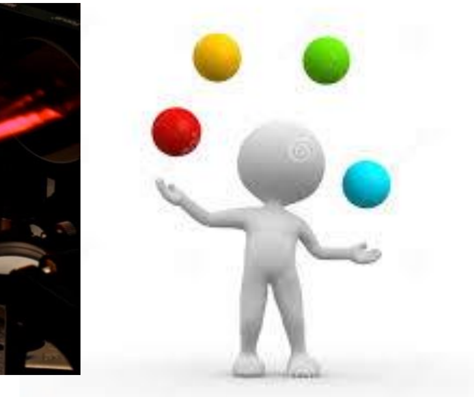
Driving laser source:
22 fs pulses
15 mJ energy
1 kHz rep rate

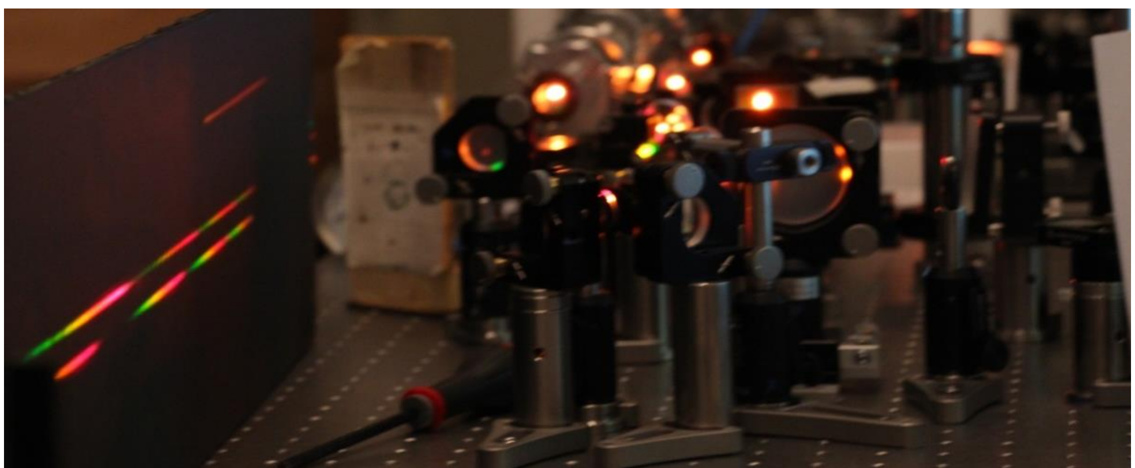
The Udyni lab @ CNR-IFN



manipulating the light







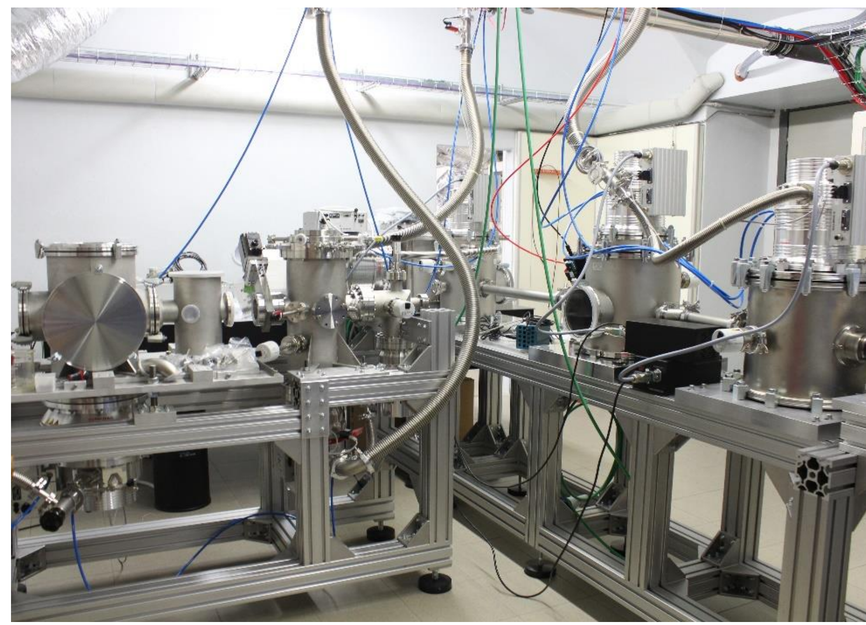
High energy OPA
<15 fs pulses
2 mJ energy
1 kHz rep rate

+ hollow fiber compression

The Udyni lab @ CNR-IFN

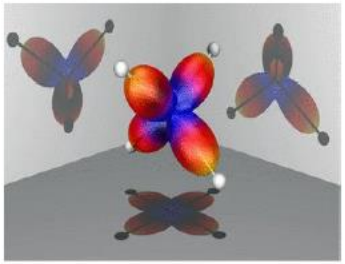
XUV spectrometer
- 80-1 nm spectral range
- stigmatic/astigmatic
- harmonic polarization detection
- large dynamical range

VMI spectrometer
for electrons
up to 200 eV



Perspectives

- time-resolved molecular orbital tomography
- time-resolved Laser Induced Electron Diffraction
- transient absorption spectroscopy
- harmonic polarimetry



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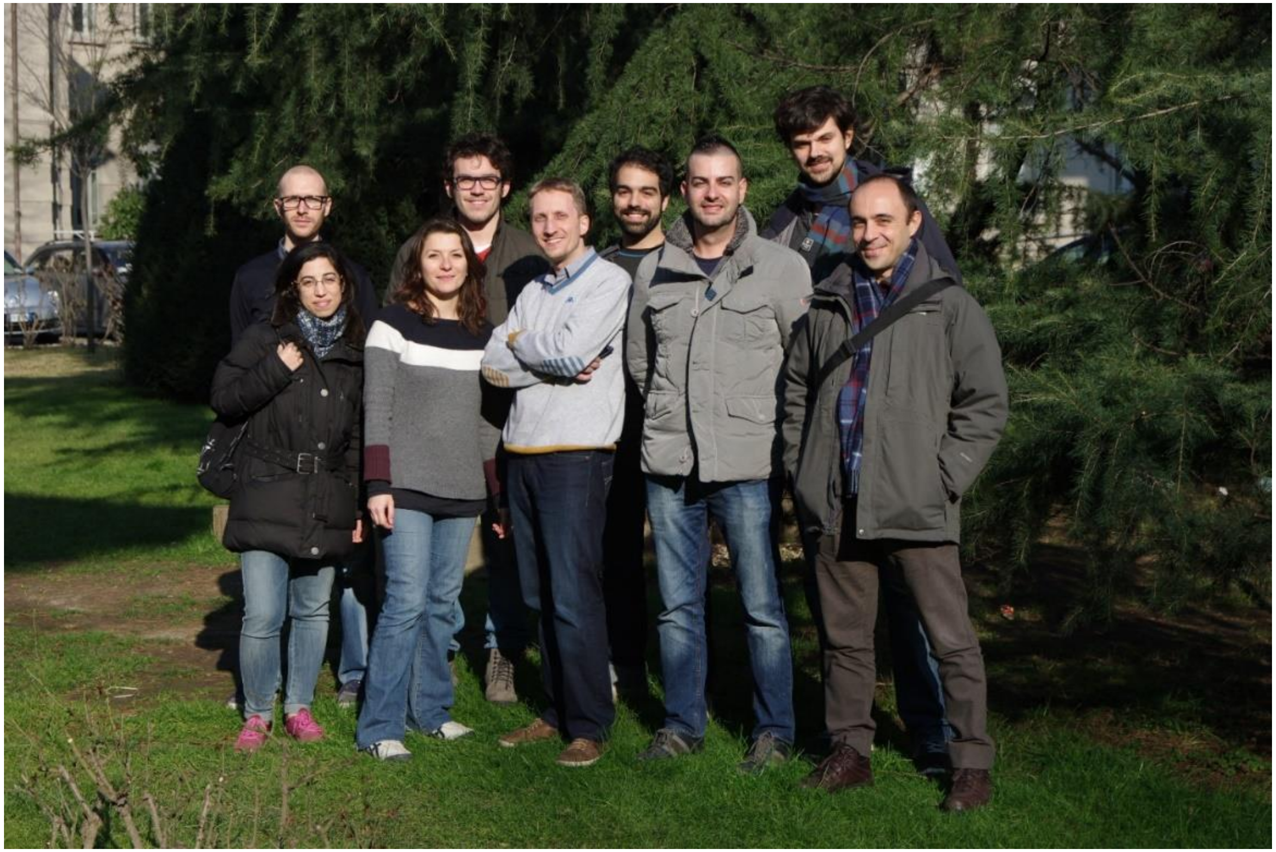

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