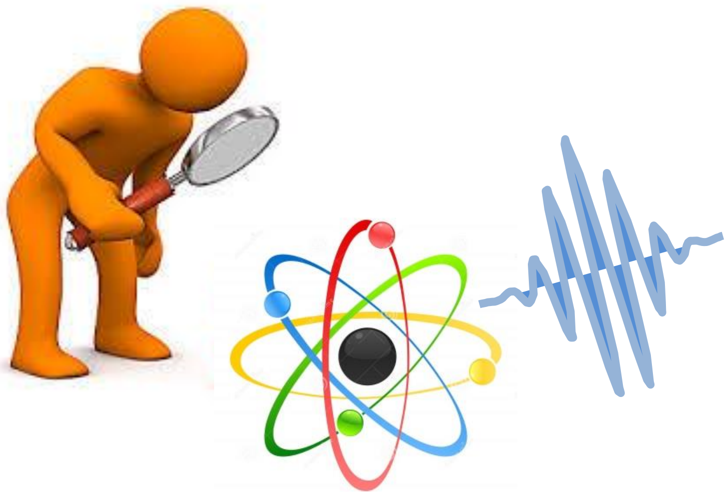
Looking into the ultrafast dynamics of electrons



G. Sansone[1,2,3]

1) Dipartimento di Fisica Politecnico Milano, Italy
2) Institute of Photonics and Nanotechnology, CNR Politecnico Milano Italy
3) Extreme Light Infrastructure Attosecond Light Pulse Source (ELI-ALPS) Szeged Hungary

Moscow, 24th October 2014

POLITECNICO MILANO

eli attosecond

Consiglio Nazionale delle Ricerche

Politecnico Milano, Italy

Founded in 1863

POLITECNICO DI MILANO

150°

EXPO

MILANO 2015

NUTRIRE IL PIANETA
ENERGIA PER LA VITA

Why and how ultrafast?

Quantum mechanics: observables

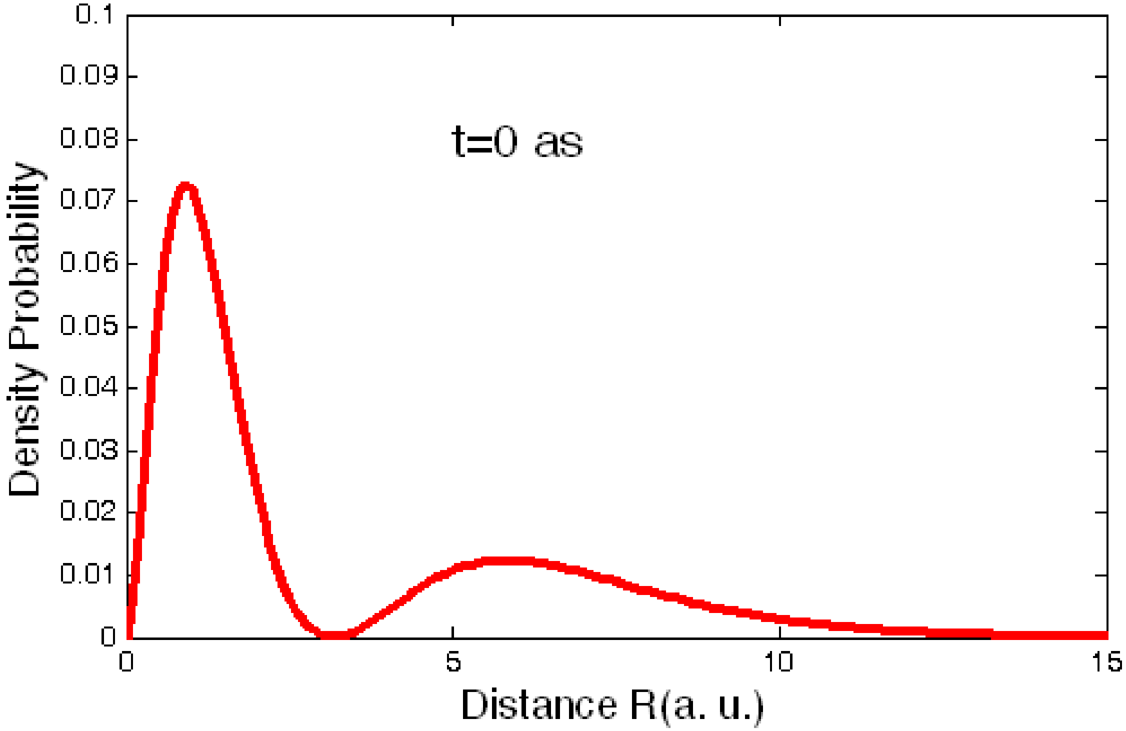
$$r(t) = \langle \psi(t) | r | \psi(t) \rangle$$

Dynamics: superposition of eigenstates

$$\psi(r,t) = \sum_n c_n(t) \psi_n(r)$$

where

$$H\psi_n(r) = E_n \psi_n(r)$$



1s-2s coherent superposition in hydrogen

Why and how ultrafast?

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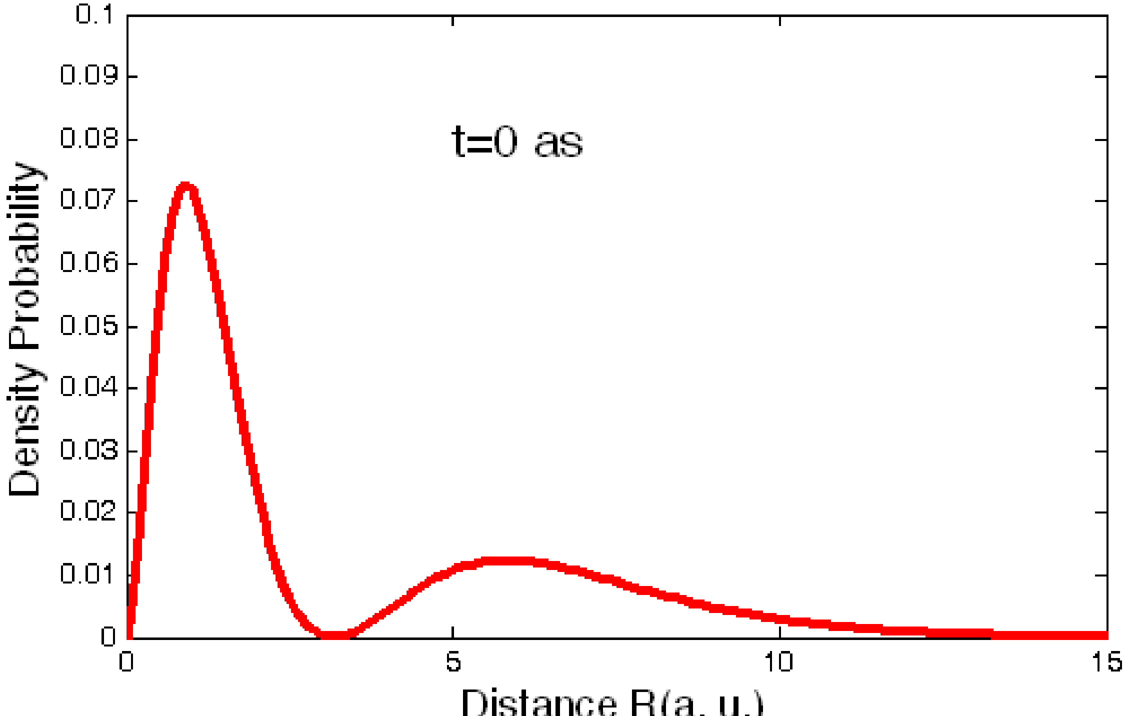


1s-2s coherent superposition in hydrogen

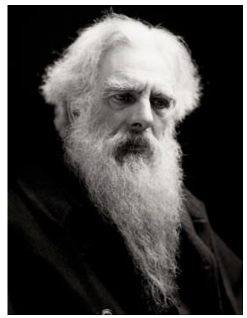
$T \approx 402$ as

Attosecond timescale

Ultrafast stopwatch

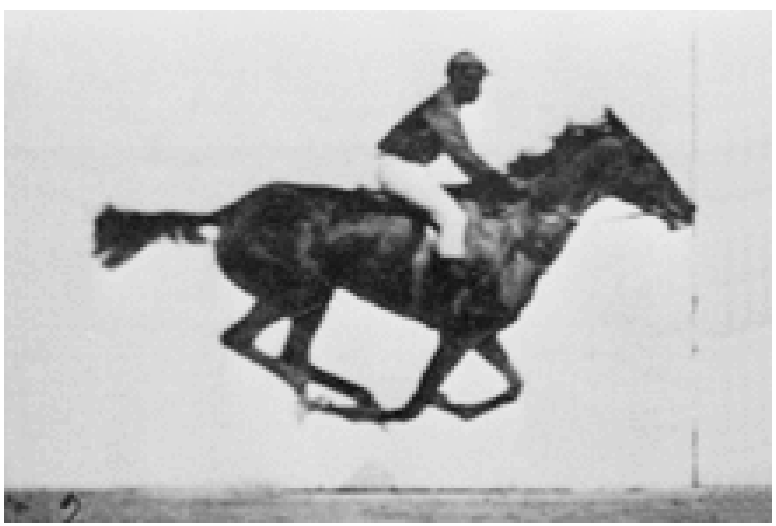
From milliseconds (10^{-3} s)

Eadweard James Muybridge



After Stanford's request 1878

"whether all four feet of a horse were off the ground at the same time during a gallop"

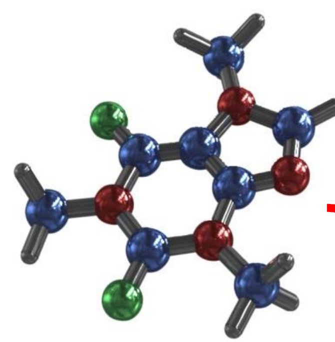


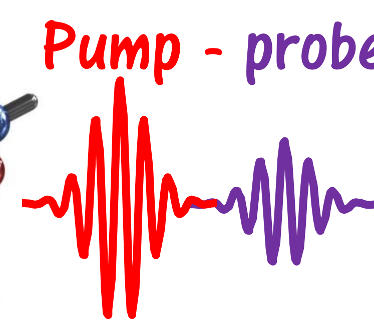
To femtoseconds (10^{-15} s)

Ahmed H. Zewail

The Nobel prize in Chemistry 1999

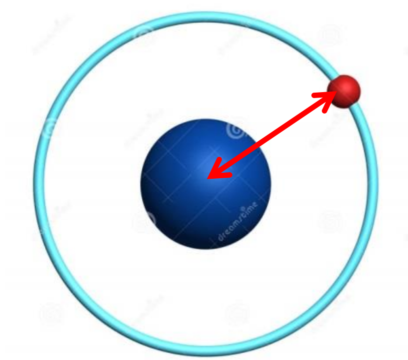
"for his studies of the transition states of chemical reactions using femtosecond spectroscopy"





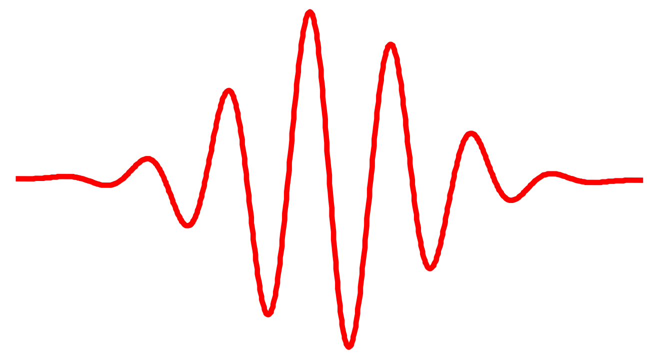
Attosecond domain: atoms in intense femtosecond laser fields

Coulomb field *vs* *Laser field*



$E=5.14x10^{11}$ V/m

Atomic unit of electric field



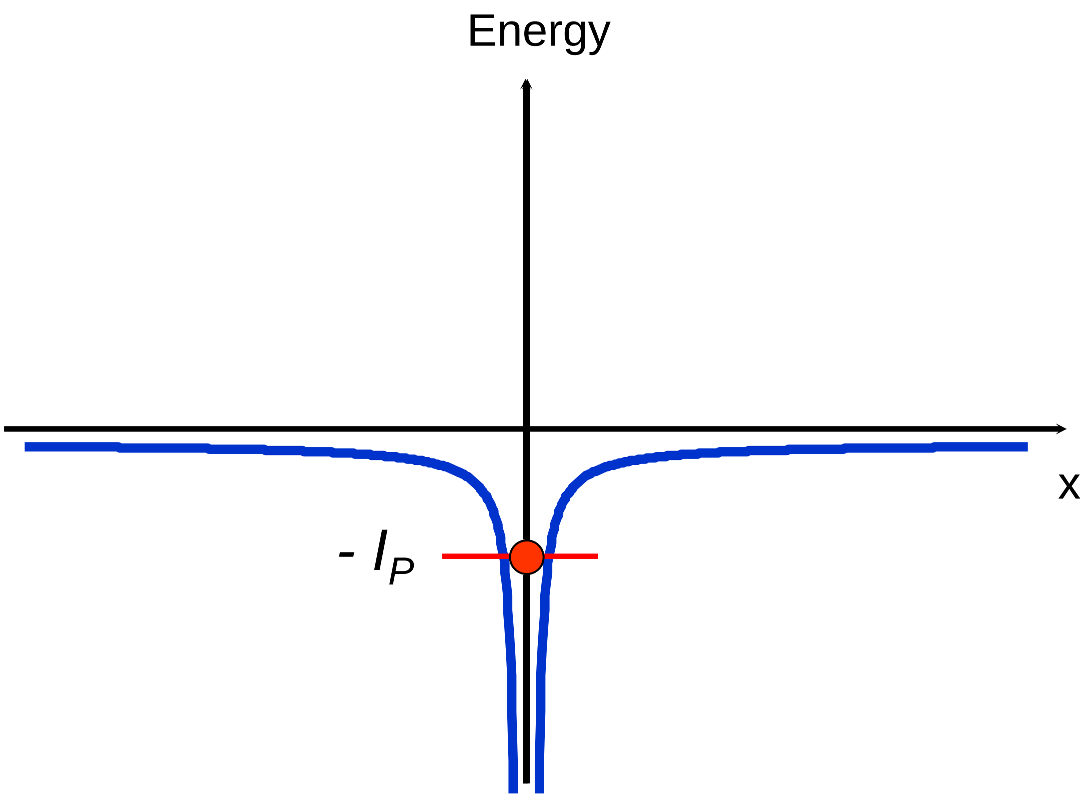
$I=10^{15}$ W/cm^2

$E=8.6 \times 10^{10}$ V/m

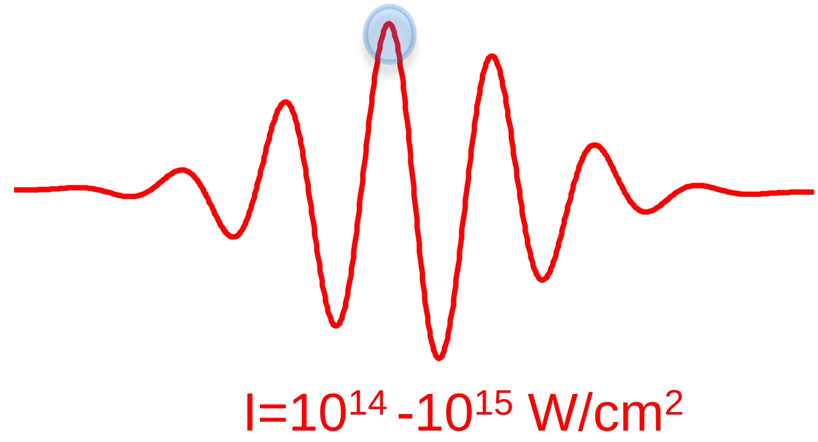
- **Extreme nonlinear optics: processes dependent on the electric field**

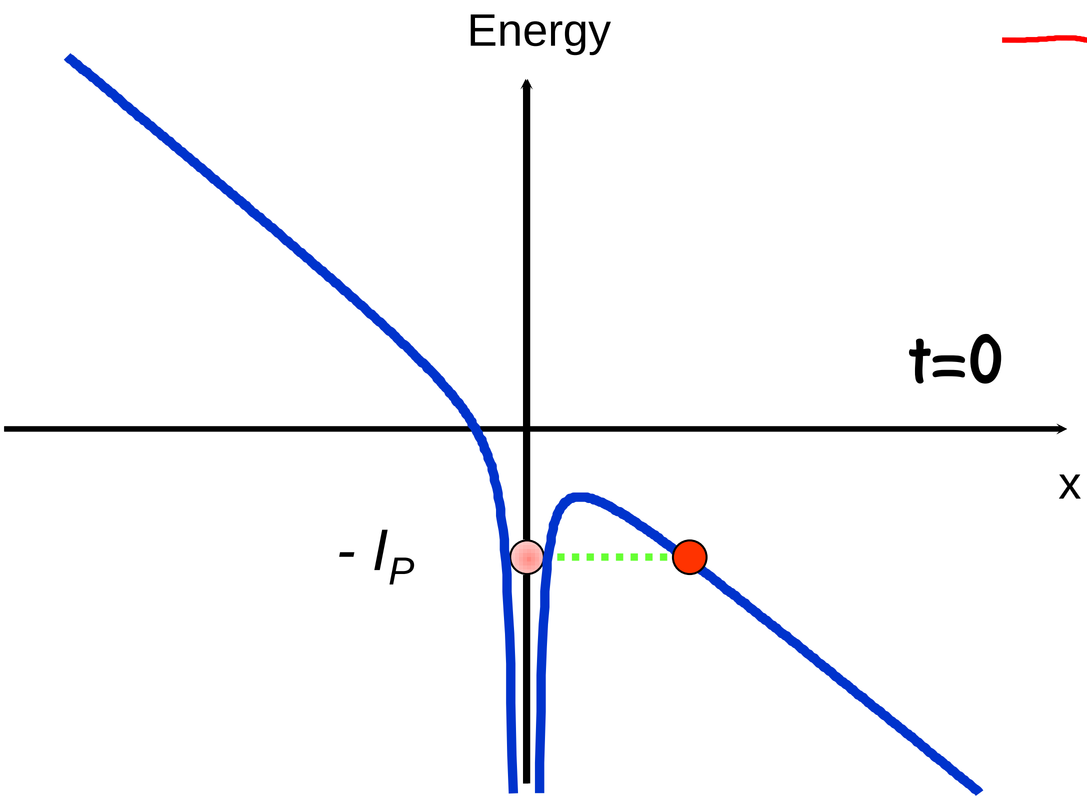
Above threshold ionization (ATI)   *Laser induced electron diffration (LIED)*
Non-sequential double ionization (NSDI)   ***High-order harmonic generation (HHG)***

Atoms in intense laser fields



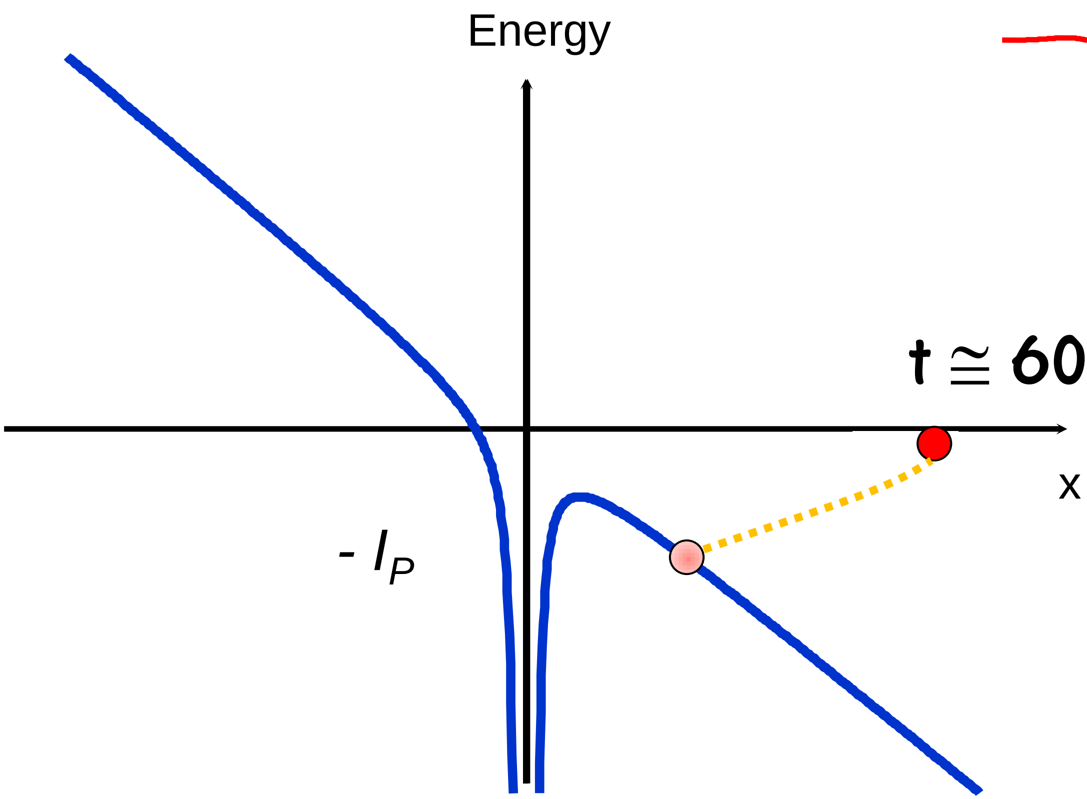
Atoms in intense laser fields

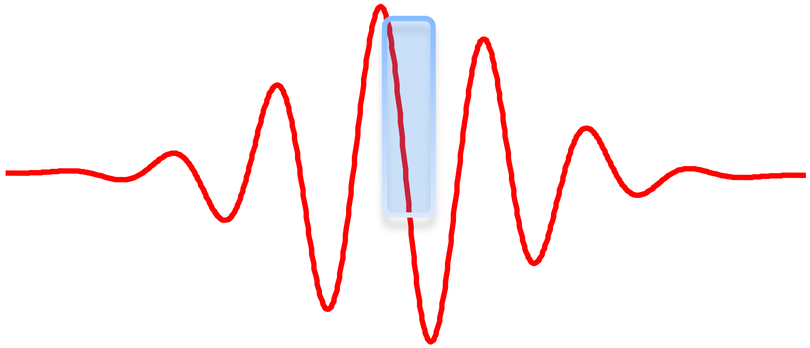




1) Tunnel ionization

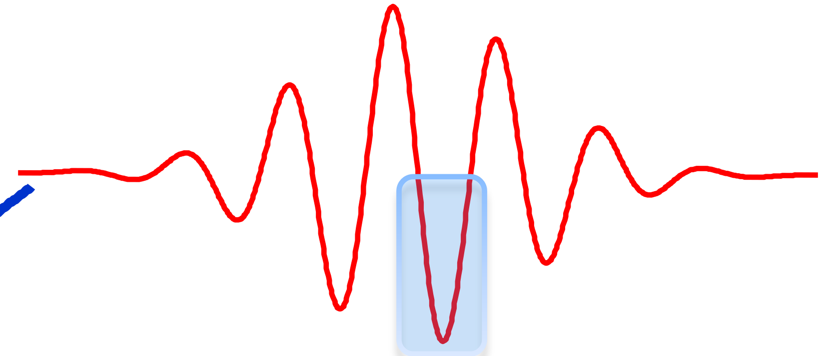
Atoms in intense laser fields

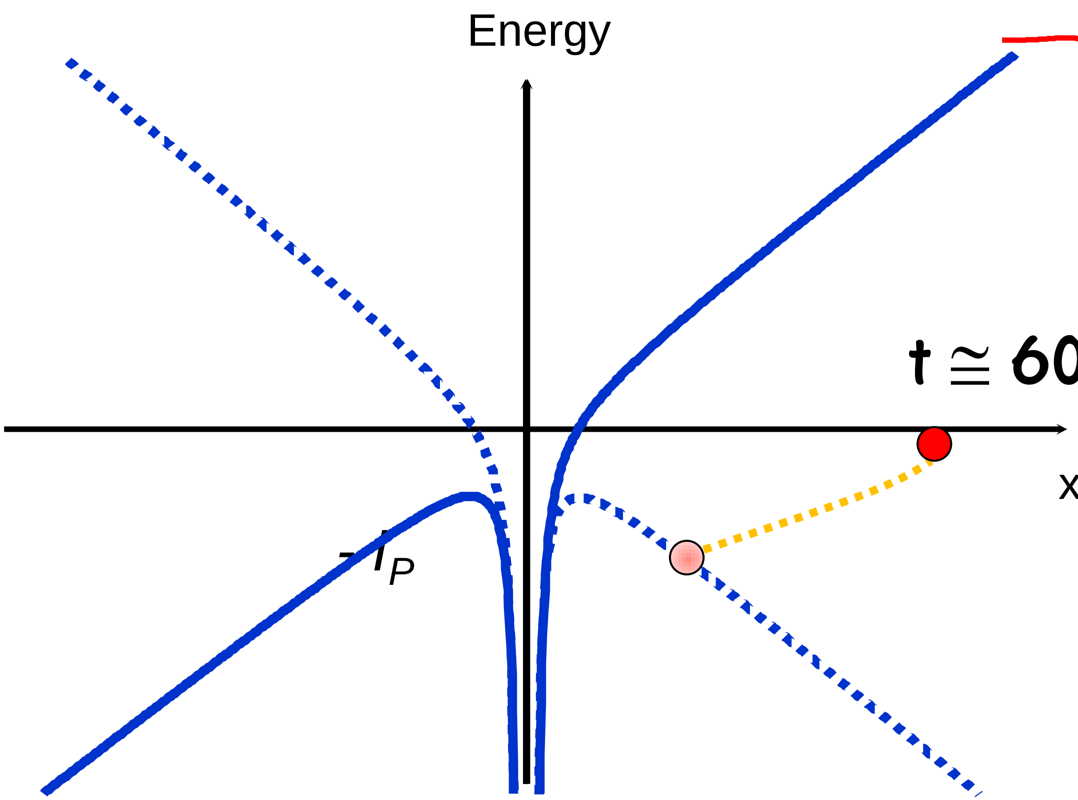




2) Acceleration in the continuum

Atoms in intense laser fields

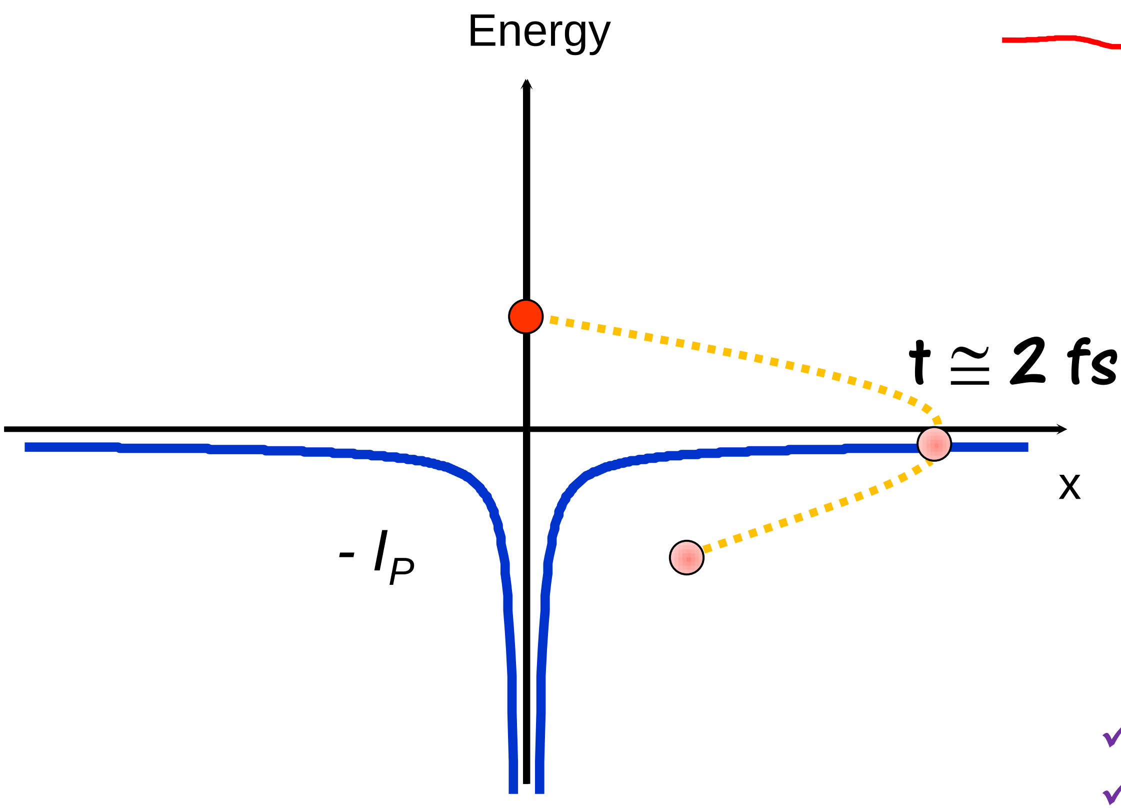


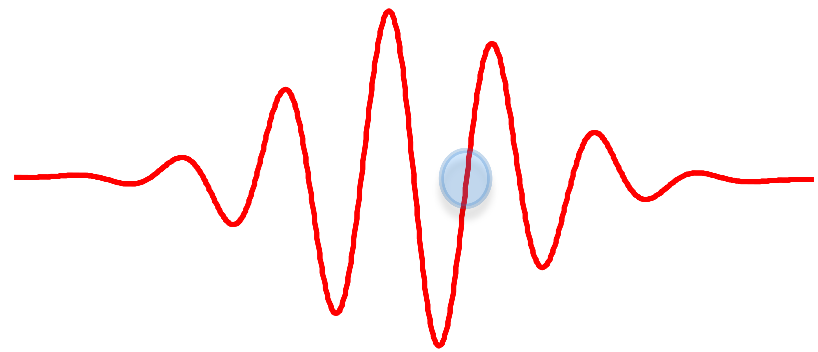


t ≅ 600 as

2) Acceleration in the continuum

Atoms in intense laser fields

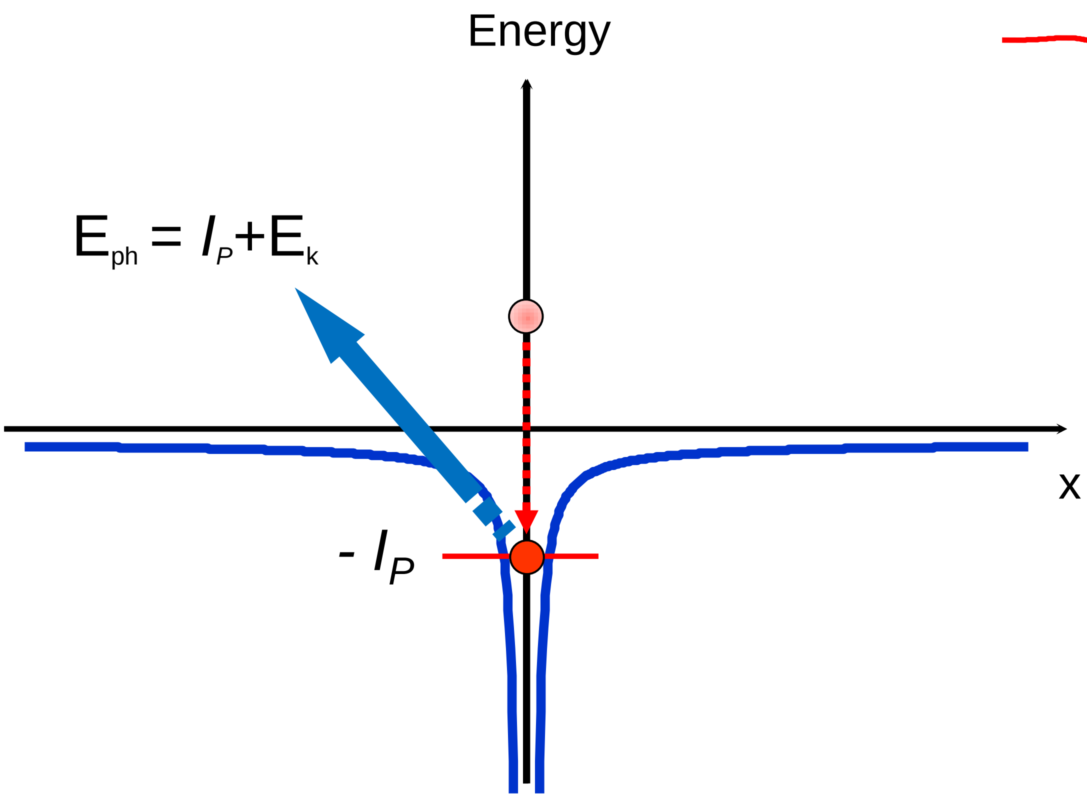


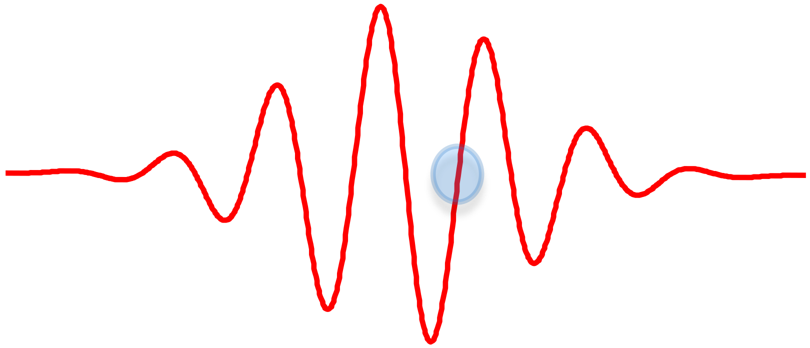


3) Recollision

- ✓ *Diffraction*
- ✓ *Nonsequential double ionization*

High-order harmonic (HHG) generation





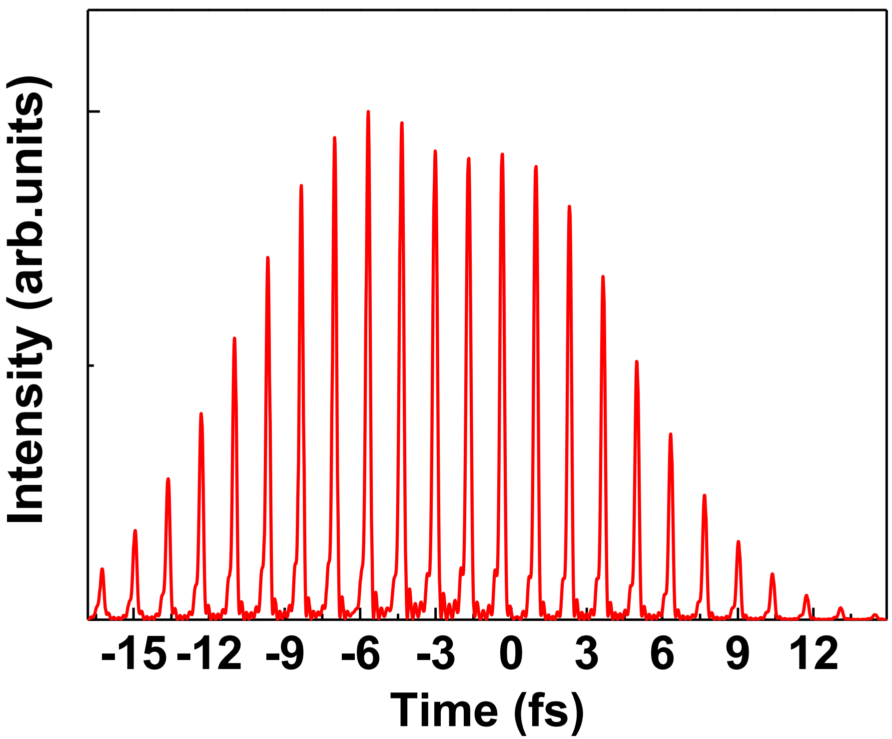
I=10^{14} -10^{15} W/cm^2

3) Recollision

✓ ***High-order harmonic generation***

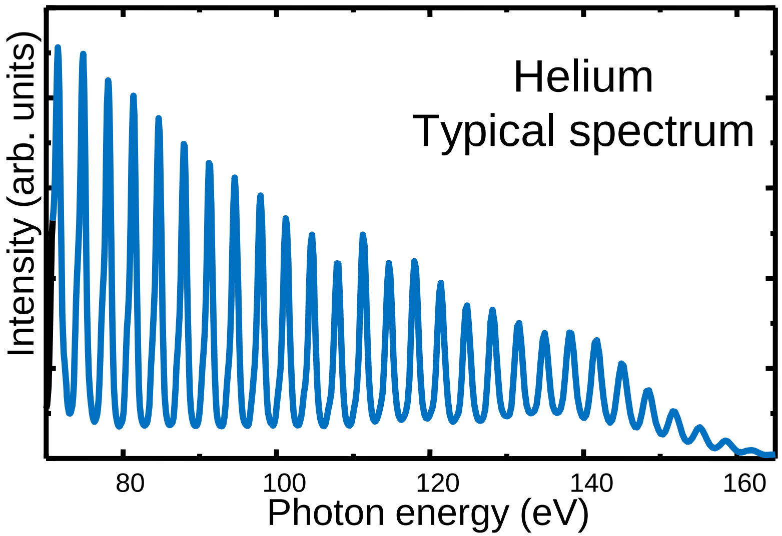
HHG: temporal and spectral domain

Temporal domain



Train of **attosecond pulses**

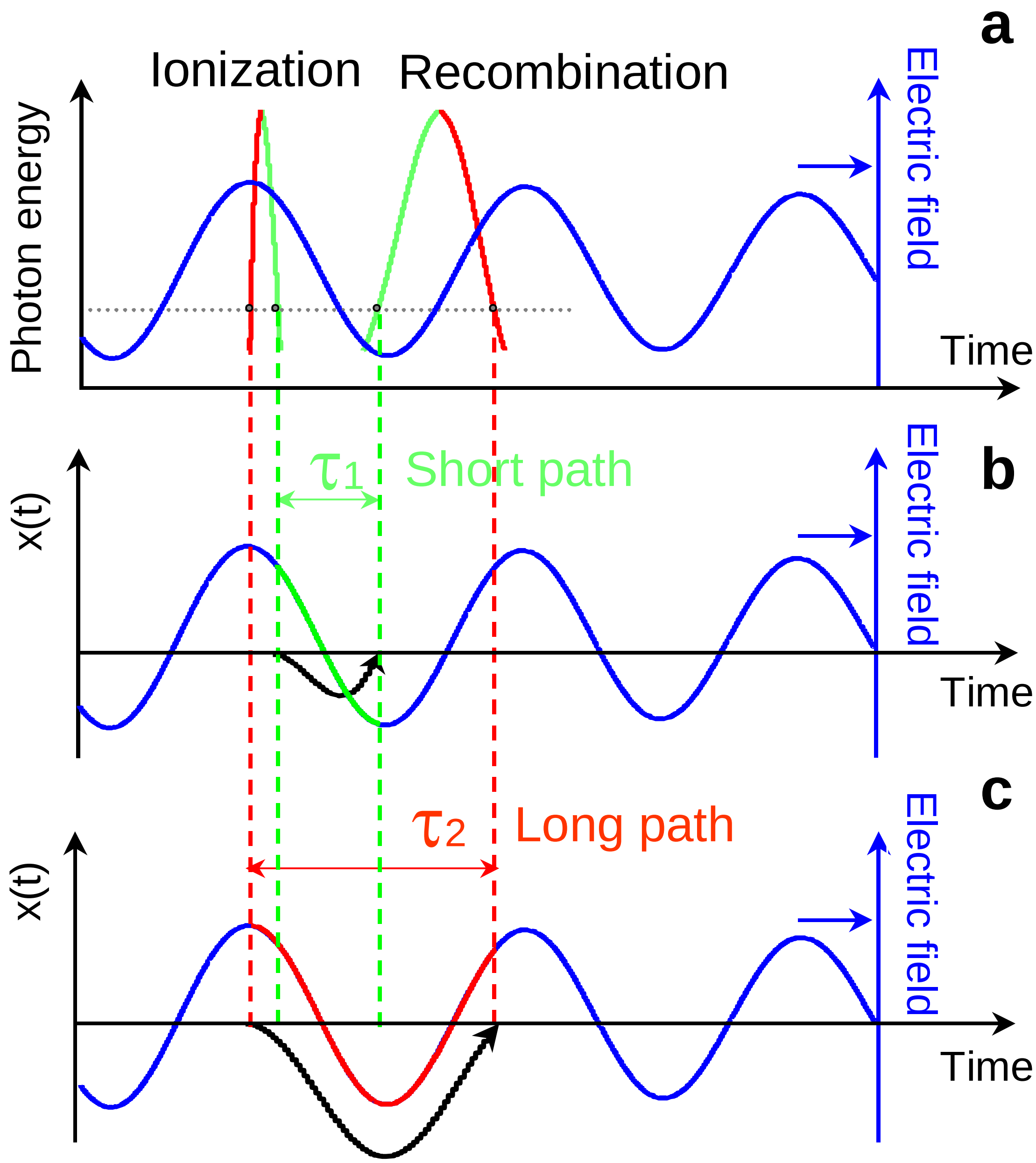
Spectral domain



*Emission of the **odd harmonics** of the fundamental frequency*

How can we obtain an *isolated* attosecond pulse?

Quantum trajectories: short and long paths

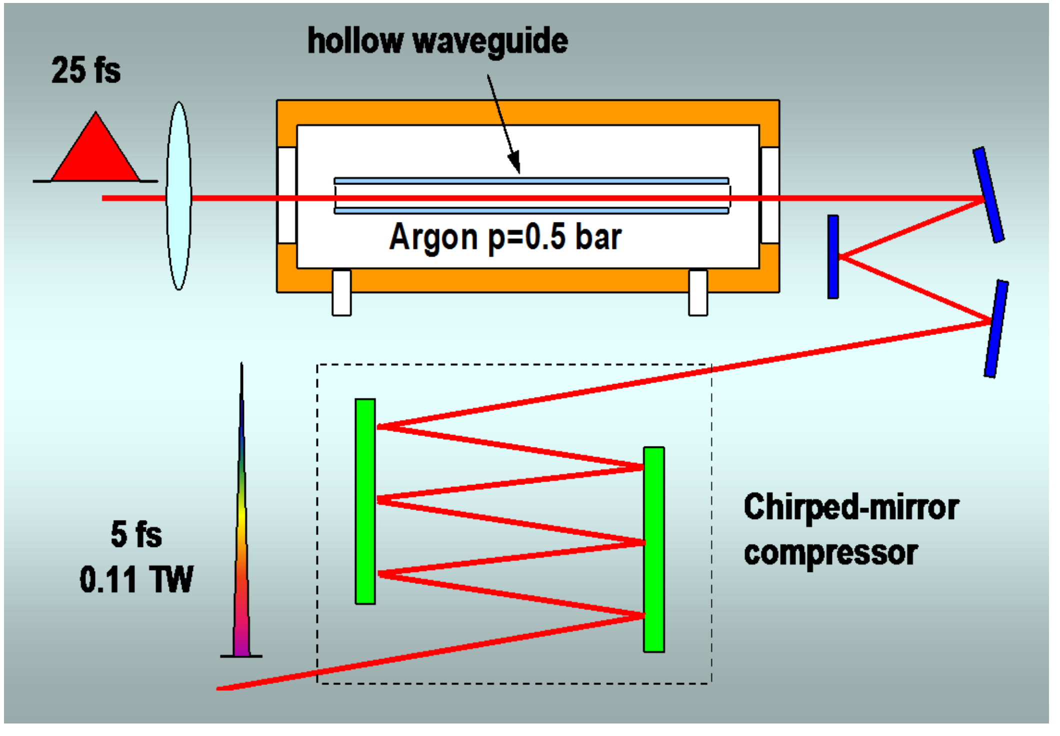


Phase-matching condition
(macroscopic effect)
select the short path contribution

How can we isolate a single attosecond pulse?

Few-cycle driving pulse for HHG

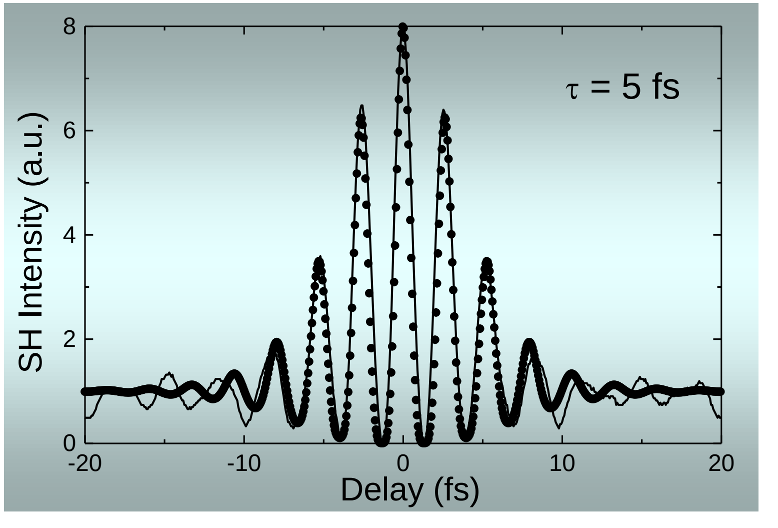
- ***Hollow-fiber compression technique***



⇒ Guiding medium with a large diameter mode and a fast nonlinear medium with high damage threshold

⇒ Ultrabroad-band dispersion control by chirped-mirrors

Interfermetric AutoCorrelation of ***5 fs*** *pulses*

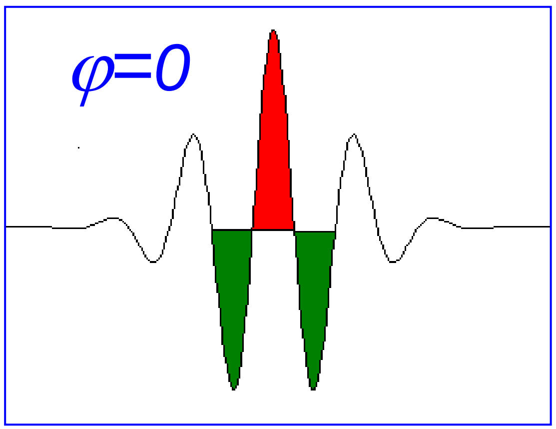


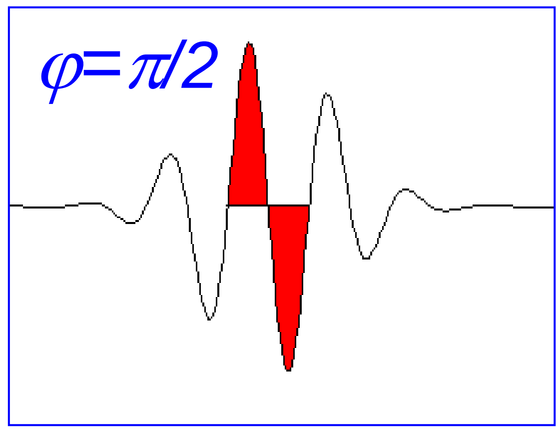
M. Nisoli *et al.*, Appl. Phys. Lett. **68**, 2793 (1996)
M. Nisoli *et al.*, Opt. Lett. **22**, 522 (1997)

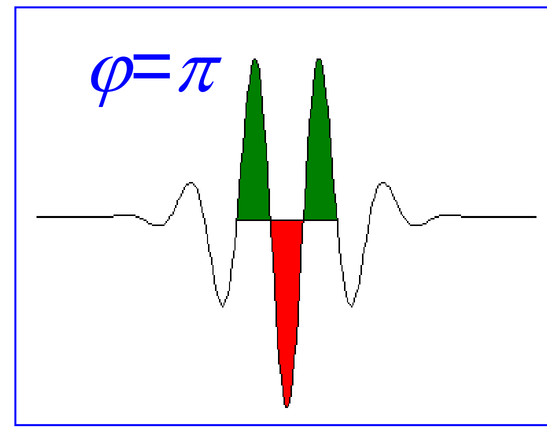
Few-cycle pulses… but

- **Time variation of the electric field of few-cycle pulses depends on the carrier envelope phase (CEP)** φ

$$E(t) = A(t)\cos(\omega t + \varphi)$$







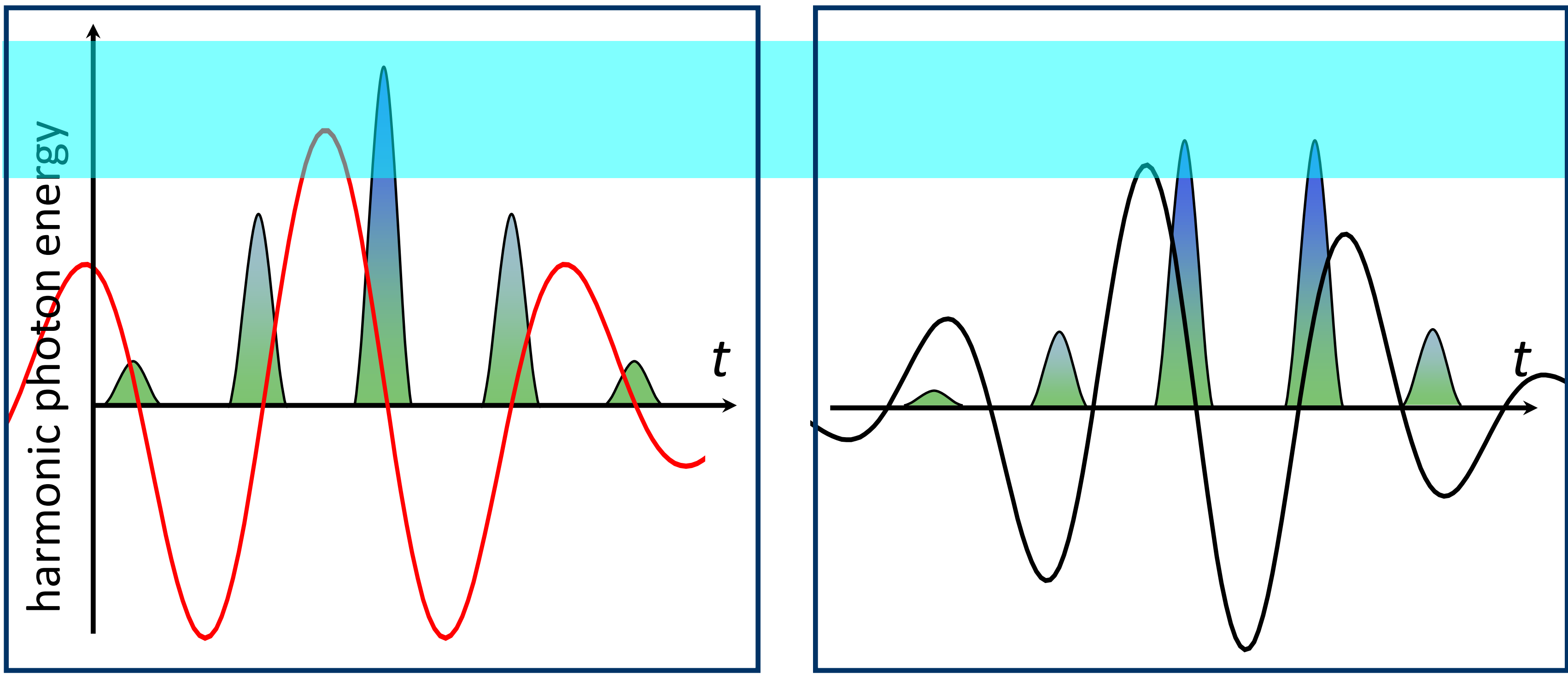
John L. Hall

Theodor W. Hänsch

Nobel Prize in Physics (2005)
"*for their contributions to the development of laser-based precision spectroscopy, including the optical frequency comb technique*".

Isolated attosecond pulses: spectral filter

$$E(t) = E_0 \cos(\omega_0 t) \qquad E(t) = E_0 \sin(\omega_0 t)$$

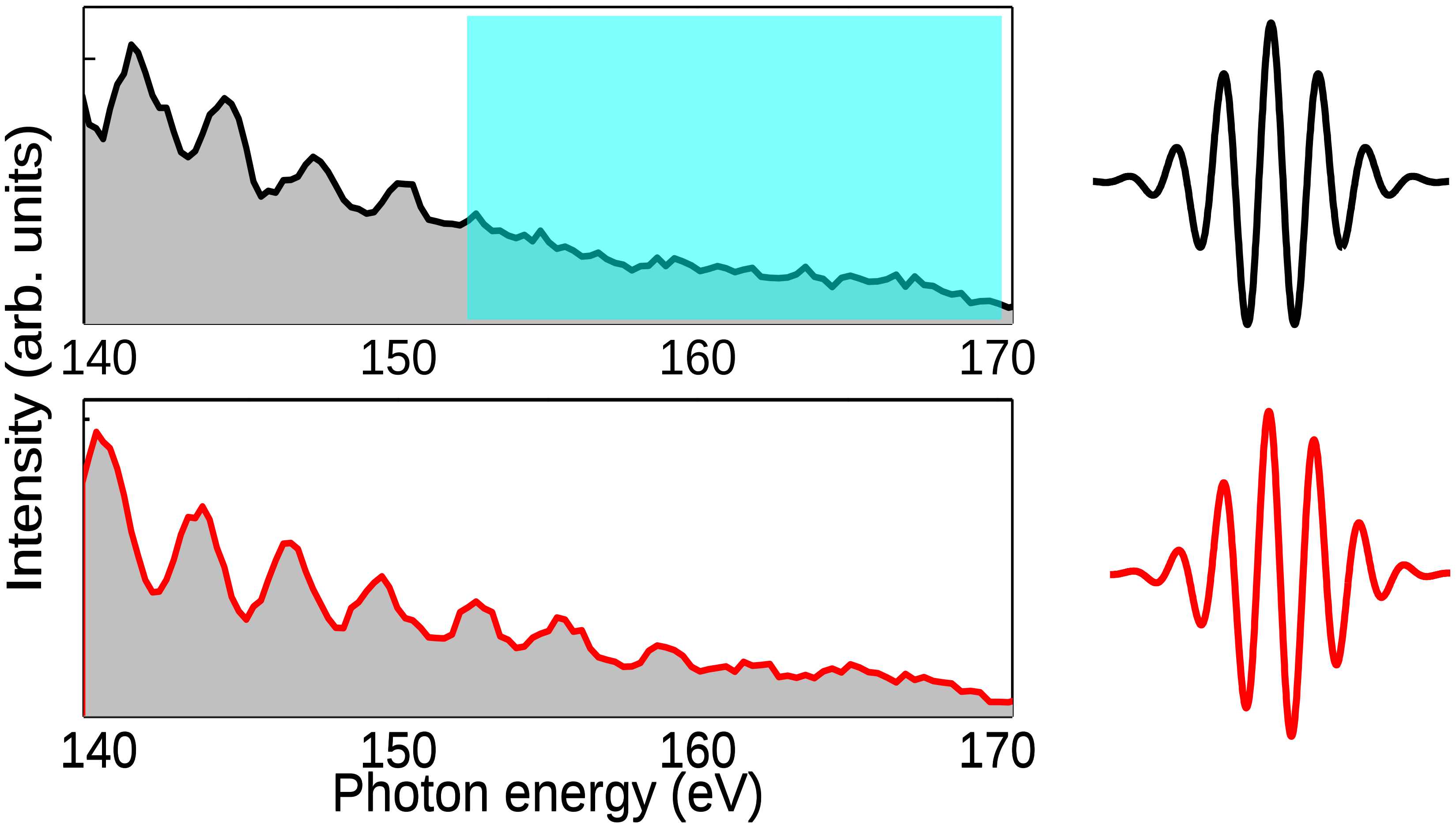


- ➔ **Spectral selection of cutoff photons leads to generation of one or two attosecond pulses**
- ➔ **Requirements: sub-5-fs phase-stabilized driving pulses (linear polarization)**

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

Few-cycle linearly polarized pulses

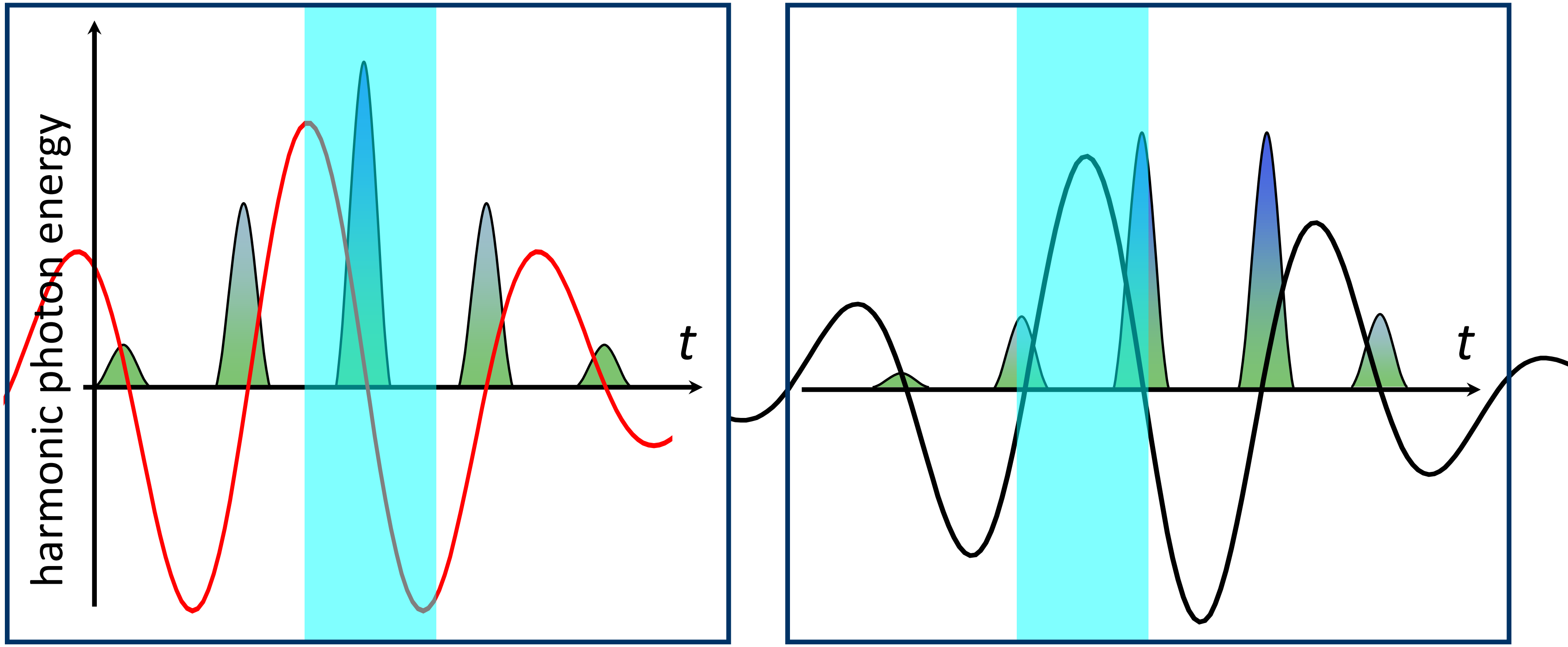
- HHG in Neon: < 5 fs; stabilized CEP



➔ **Broad continuum only in the cut-off**

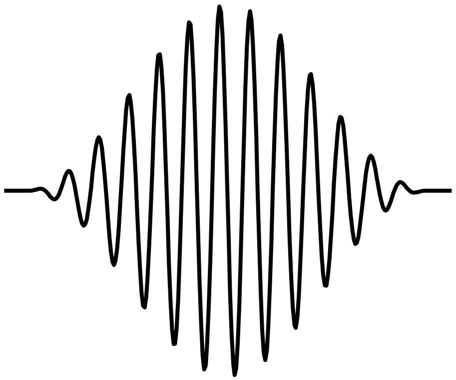
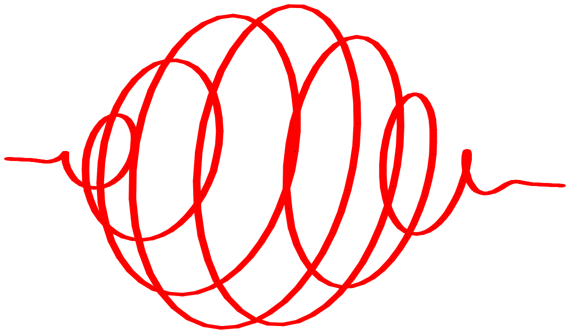
Isolated attosecond pulses: temporal filter

$$E(t) = E_0 \cos(\omega_0 t) \qquad E(t) = E_0 \sin(\omega_0 t)$$



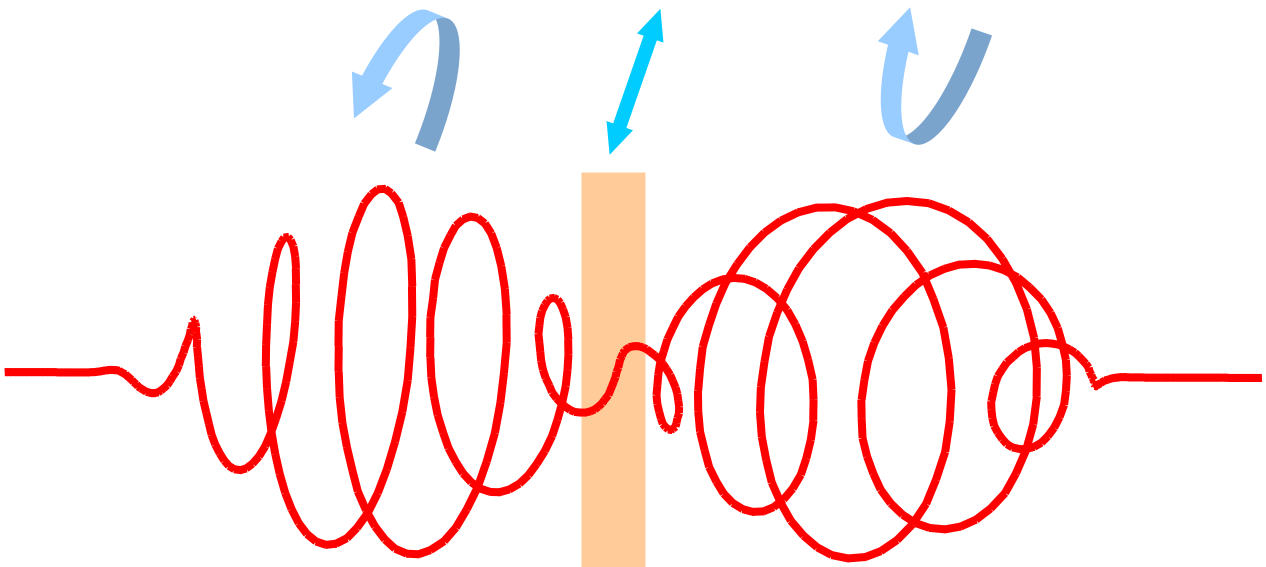
- ➔ **Temporal gating**
- ➔ **Requirements: phase-stabilized driving pulses**

HHG polarisation 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

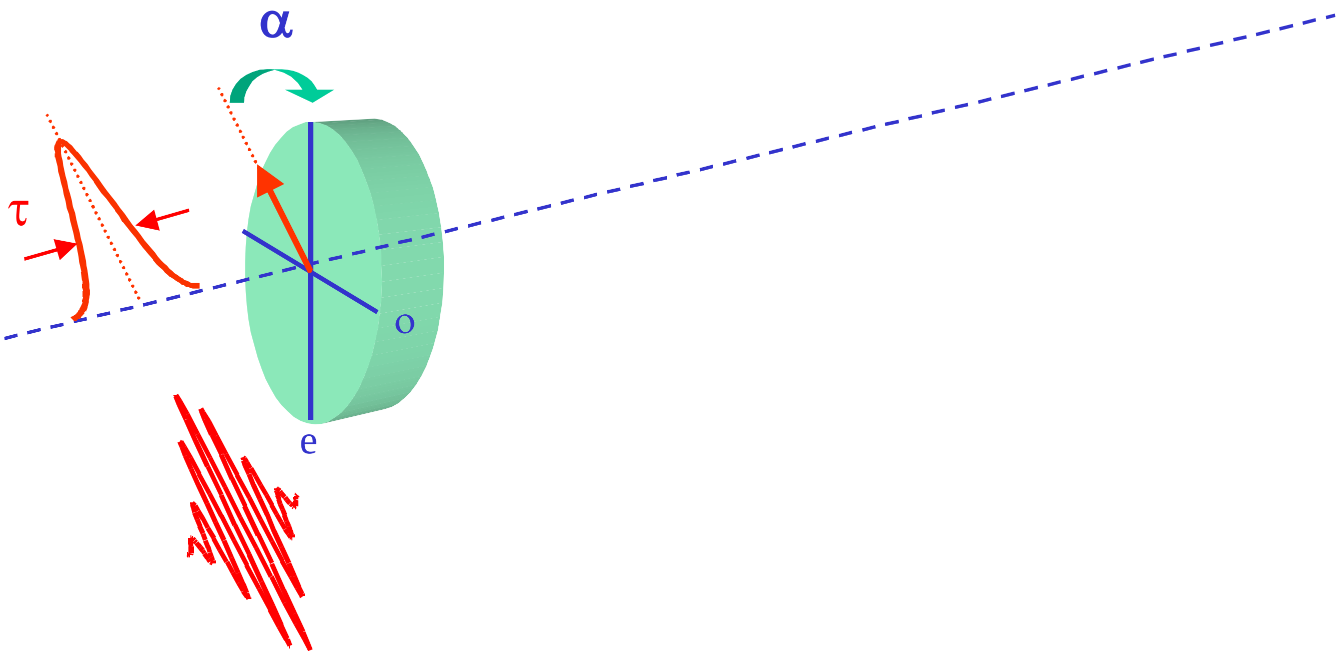
- Time-dependent polarization



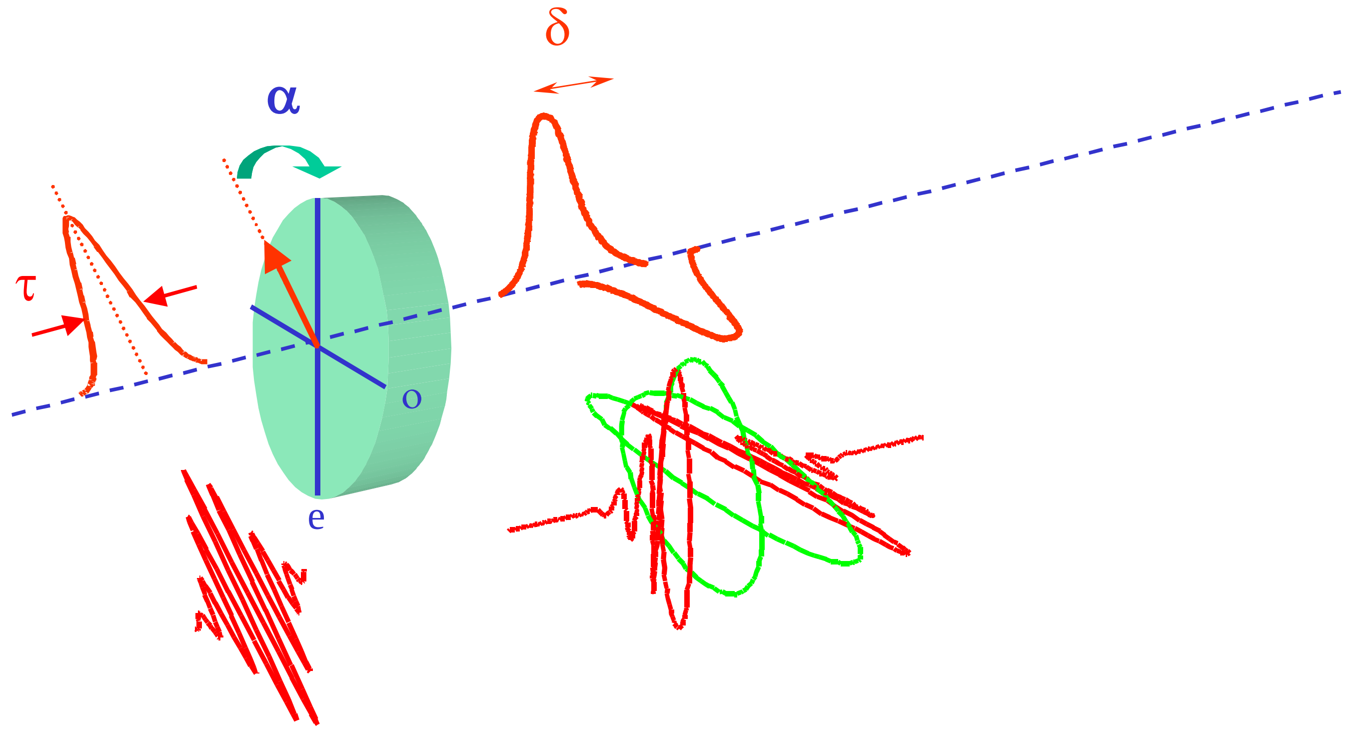
P. Corkum *et al.,* Opt. Lett. **19,** 1870 (1994)
O. Tcherbakoff *et al.,* Phys. Rev. A **68,** 043804 (2003)

Generation of XUV continuum with PG requires:
few-cycle pulses
CEP stabilization

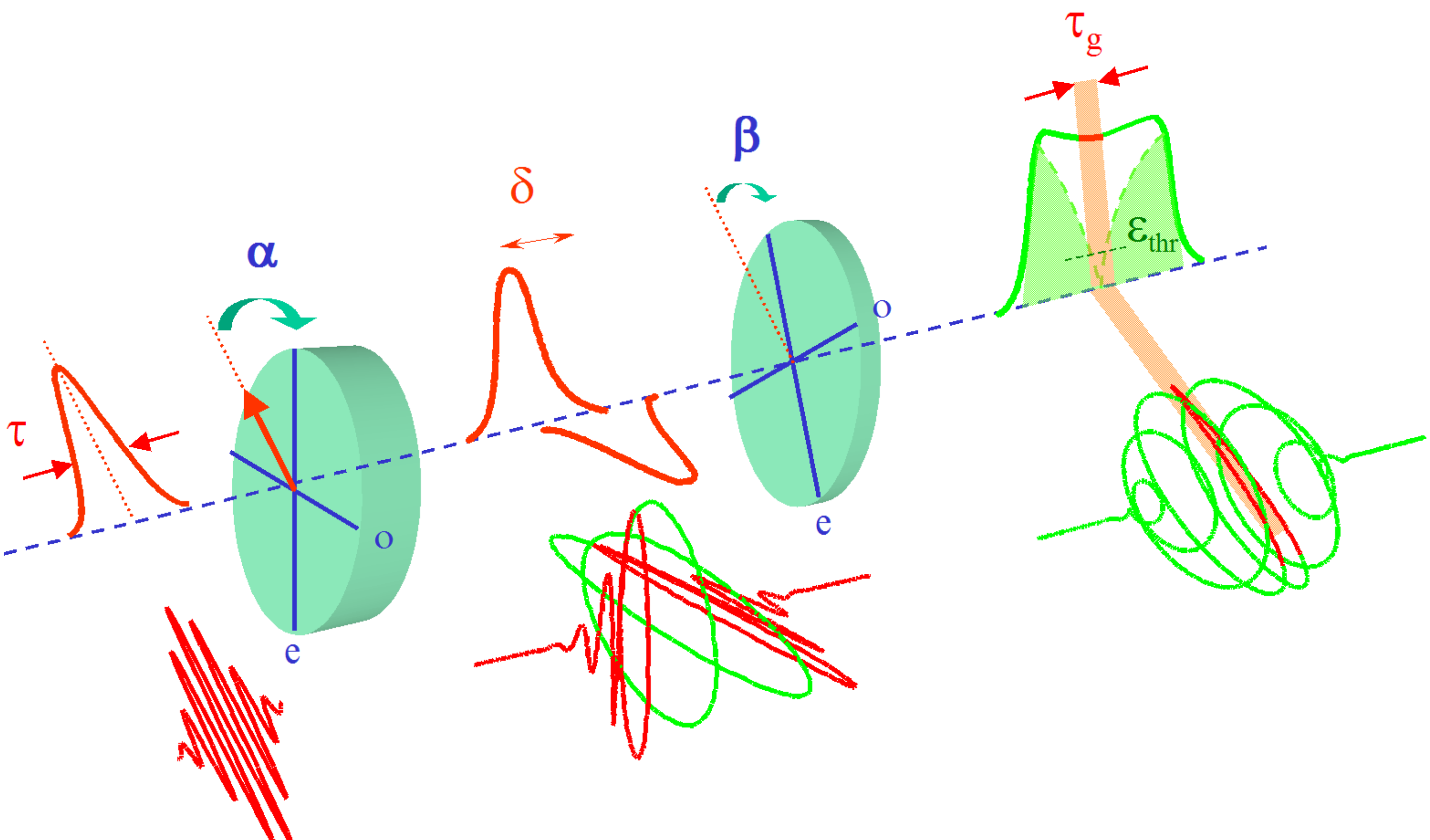
Polarization gating



Polarization gating

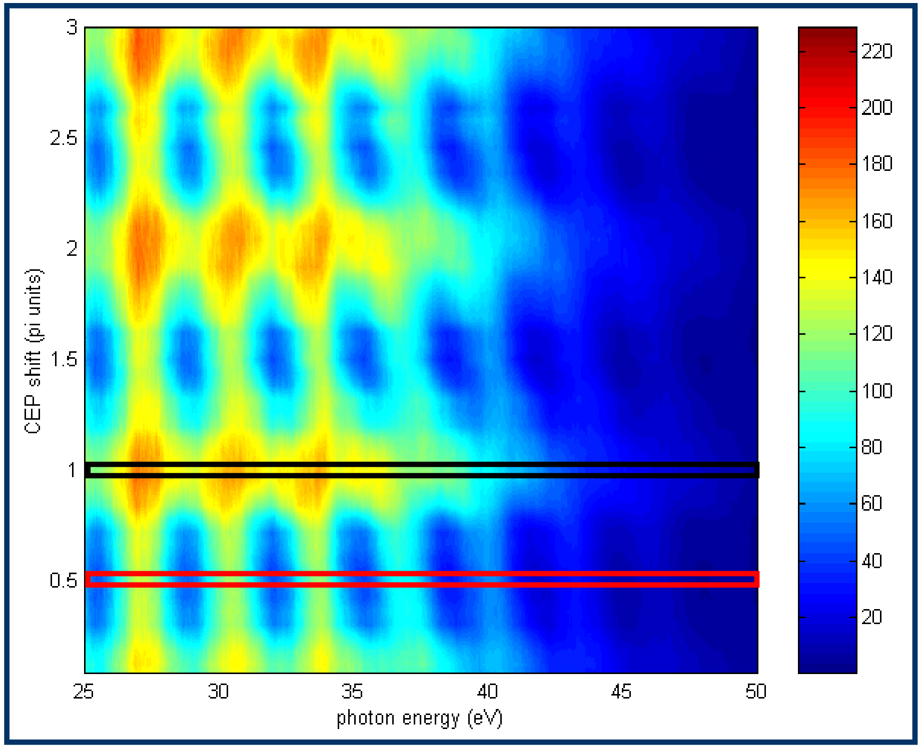


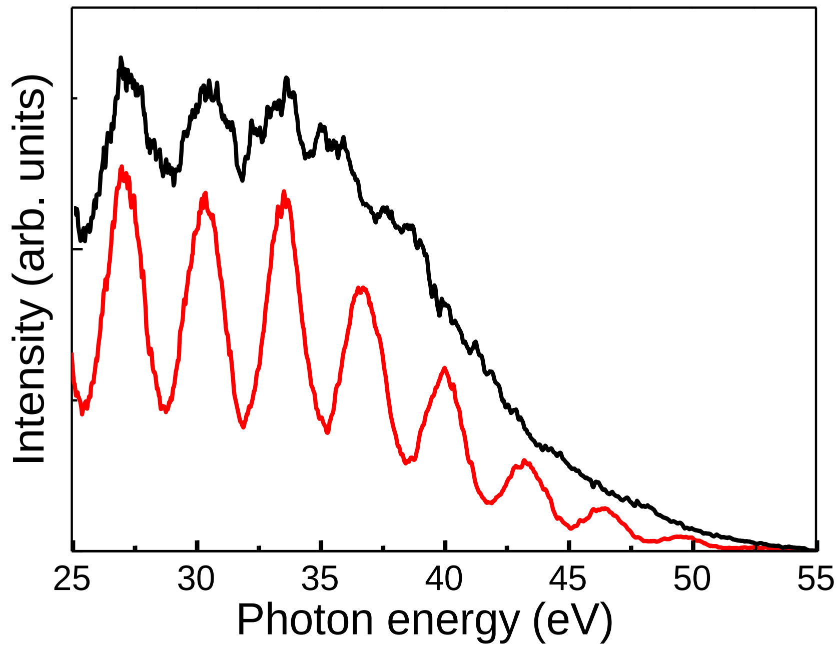
Polarization gating



Experimental results:Argon

- pulse duration τ = 5 fs; delay δ = 6.2 fs; $\psi_0 < \psi < \psi_0 + 3\pi$





➔ **Periodic change of amplitude and shape for $\Delta\psi = \pi$**

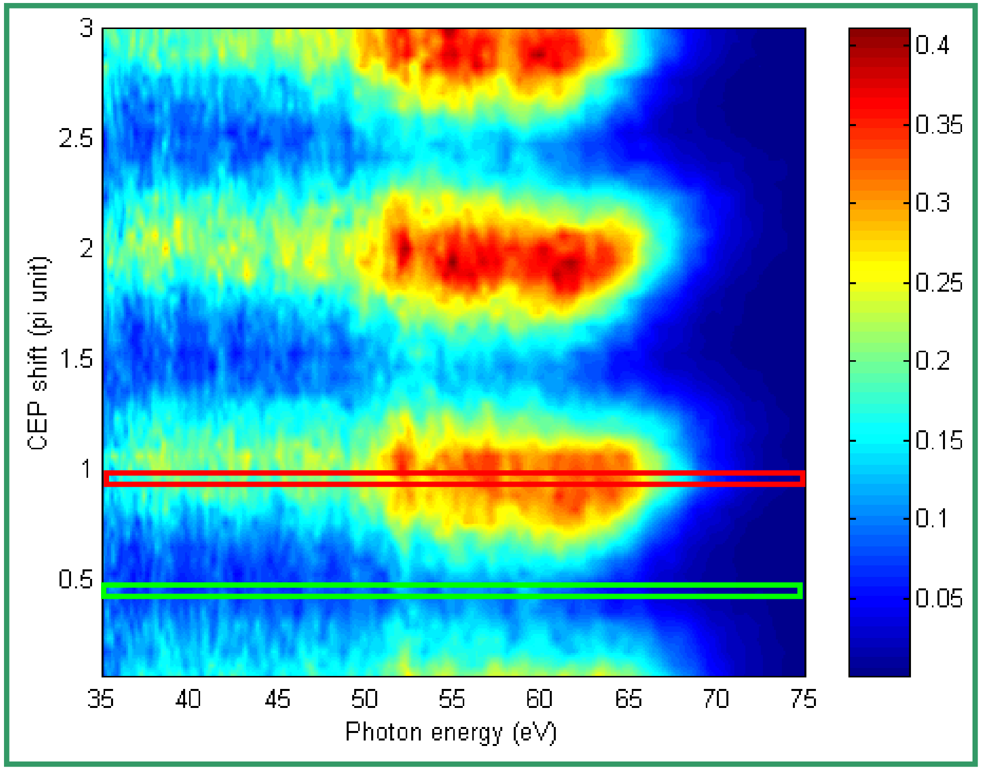
➔ **Continuous spectra from 30 eV to 55 eV for particular ψ**

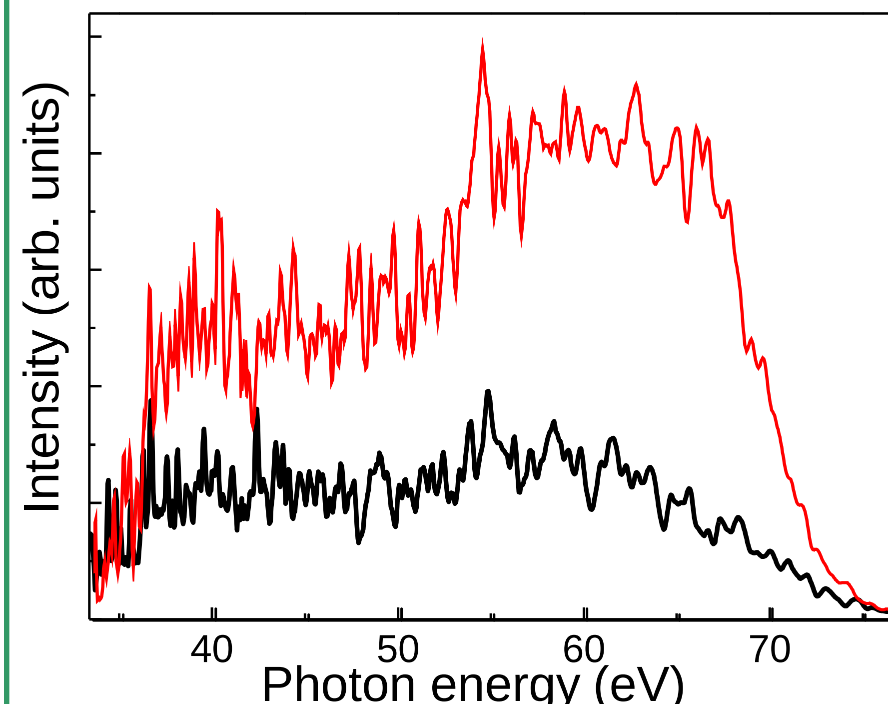
➔ **CEP drives transition from double to single emission**

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

Experimental results:Neon

- pulse duration τ = 5 fs; delay δ = 6.2 fs; $\psi_0 < \psi < \psi_0 + 3\pi$





➔ **Strong periodic modulation of emission efficiency for $\Delta\psi = \pi$**

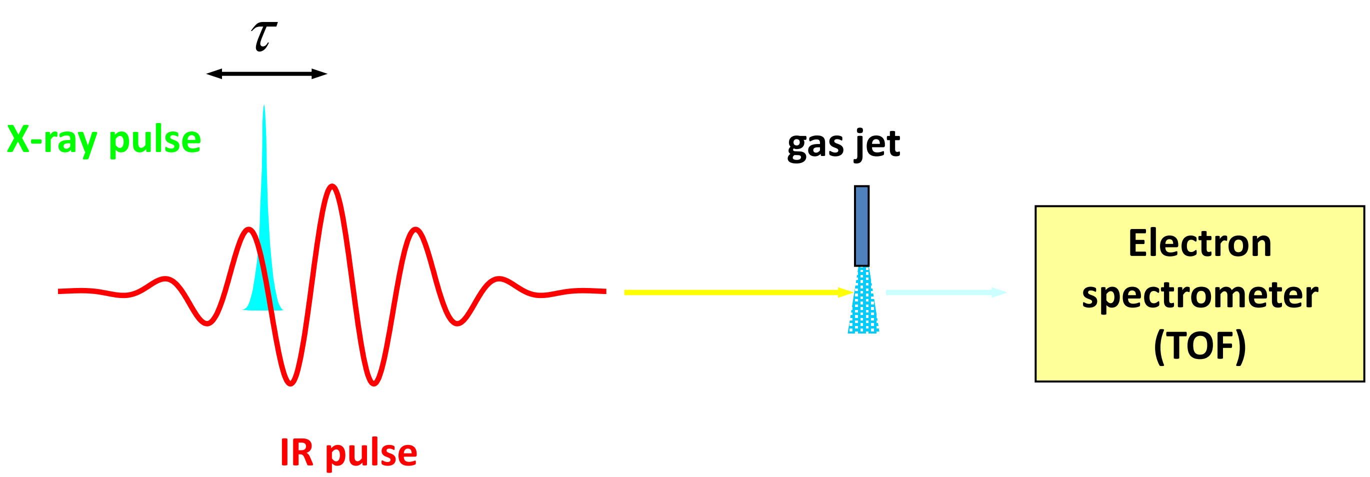
➔ **Continuous spectra from 30 eV to 75 eV for all CEPs**

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

Temporal characterisation: 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

Frequency-Resolved Optical Gating for Complete Reconstruction of Attosecond Bursts

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

➔ **Photoionization spectrum:**

delay between IR and XUV pulses

dipole transition element (assumed constant)

XUV field

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

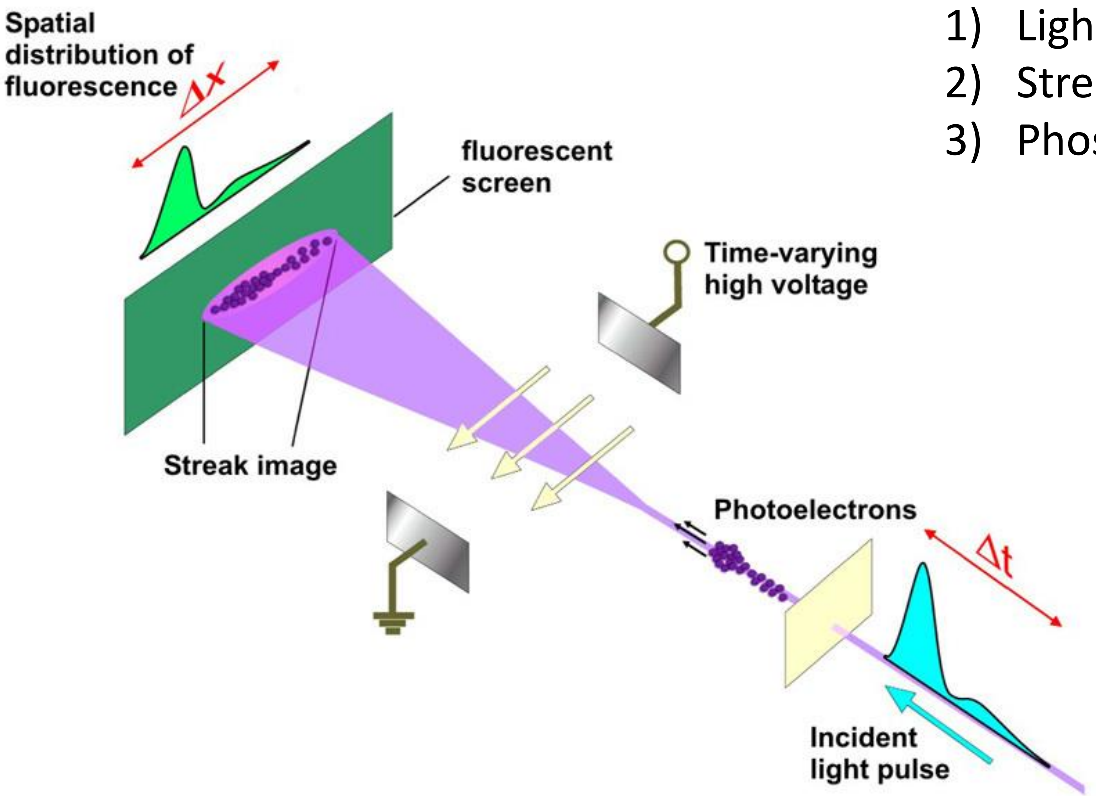
phase gate *G*(*t*)

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

v: final electron velocity
A(t): IR vector potential

➔ **The IR laser field provides a phase gate for FROG measurements on attosecond bursts**

Characterization of isolated attosecond pulses: streak camera

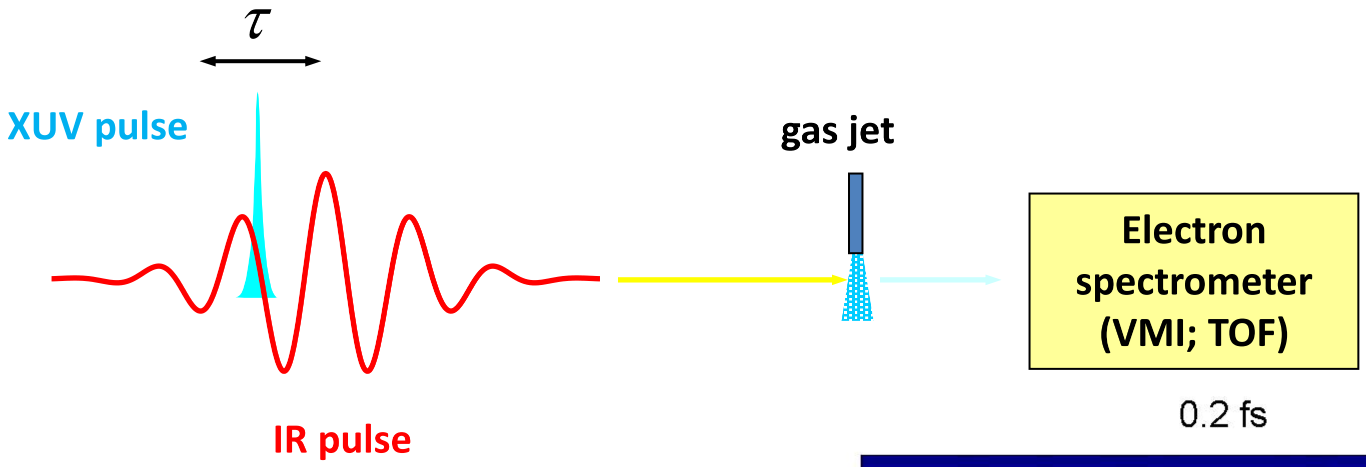


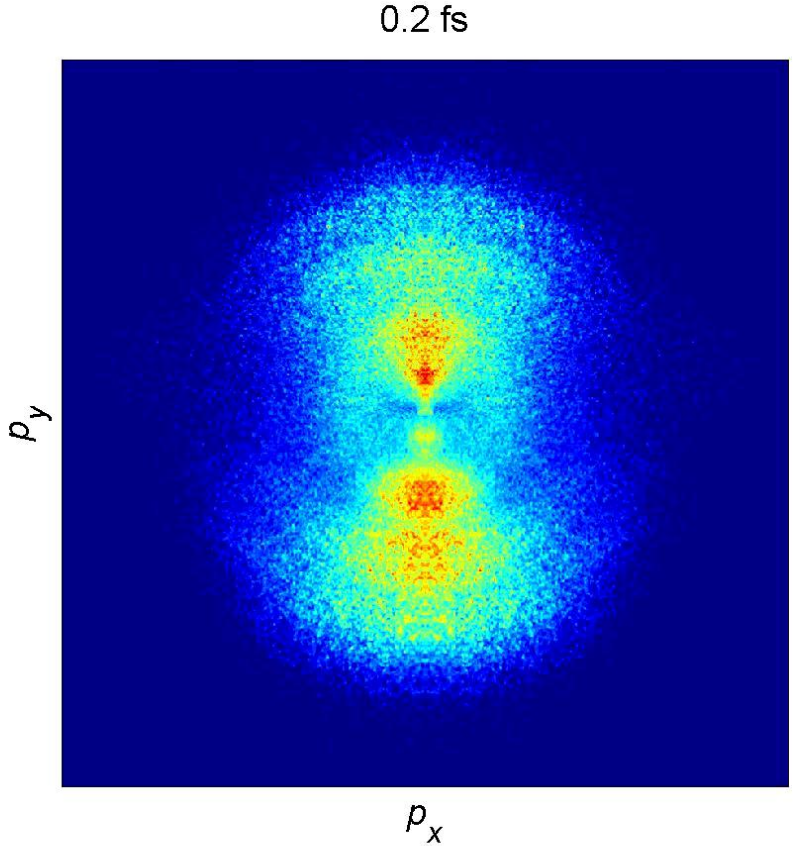
1) Light pulse→ electron bunch
2) Streaking field (time-varying high voltage)
3) Phosphor screen and streaked image

http://www.mpg.de/495195/pressRelease200402241

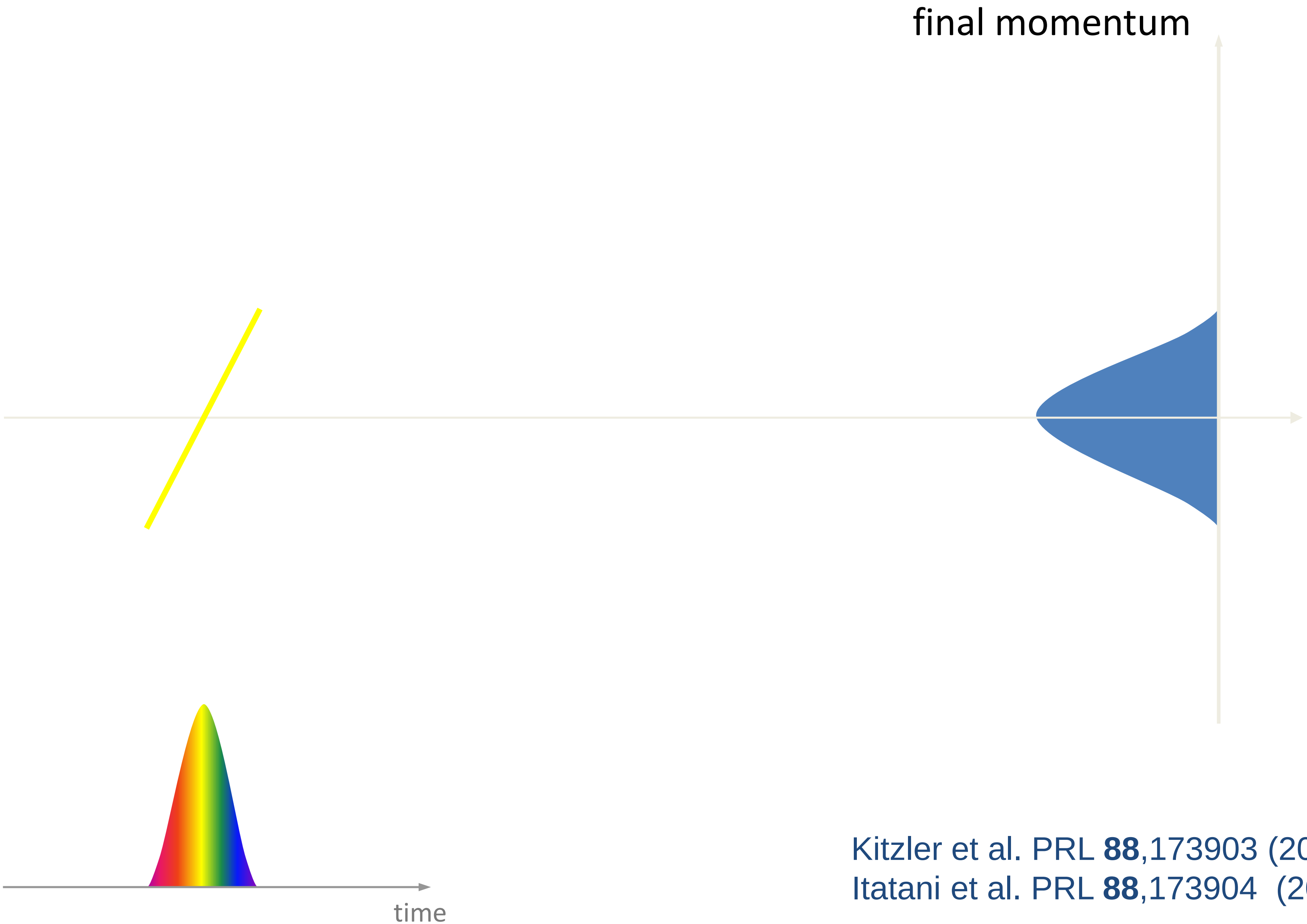
Does it work in the attosecond domain?

Characterization of isolated attosecond pulses



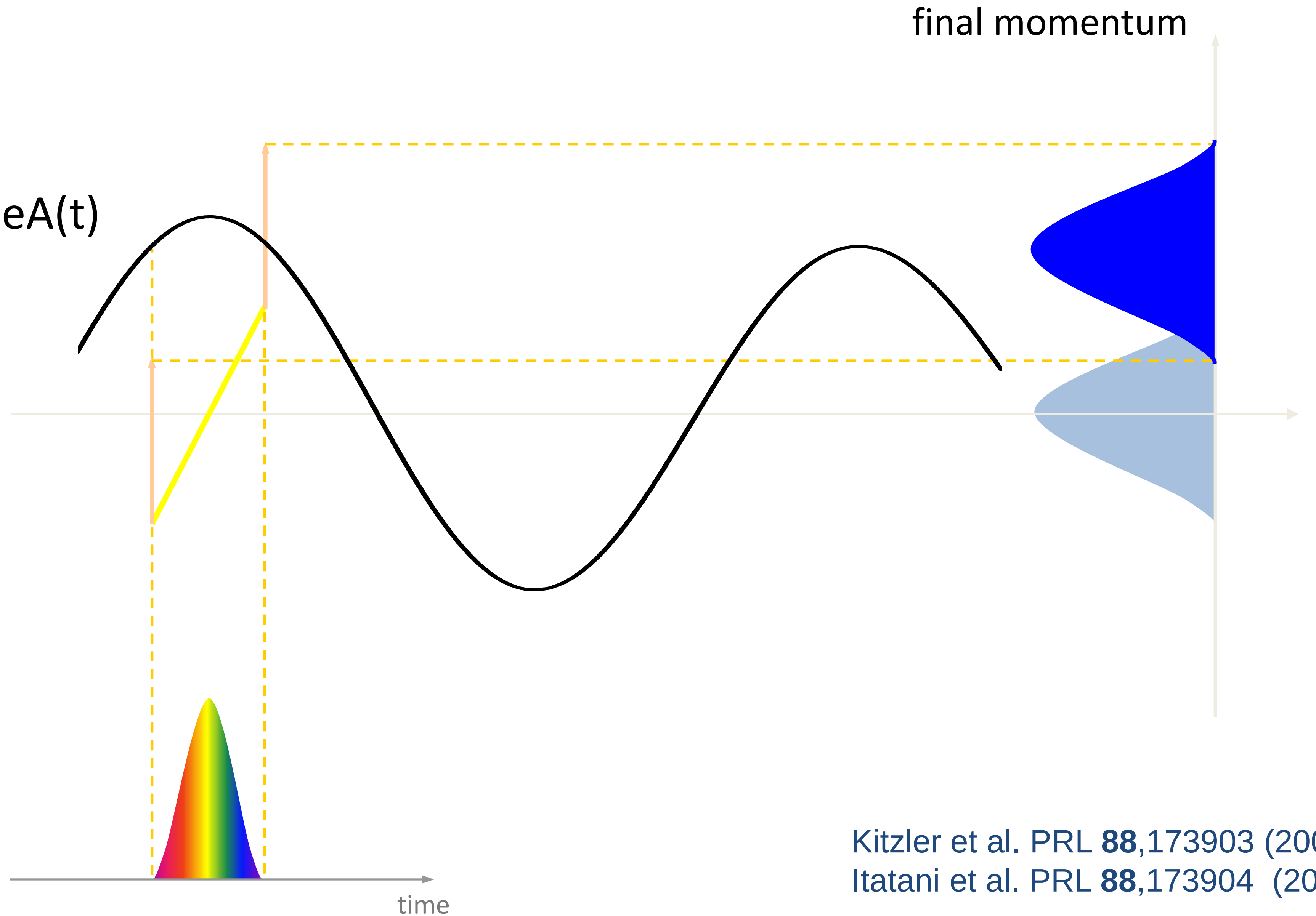


Attosecond streak camera



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

Attosecond streak camera



Kitzler et al. PRL **88**,173903 (2002)
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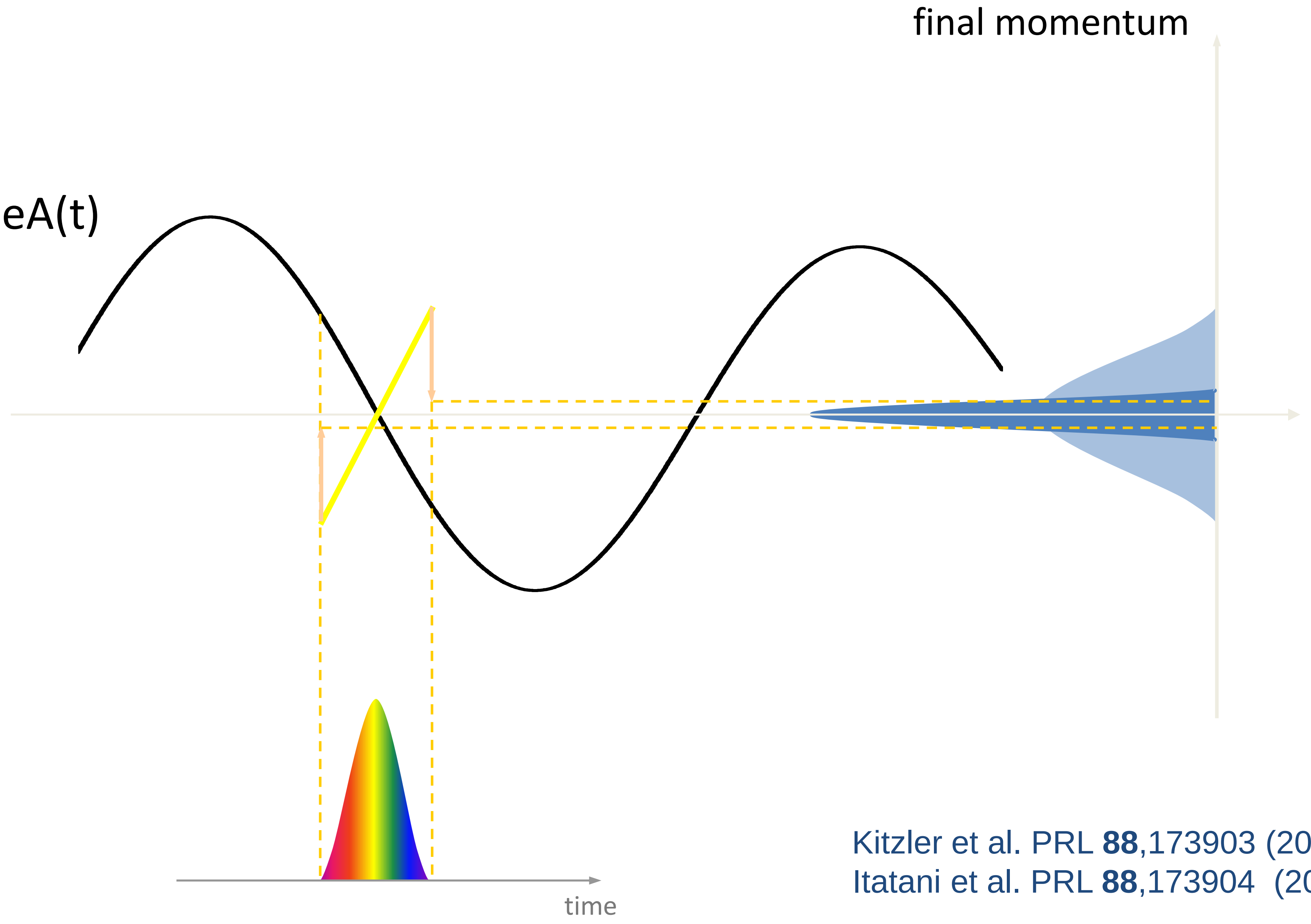
Attosecond streak camera



Kitzler et al. PRL **88**,173903 (2002)
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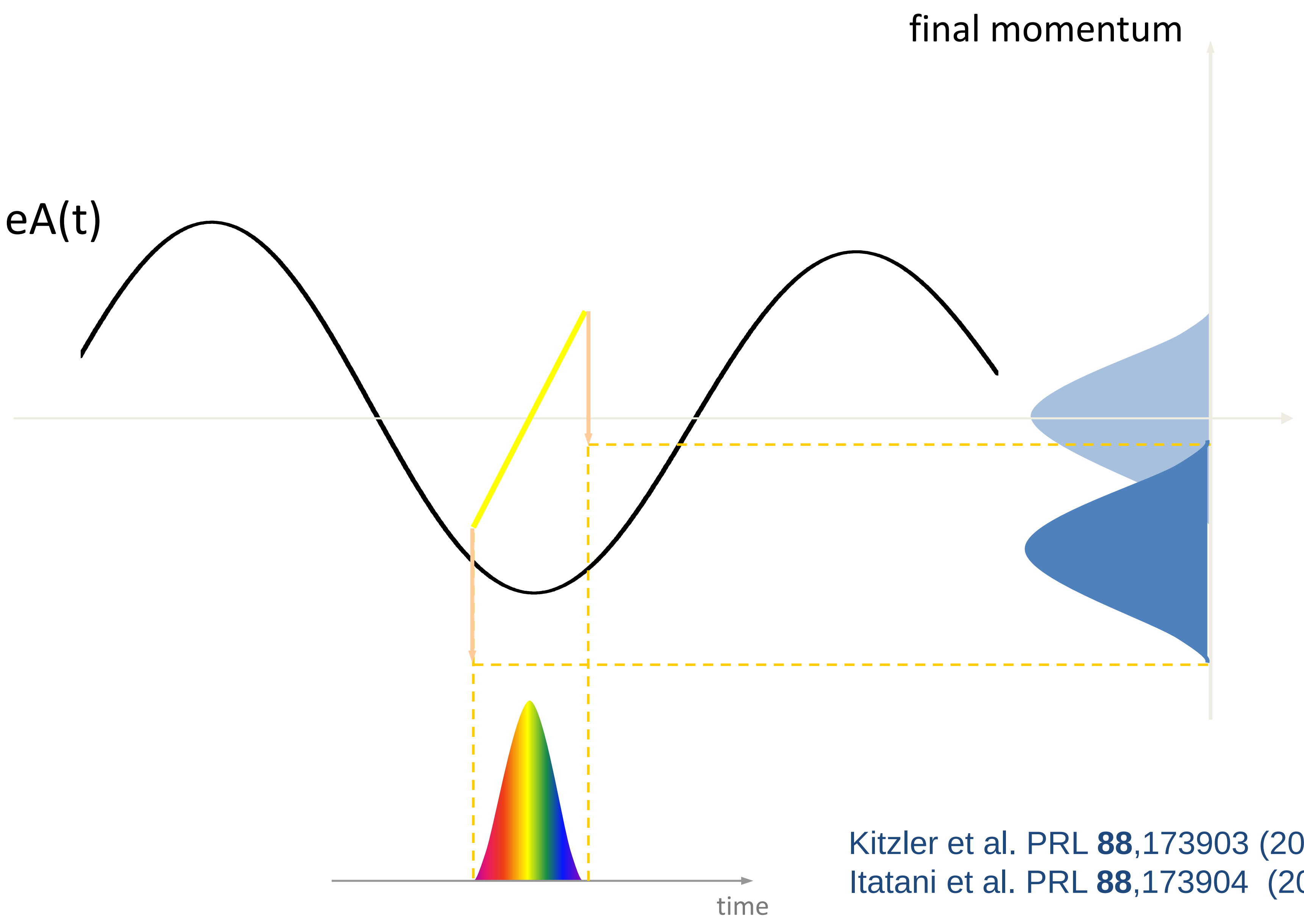
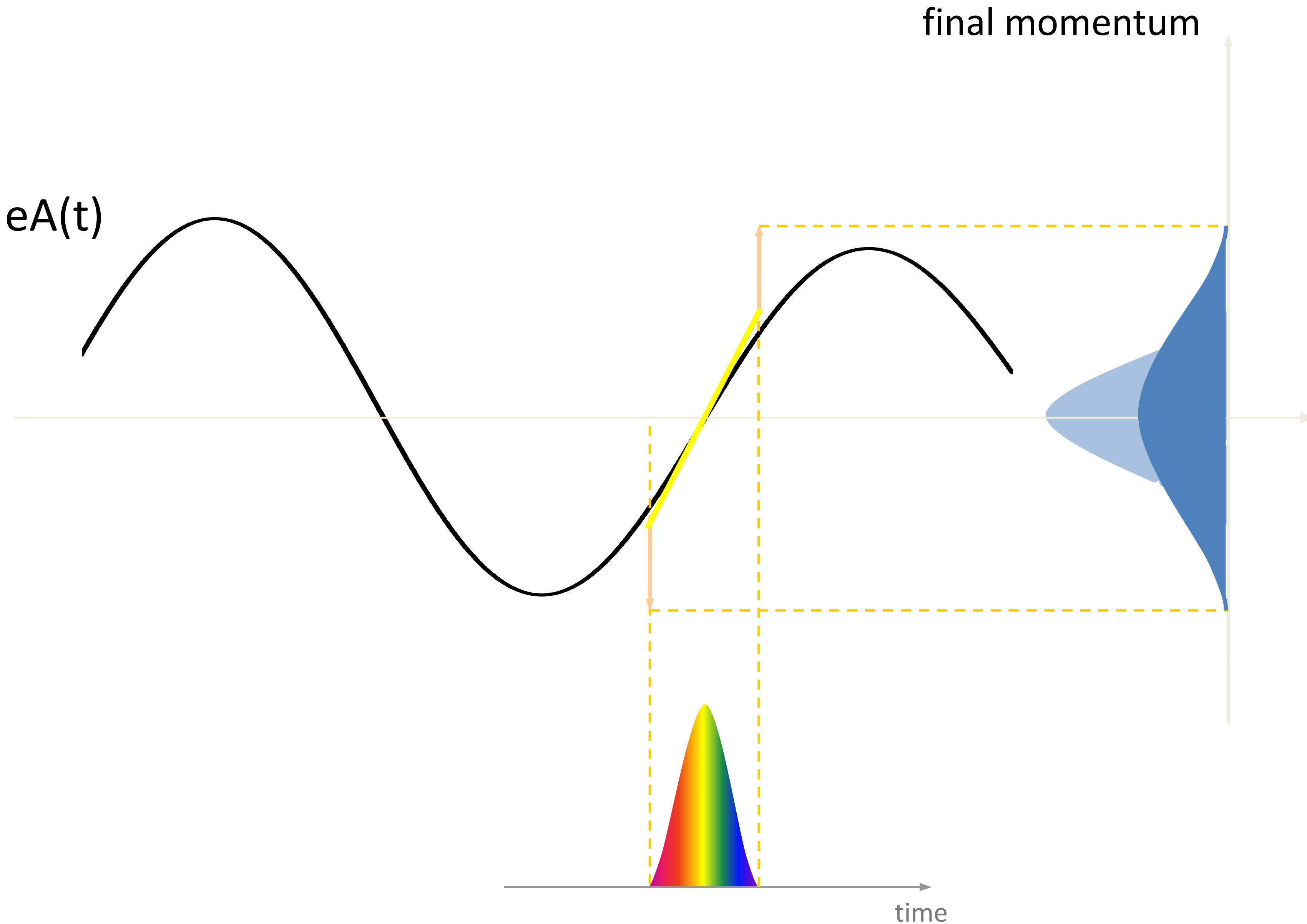
Attosecond streak camera

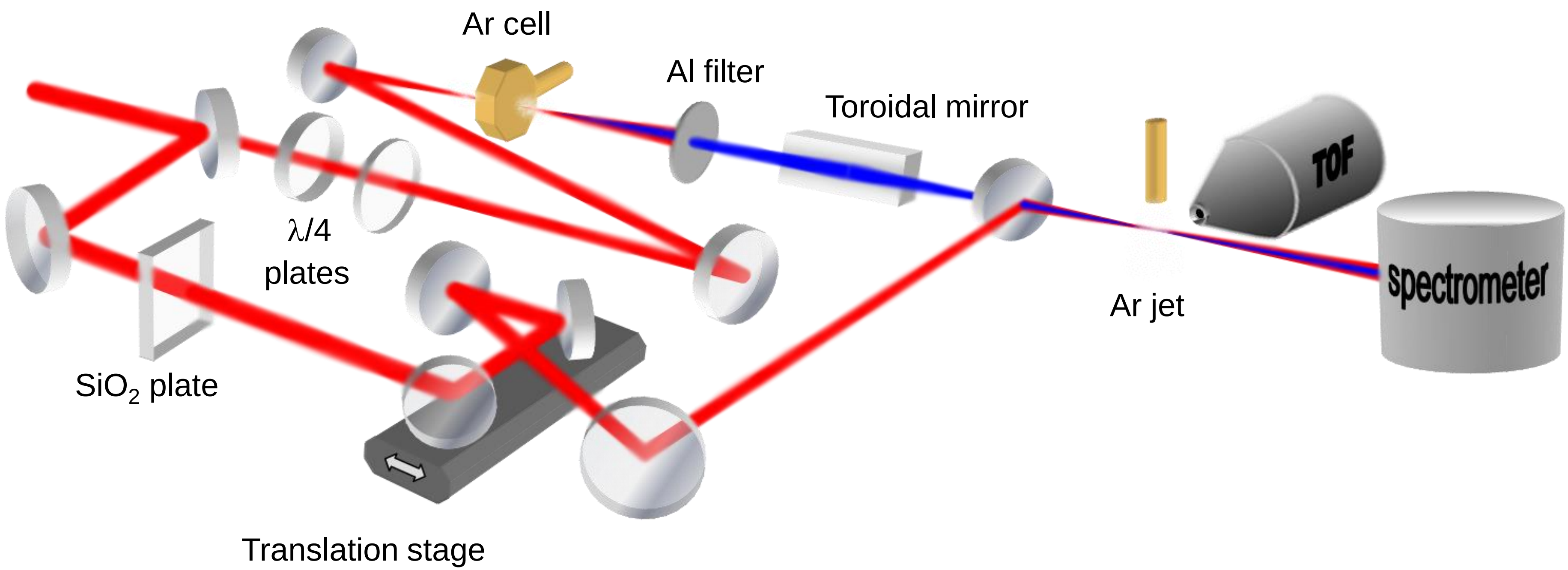


Kitzler et al. PRL **88**,173903 (2002)
Itatani et al. PRL **88**,173904 (2002)

Attosecond streak camera

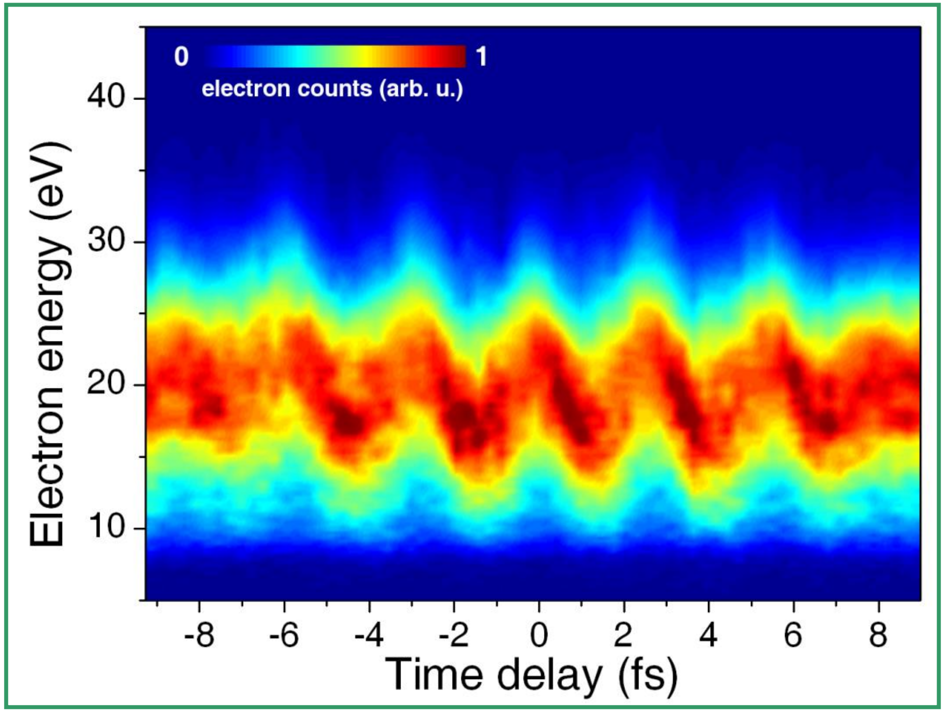


Experimental setup

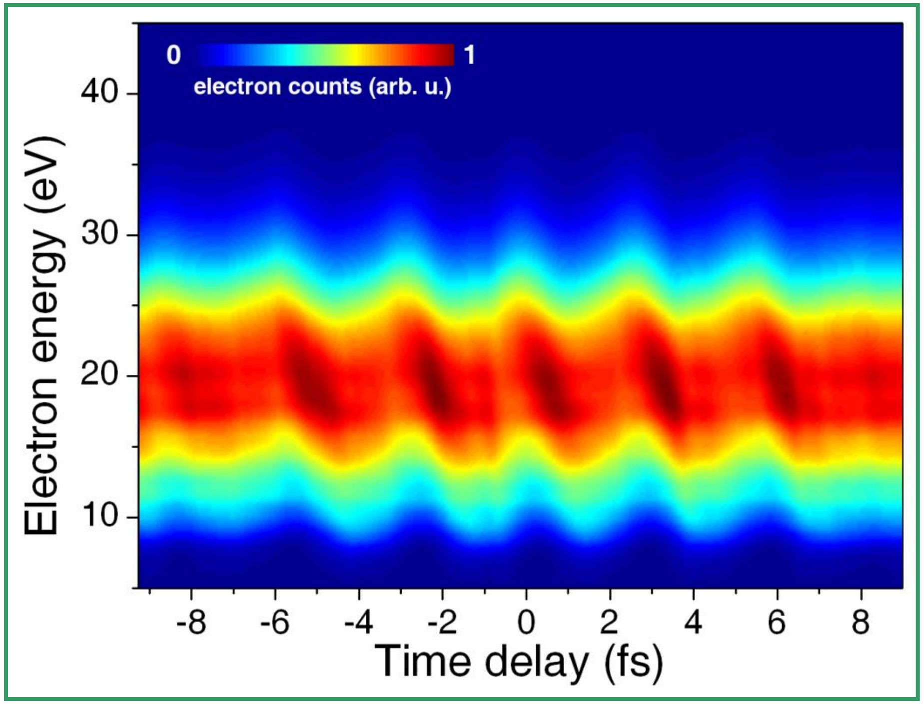


Temporal characterisation

- 100-nm Aluminum filter

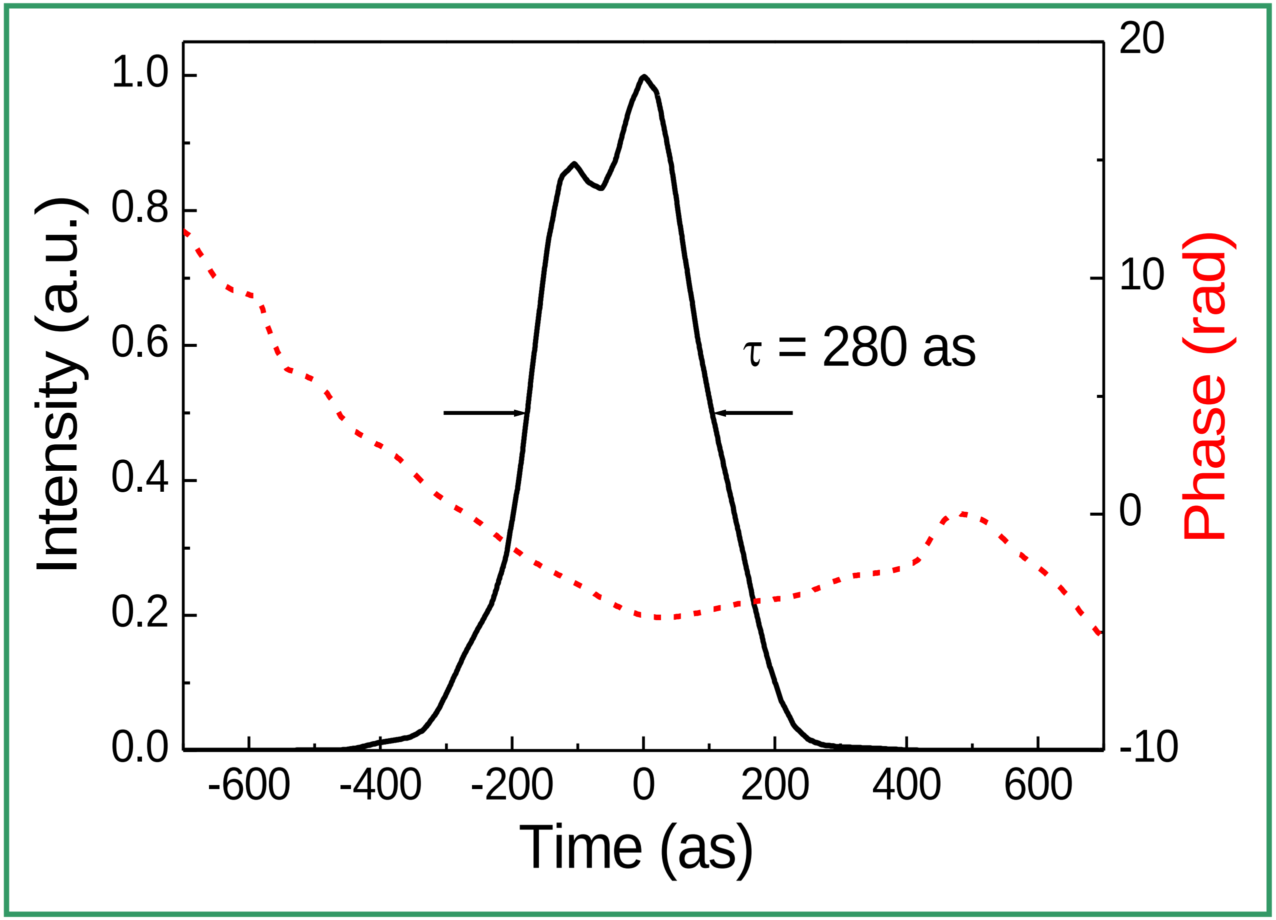


Retrieved CRAB trace



➢ **Positive chirp**

Intensity profile and phase

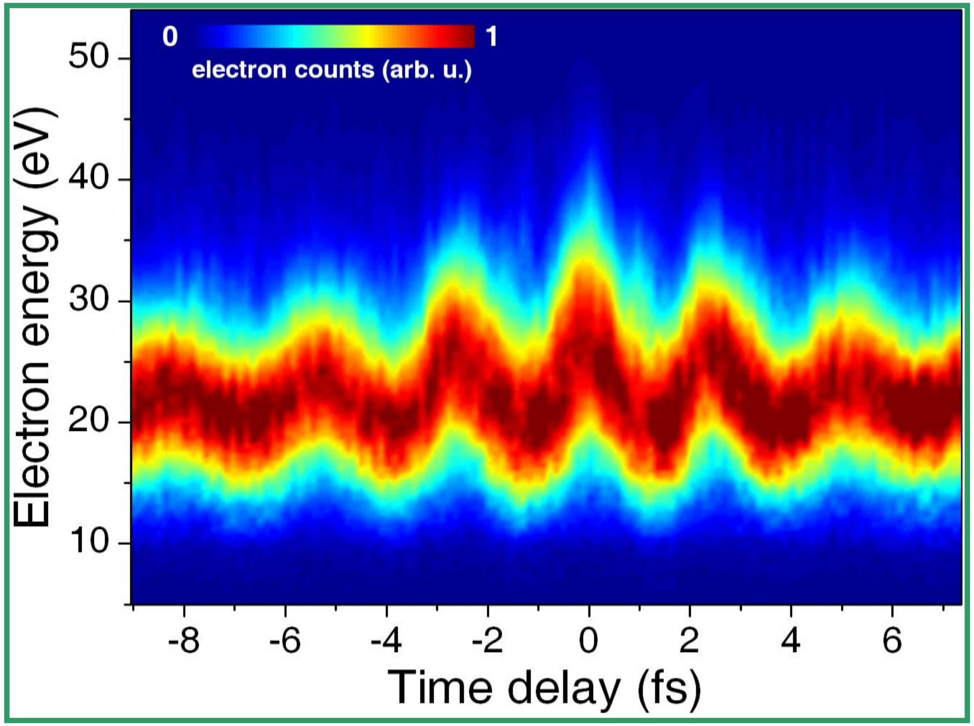


- 2.5 optical cycles
- Positive chirp (atto chirp)

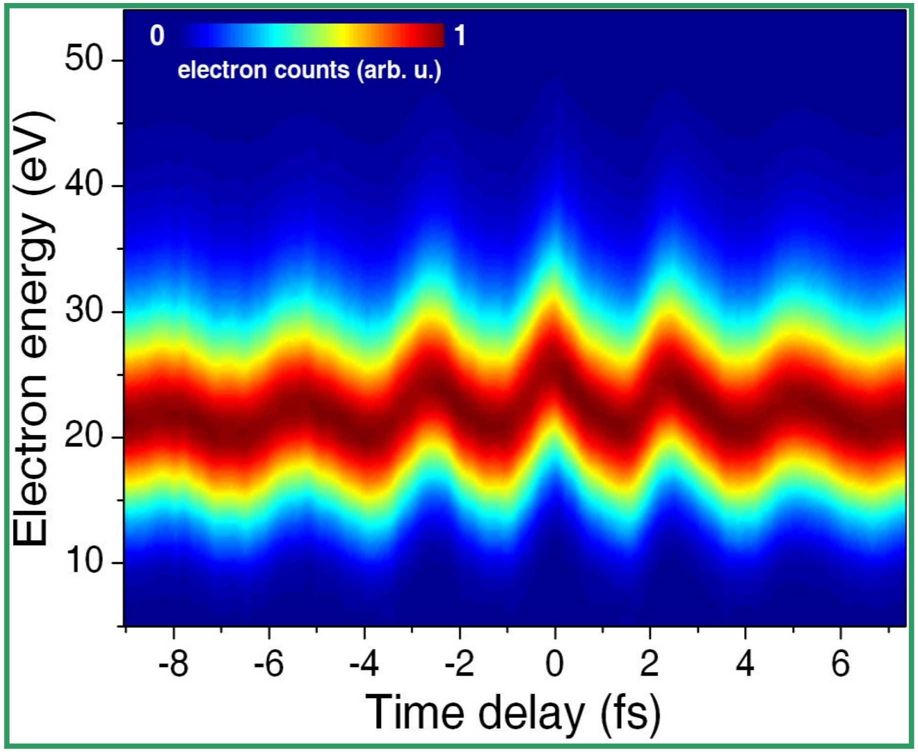
Dispersion compensation

- Use of Aluminum foils

➔ **300-nm Aluminum filter**

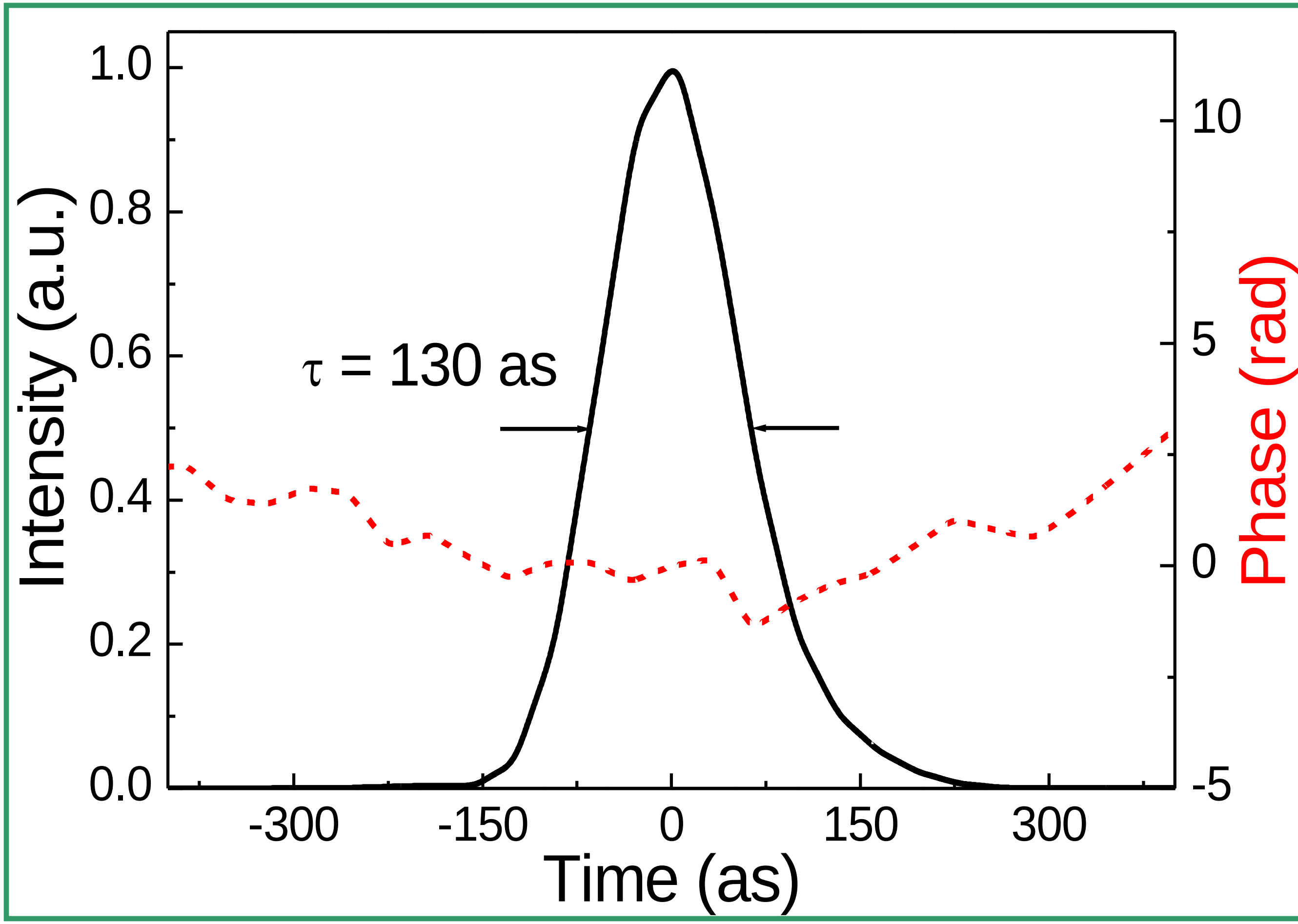


Retrieved CRAB trace



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

Intensity profile and phase

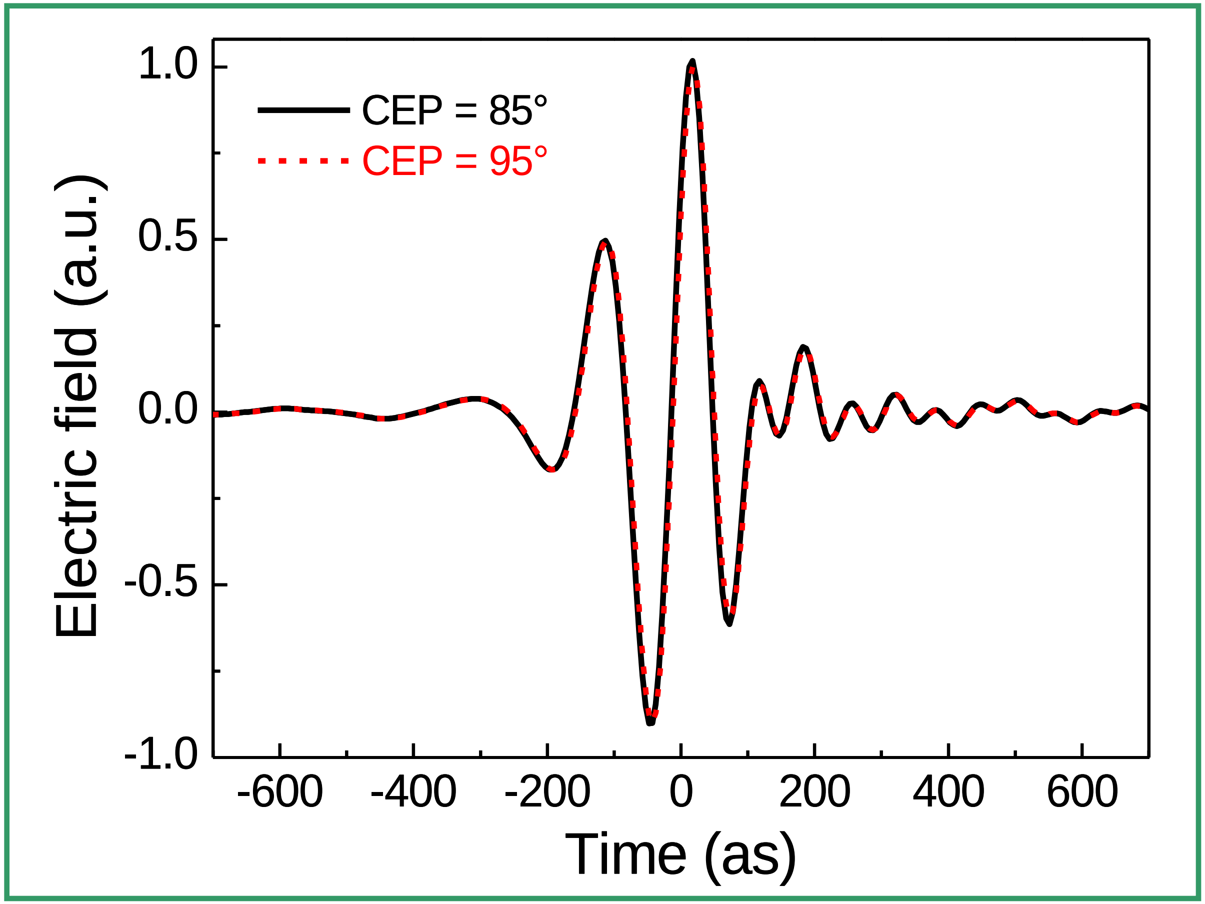


G. Sansone *et al.* Science **314**, 443 (2006).

- ➔ **Good dispersion compensation**
- ➔ **Near-single cycle pulse**

Stability of as electric field :CEP variation

- Nonadiabatic saddle-point simulations



➔ **Negligible influence in the quite broad CEP range giving rise to isolated pulses**

MARIE SKŁODOWSKA-CURIE ACTION: INNOVATIVE TRAINING NETWORKS (ITN)

"MEDEA"

Molecular Electron Dynamics investigated by IntensE Fields and Attosecond Pulses

15 ESRs position for working in the field of electronic/nuclear molecular dynamics excited by attosecond and XUV FELs pulses

- ✓ 11 research institutions
- ✓ 6 companies
- ✓ 1 museum
- ✓ 1 outreach institute
- ✓ 1 FELs institution