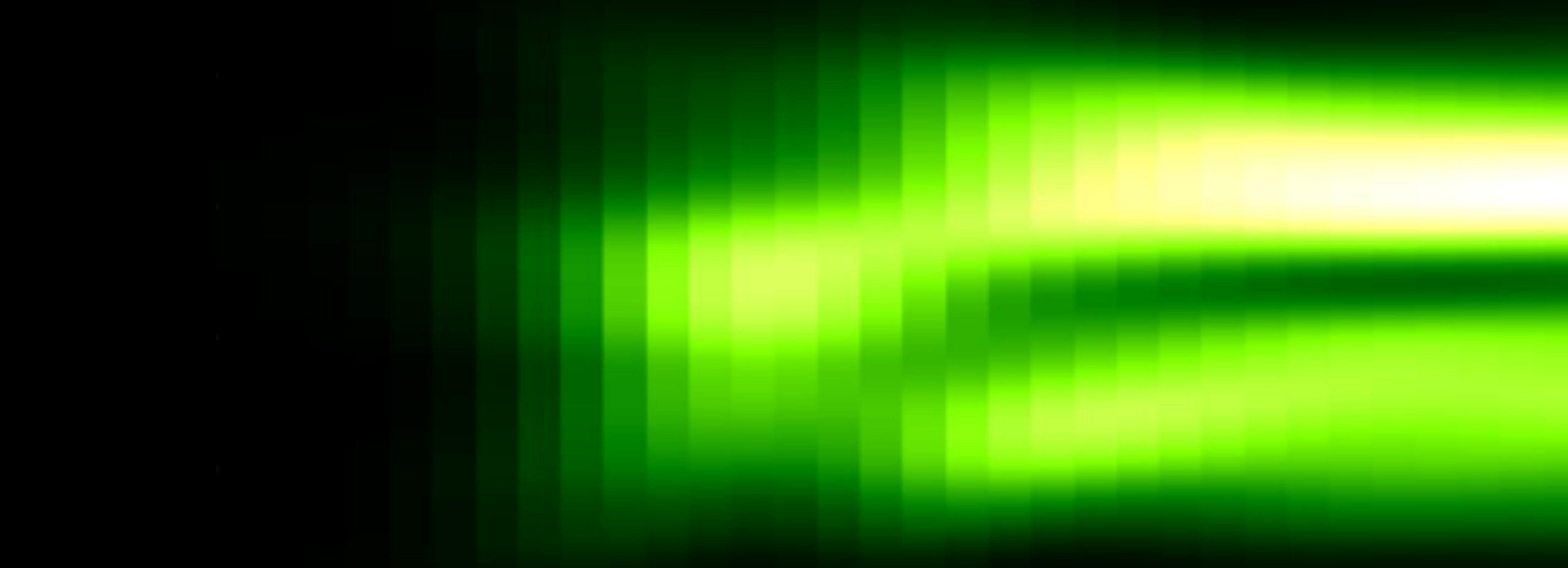


ATTOLab-Orme: a platform for the study of attosecond dynamics

Pascal Salières

LIDYL / Attophysics Group
CEA-Saclay
France

Outline

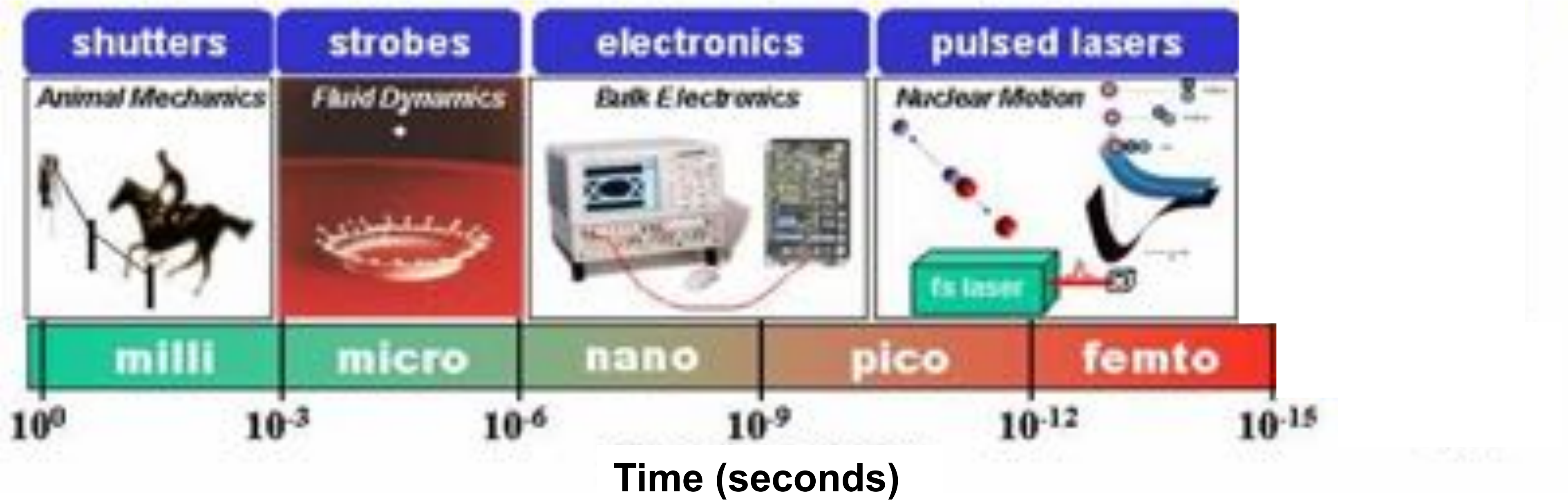
1) Ultrafast dynamics and attosecond light tools

2) Attosecond facility at Paris-Saclay University: ATTOLab-Orme

3) Attosecond photoionization spectroscopy

=> Shooting the attosecond movie of photoemission

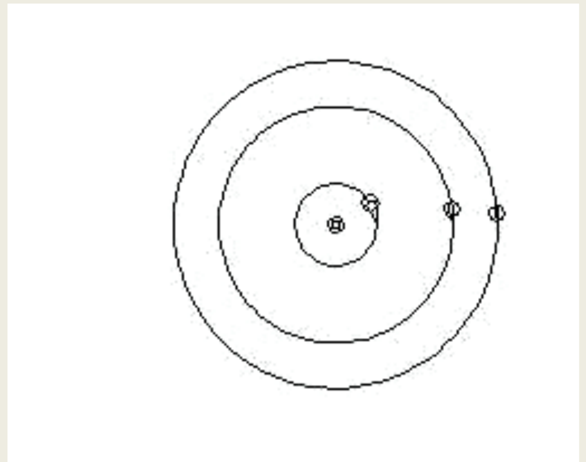
A race against time…



-Femtosecond: timescale of atomic movement inside molecules
=> femtochemistry: Nobel Prize Hamed Zewail (1999)
'pump – probe' (measurement) technique

-Attosecond: timescale of electron movements
in the core of atoms and mol.

Period of the 1ère Bohr orbit = 150 as



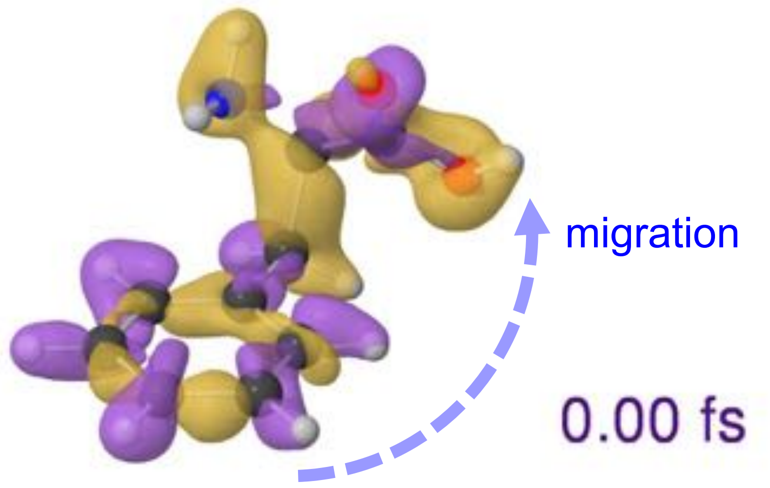
Coherent wavepacket dynamics on attosecond timescale

Understanding and controlling in ‘real time’ (atto –femto)

the evolution of electronic & nuclear wavepackets localized at atomic scale (Å –nm)

- **a unifying concept**
- **efforts & competition worldwide**

Atoms & Molecules

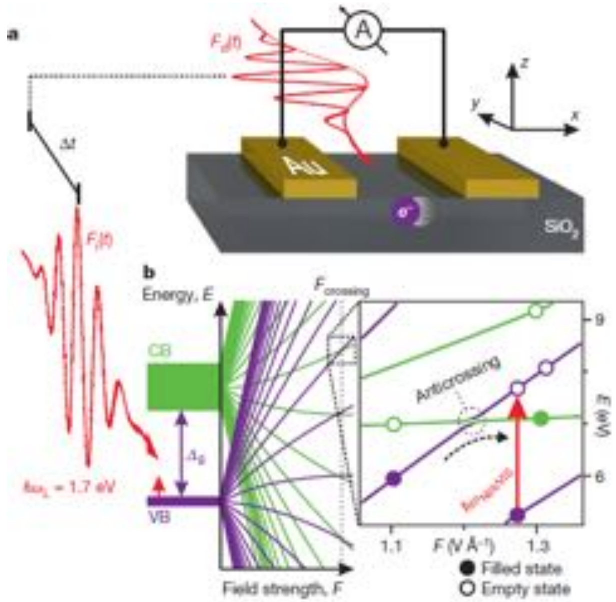


Ultrafast charge migration

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

F. Remacle and R. D. Levine *PNAS* 103, 6793 (2006)

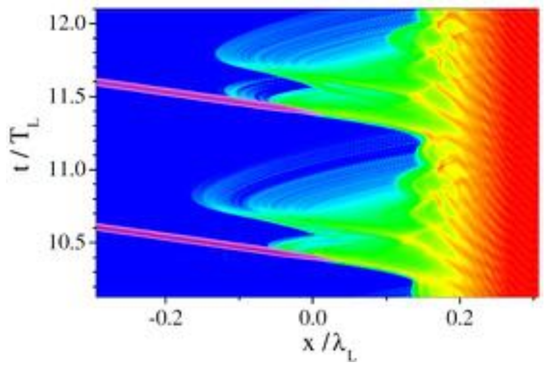
Solids



Attosecond current switch

Schiffrin et al., *Nature* 493, 70 (2013)

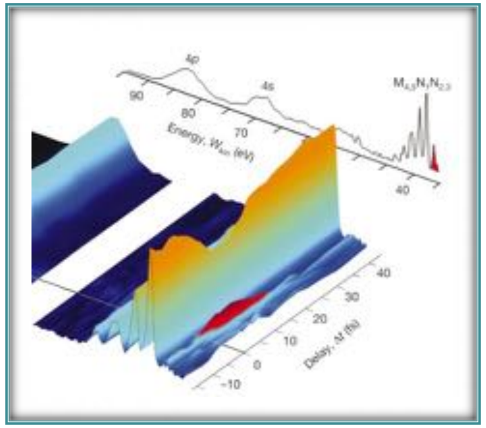
Plasmas



Ultrafast beams of e, protons, Xrays

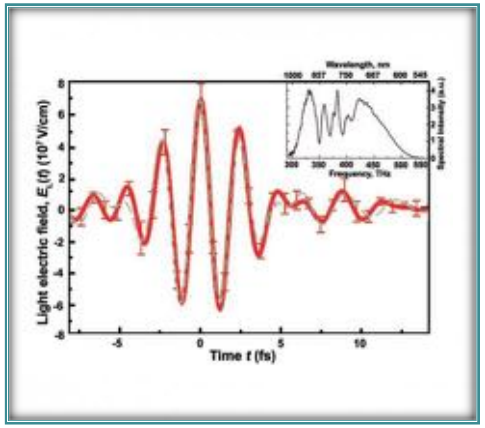
M. Thévenet, *Nature Physics* 12, 355 (2016)

2001-2021: 20 years of attosecond science



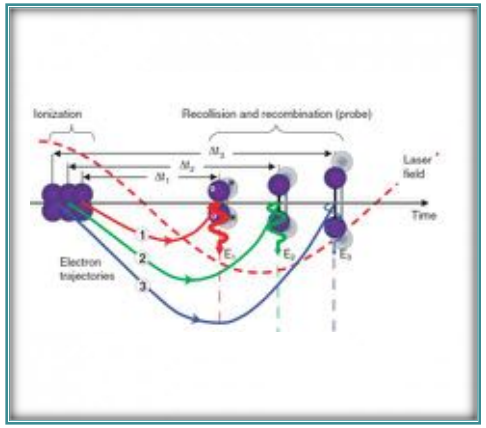
Time-resolved
few-fs Auger decay

[Drescher *et al. Nature* (2002)]



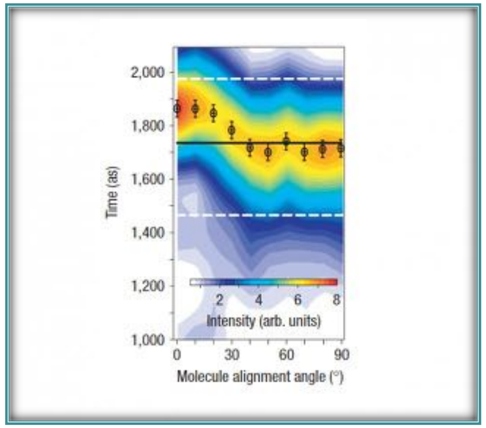
Direct measurement
of light waveforms

[Goulielmakis *et al, Science* (2004)]



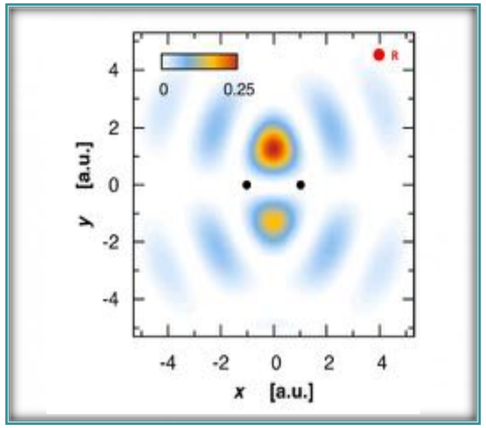
Attosecond-resolved
proton dynamics

[Baker *et al. Science* (2006)]



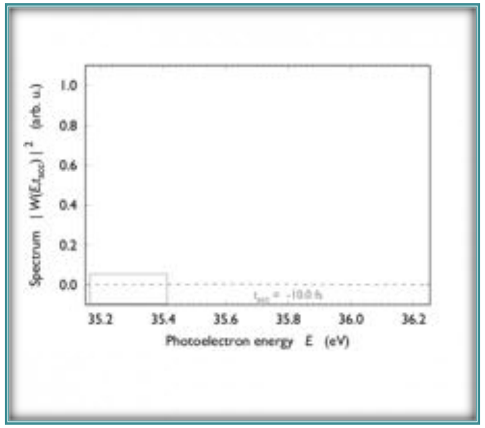
Coherent control
of attosecond emission

[Boutu *et al. Nature Phys*. (2008)]



Attosecond imaging
of electronic wavepackets

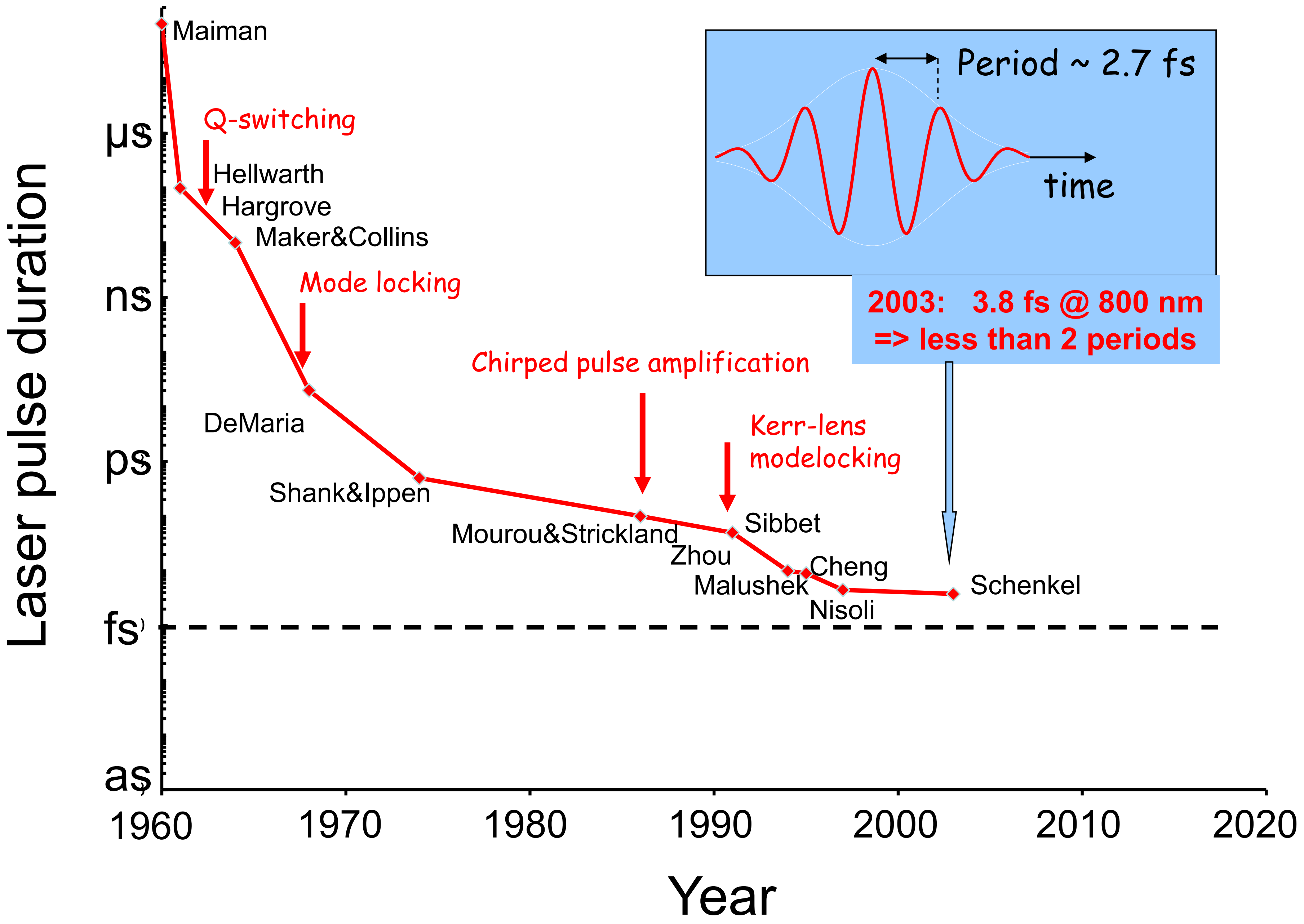
[Haessler *et al. Nature* Phys. (2010)]



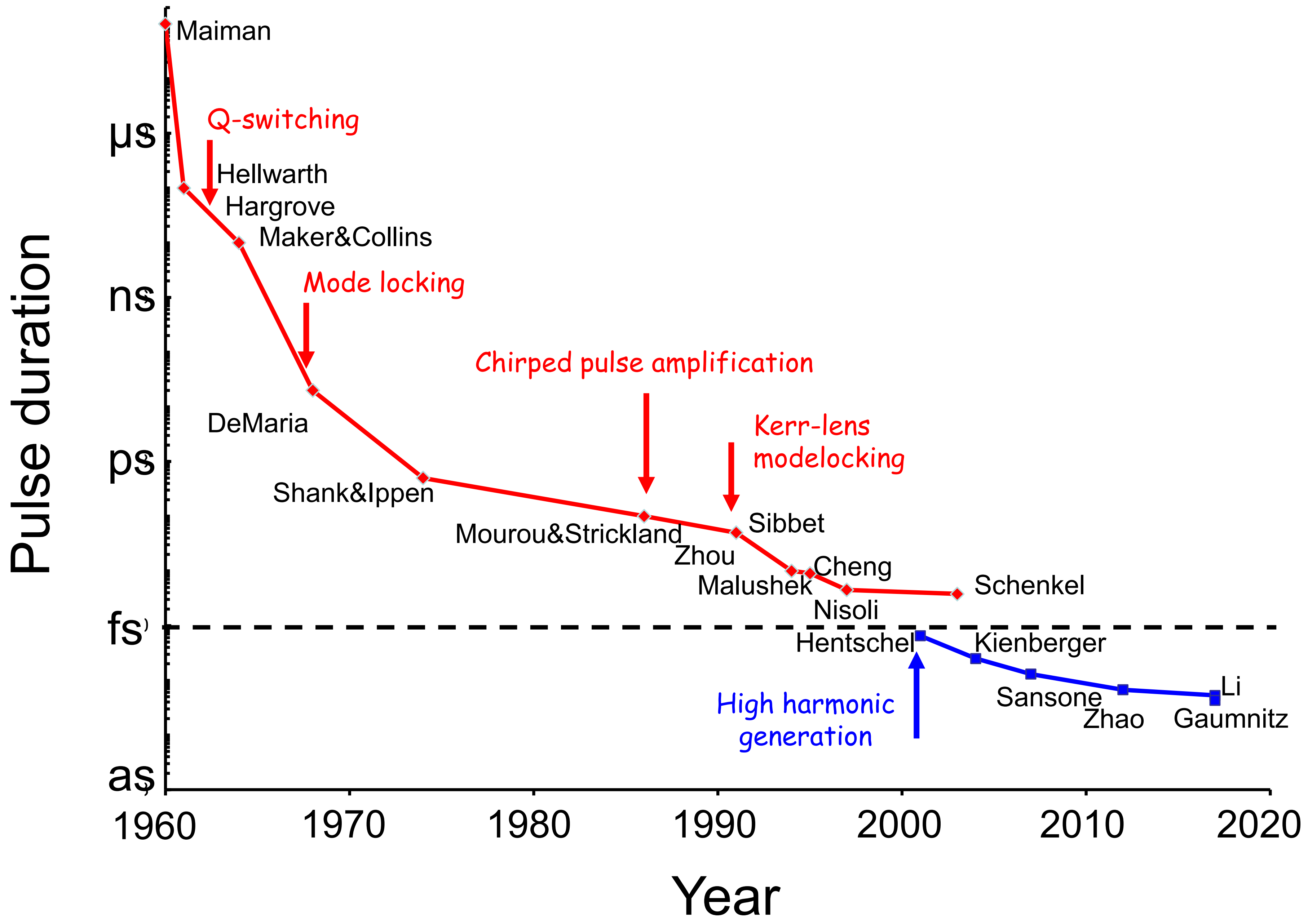
Attosecond dynamics
of photoemission

[Gruson *et al, Science* (2016)]

Light tools: lasers are limited by the femtosecond barrier

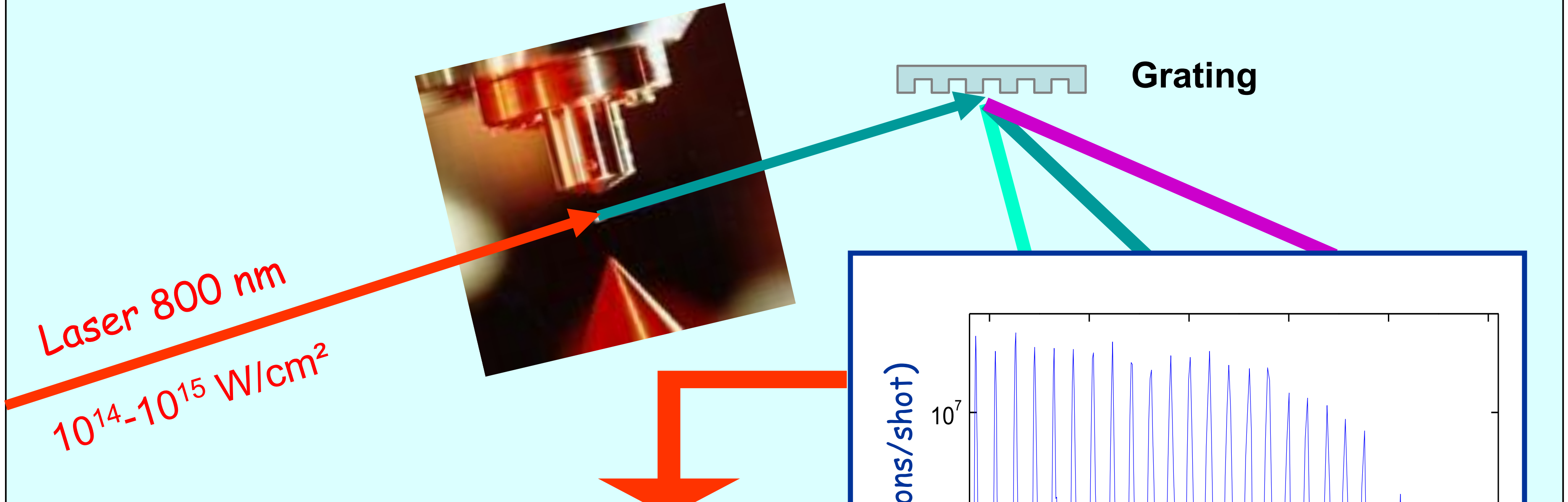


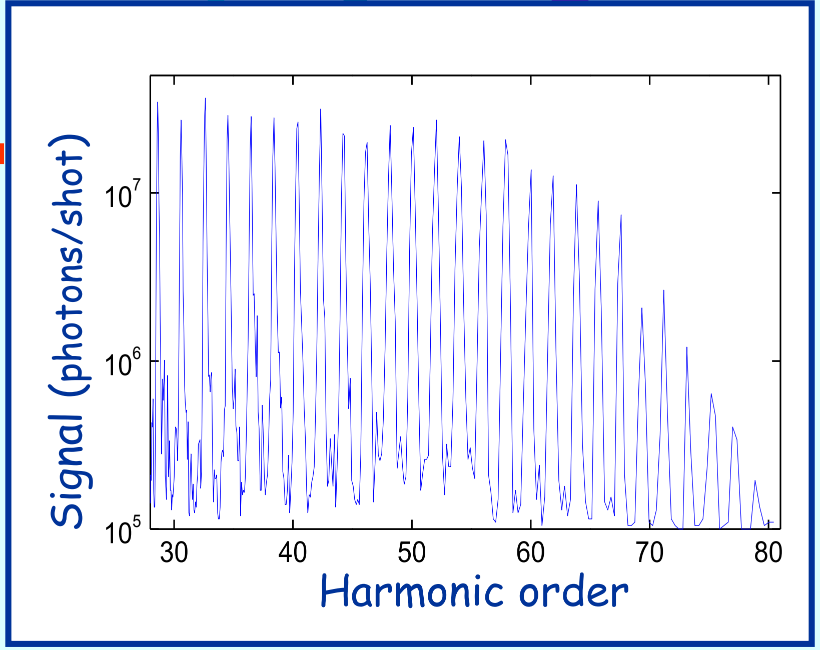
Breaking the femtosecond barrier



A broadband coherent source in the XUV

High-order Harmonic Generation *(Saclay and Chicago, 1988)*



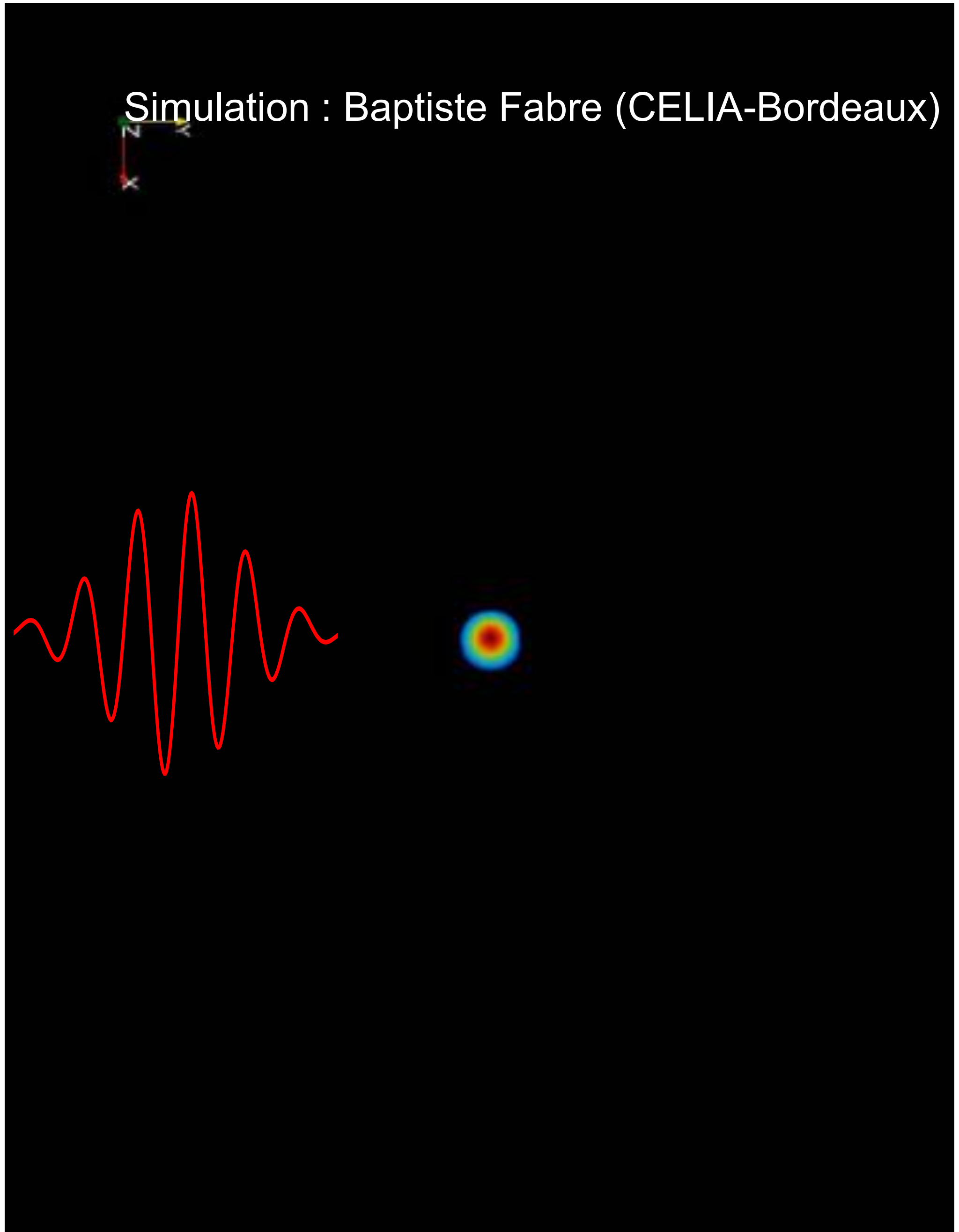


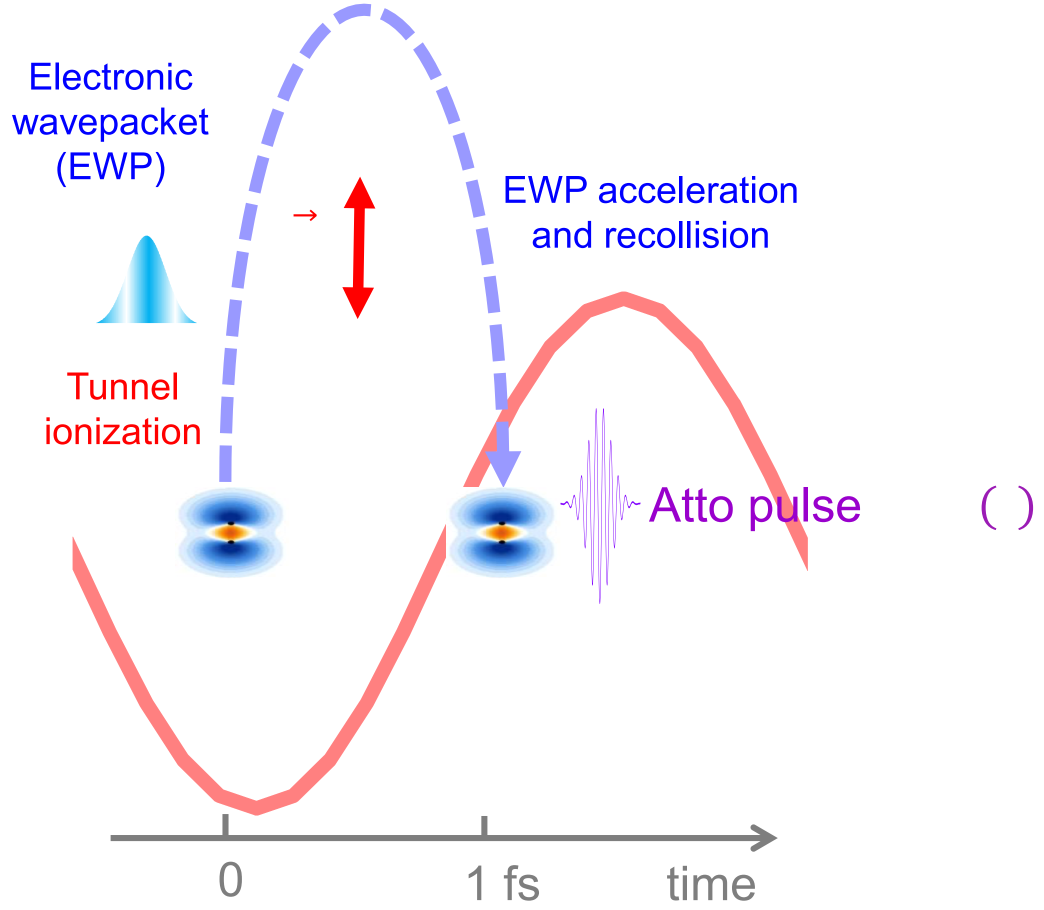
- Strong field regime:

Non-perturbative behavior

Spectrum: plateau / cutoff

Strong-field dynamics: atom in an intense laser field





$$h\nu_{cutoff} = I_p + 3U_p$$

I_p: ionization potential

U_p: ponderomotive energy ~$I\lambda^2$

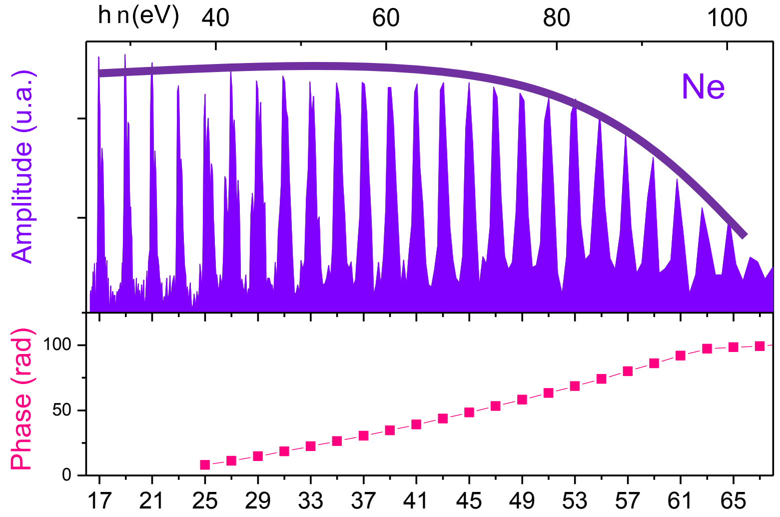
Schafer et al. PRL 1993; Corkum PRL 1993
Lewenstein et al. PRA 1994

The light tools: ultrafast gradients

Controlled "few-cycle" laser pulses

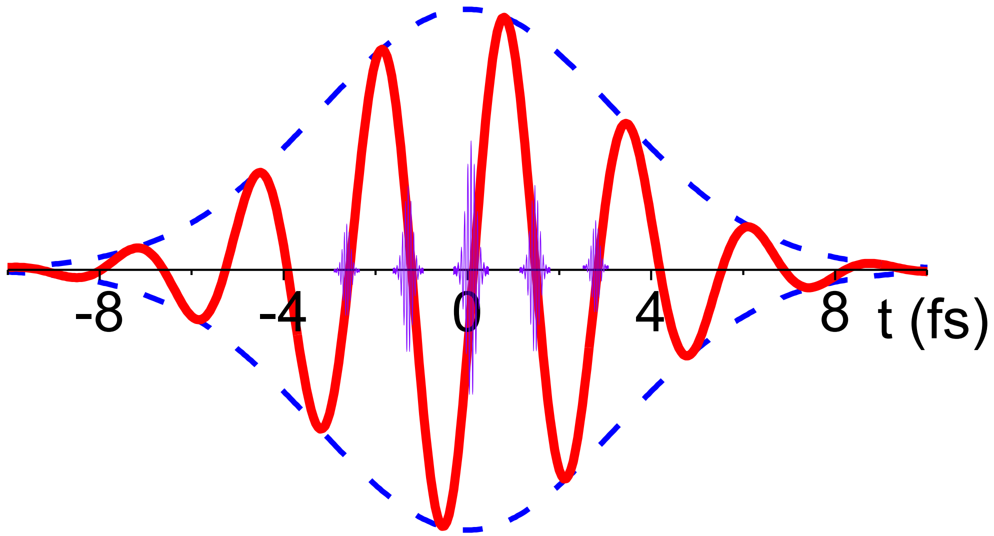
Harmonic spectrum

P.-M. Paul *et al Science 292, 1689* (2001)
S. Aseyev *et al PRL 91, 223902* (2003)



Generation of a train of atto pulses

~ 100 as / l~1-60 nm (XUV: 20 –1600 eV)



Continuous spectrum ↔ isolated atto pulse

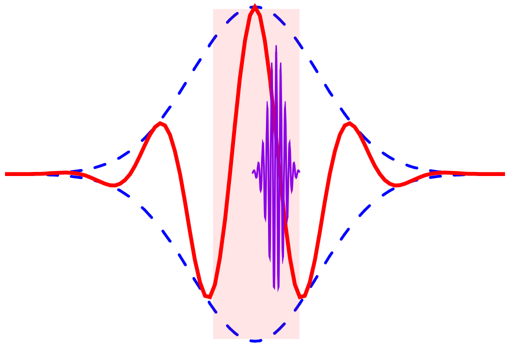
M. Hentschel et al., *Nature 414, 509* (2001)
G. Sansone et al. *Science* 314, 443 (2006)

Isolated attosecond pulses: 'amplitude gating'

laser pulse ~ 5fs with controled carrier-envelope phase F_{CEP}

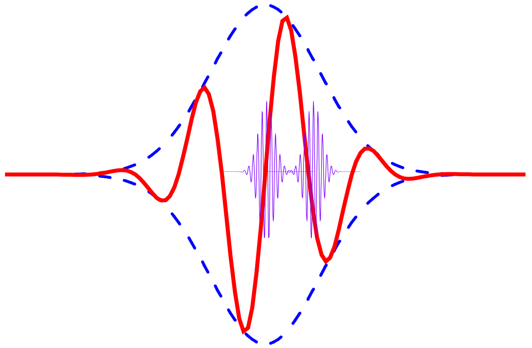
"cosine" pulse $F_{CEP} = 0$

=> emission of high orders confined to the central ½ period

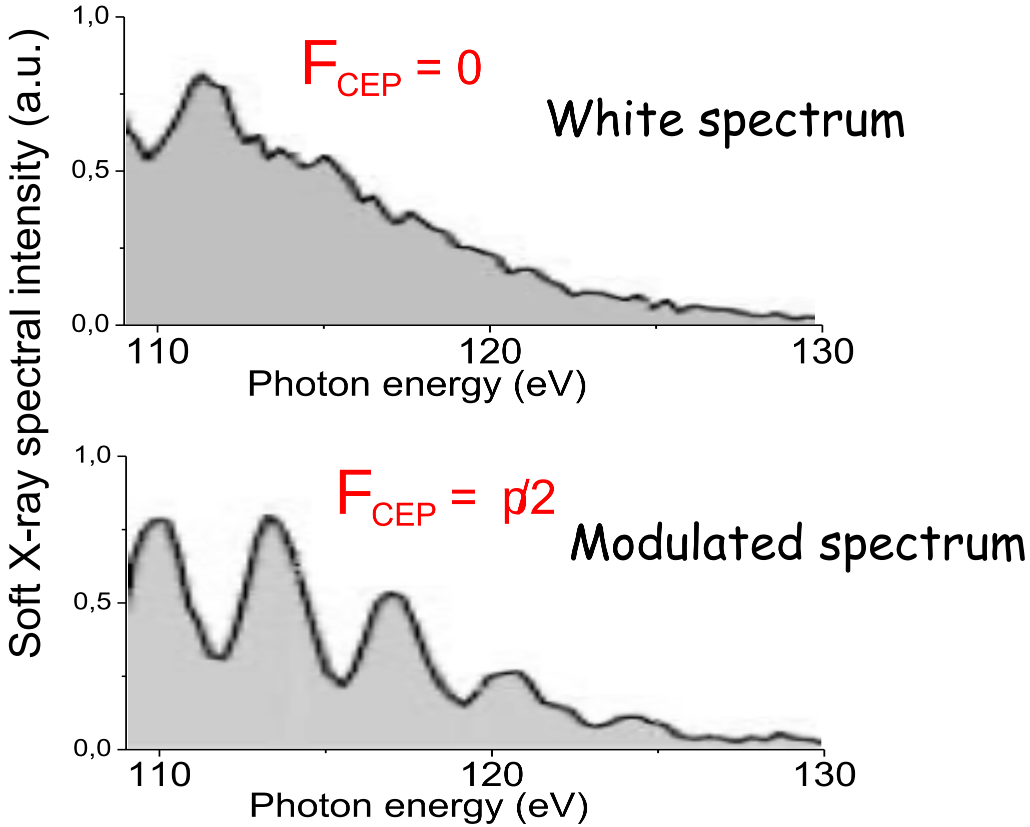


single atto pulse

"sine" pulse $F_{CEP} = p/2$



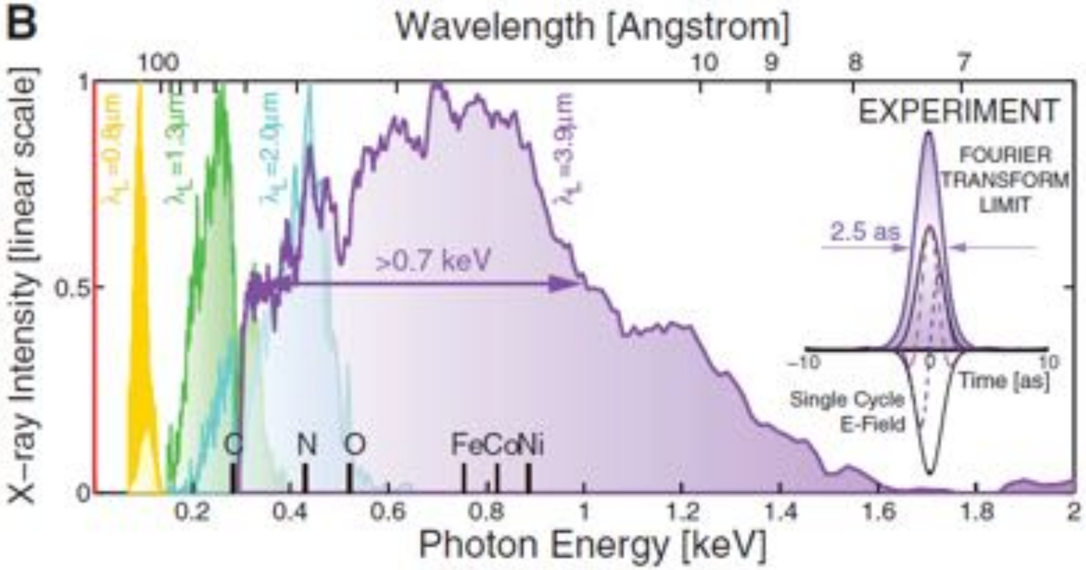
2 atto pulses



Baltuska et al. Nature 421, 611 (2003)

Attosecond light sources: state of the art

Photon energies extending to 1.6 keV



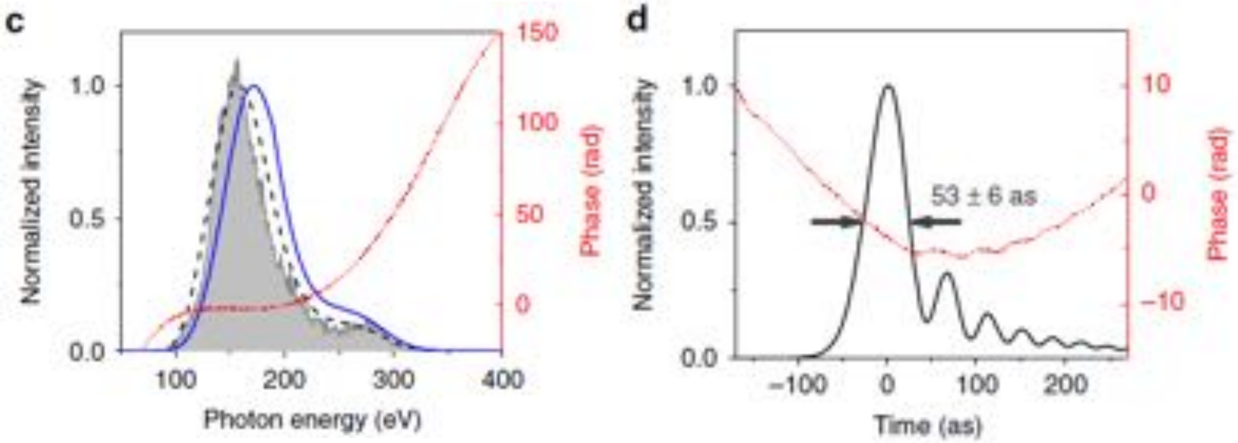
T. Popmintchev et al. *Science* 336, 1287 (2012)

700 eV bandwidth => Fourier limit: 2.5 as

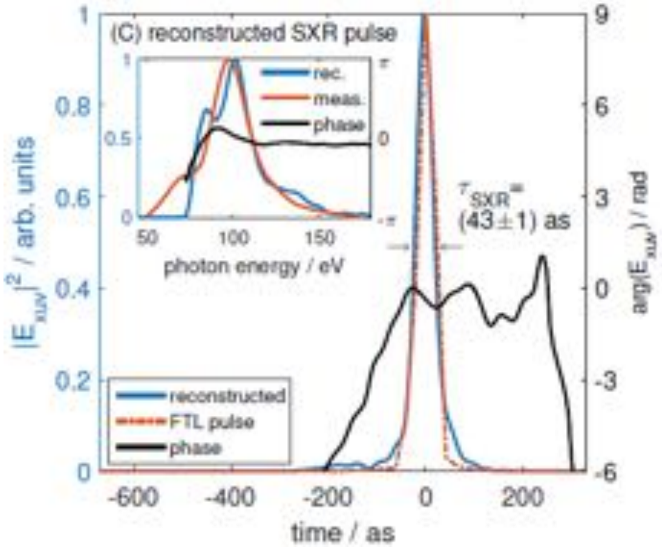
BUT: intrinsic atto-chirp => actual duration far from FT limit

Y. Mairesse et al. *Science 302, 1540 (2003)*

Shortest duration for isolated pulses of ~50 as



J. Li et al., *Nature Comm. 8, 186* (2017)



T. Gaumnitz et al., *Optics Express 25, 27506* (2017)

ATTOLab consortium

A consortium of 9 laboratories in Saclay

http://attolab.fr/

fs lasers + as XUV sources

Solid state physics

Fs laser and XUV sources with plasmas

XUV optics

Gas phase

State-of-the-art laser systems @ ATTOLab-Orme

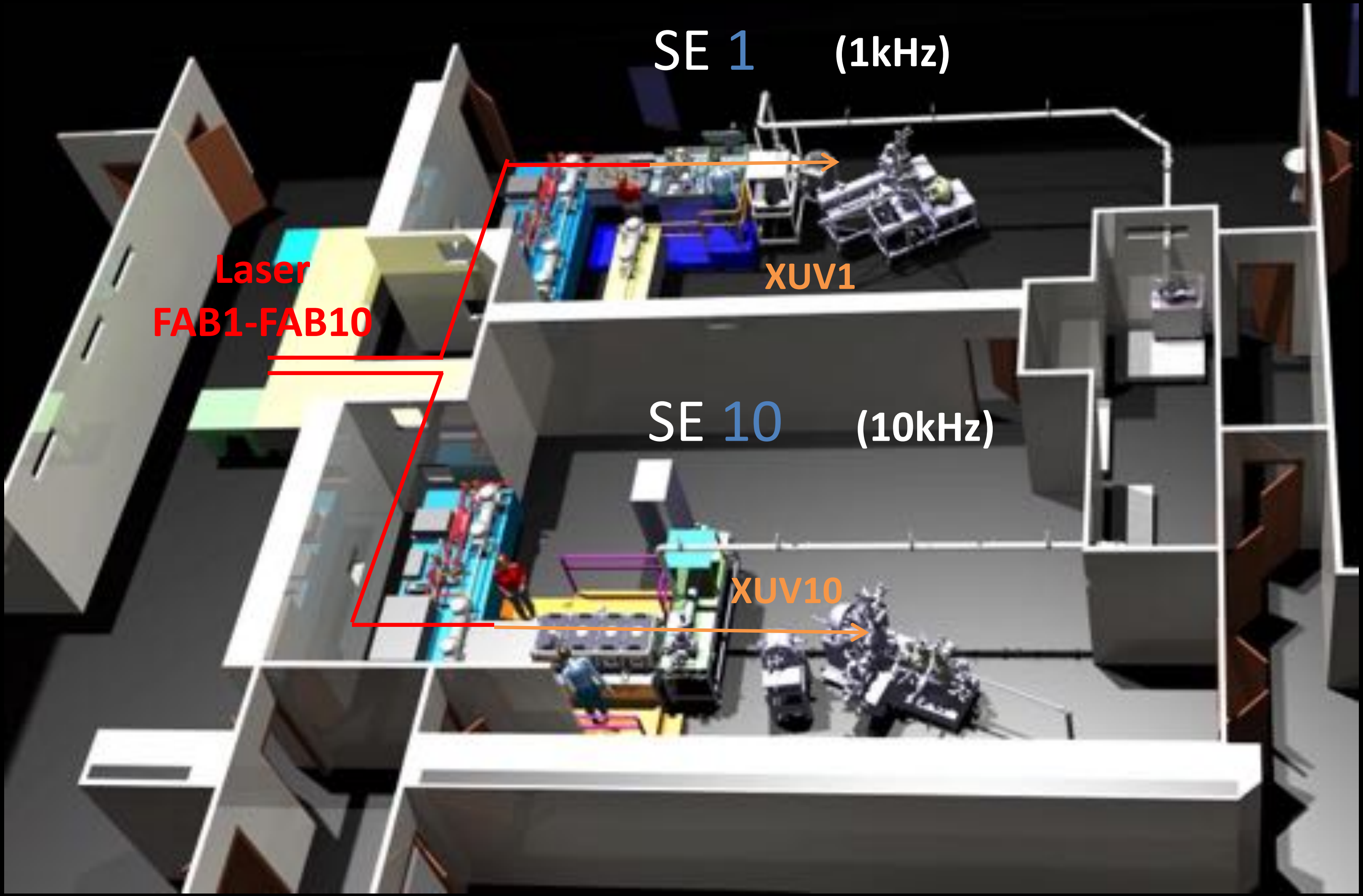
FAB 1: 15 W (15 mJ/1 kHz), 24 fs, CEP shot to shot: 350 mrad

FAB 10: 20 W (2 mJ/10 kHz), 24 fs, CEP shot to shot: 250 mrad

Titanium:Sapphire @800nm

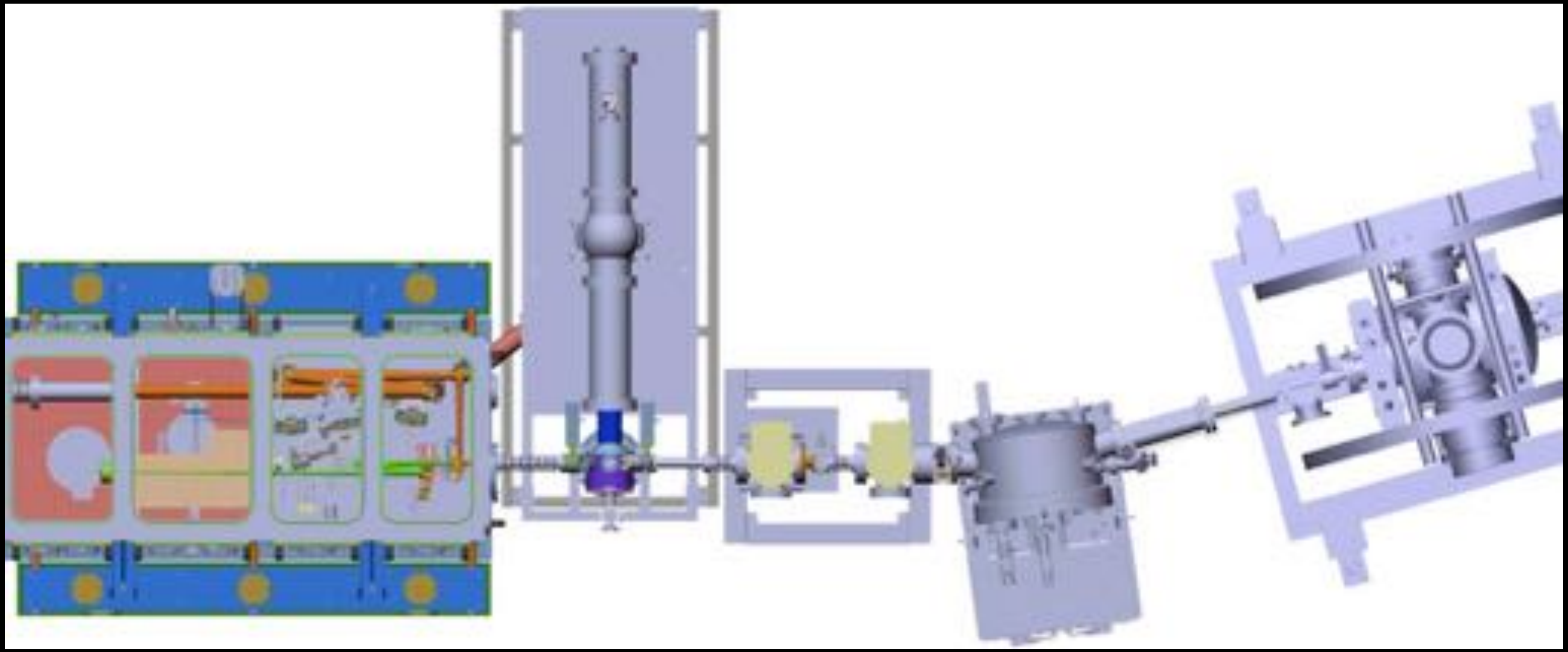
P. D'O-F. Hergott, O. Tcherbakoff *et al.*

2 (3) attosecond beamlines



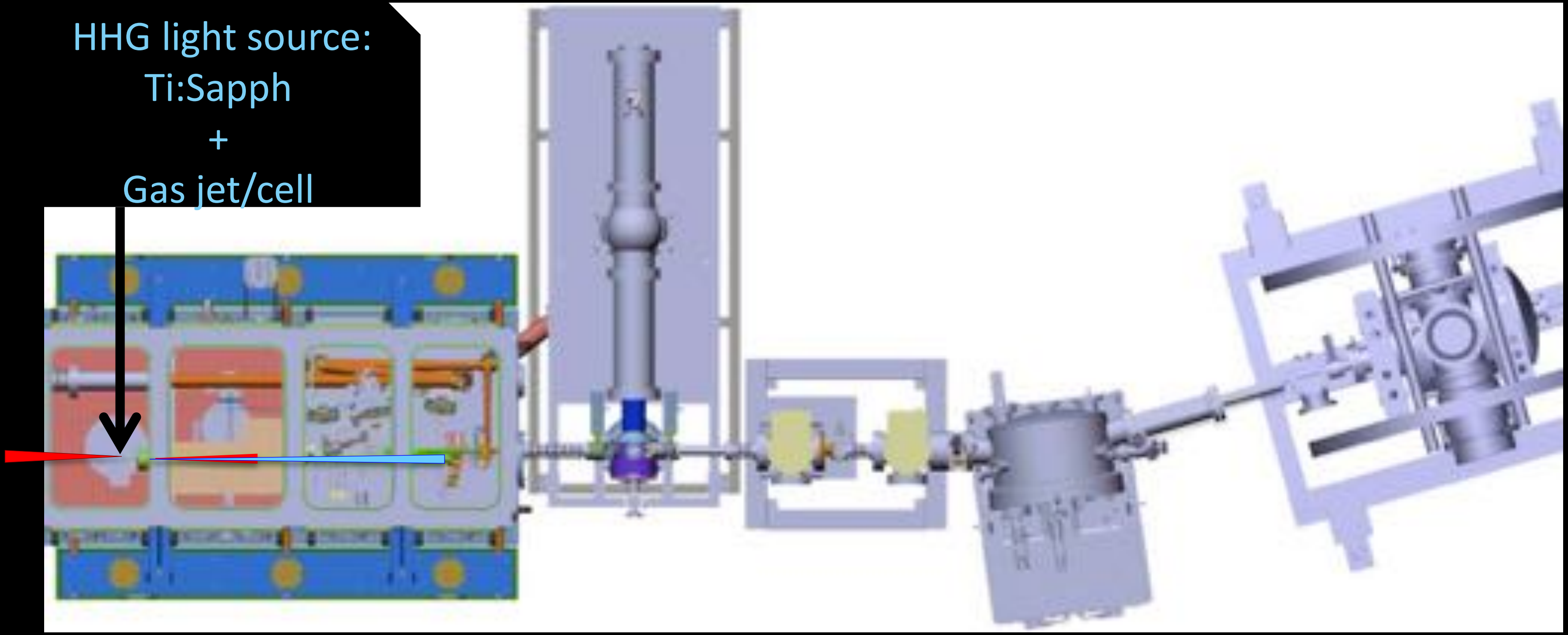
T. Ruchon, D. Bresteau, P. Salières *et al.*

SE10 high-reprate attosecond beamline



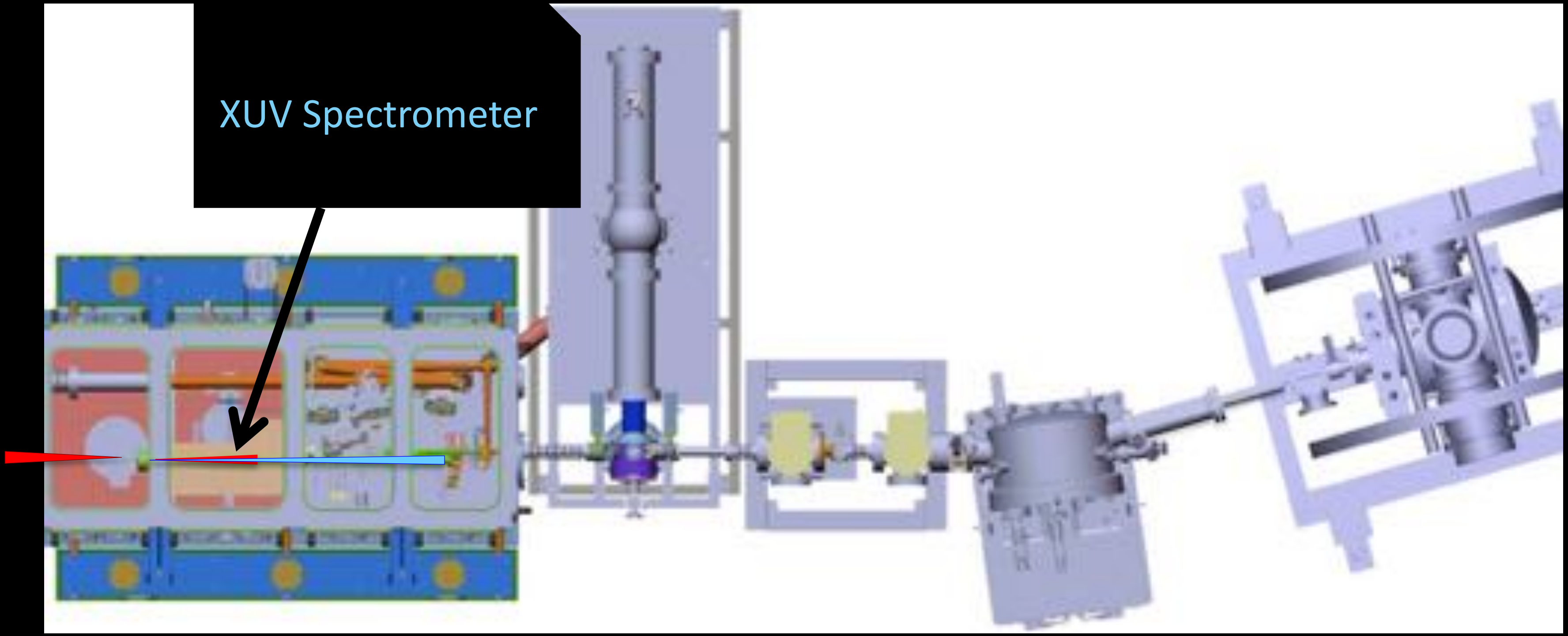
Transport, control and characterize ultrashort (0.1 - 100 fs) XUV (10 - 100 eV) pulses and carry out time resolved pump-probe applications with sub-fs resolution

SE10 high-reprate attosecond beamline



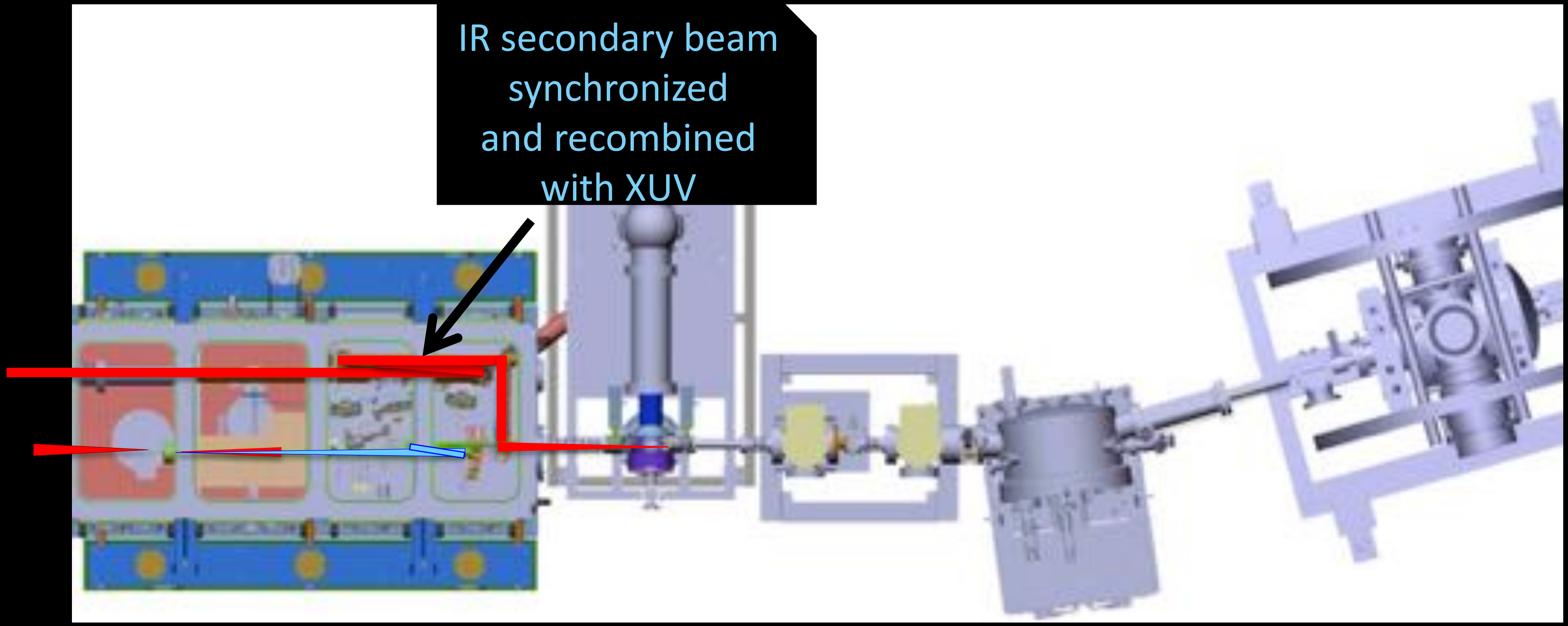
Transport, control and characterize ultrashort (0.1 - 100 fs) XUV (10 - 100 eV) pulses and carry out time resolved pump-probe applications with sub-fs resolution

SE10 high-reprate attosecond beamline



Transport, control and characterize ultrashort (0.1 - 100 fs) XUV (10 - 100 eV) pulses and carry out time resolved pump-probe applications with sub-fs resolution

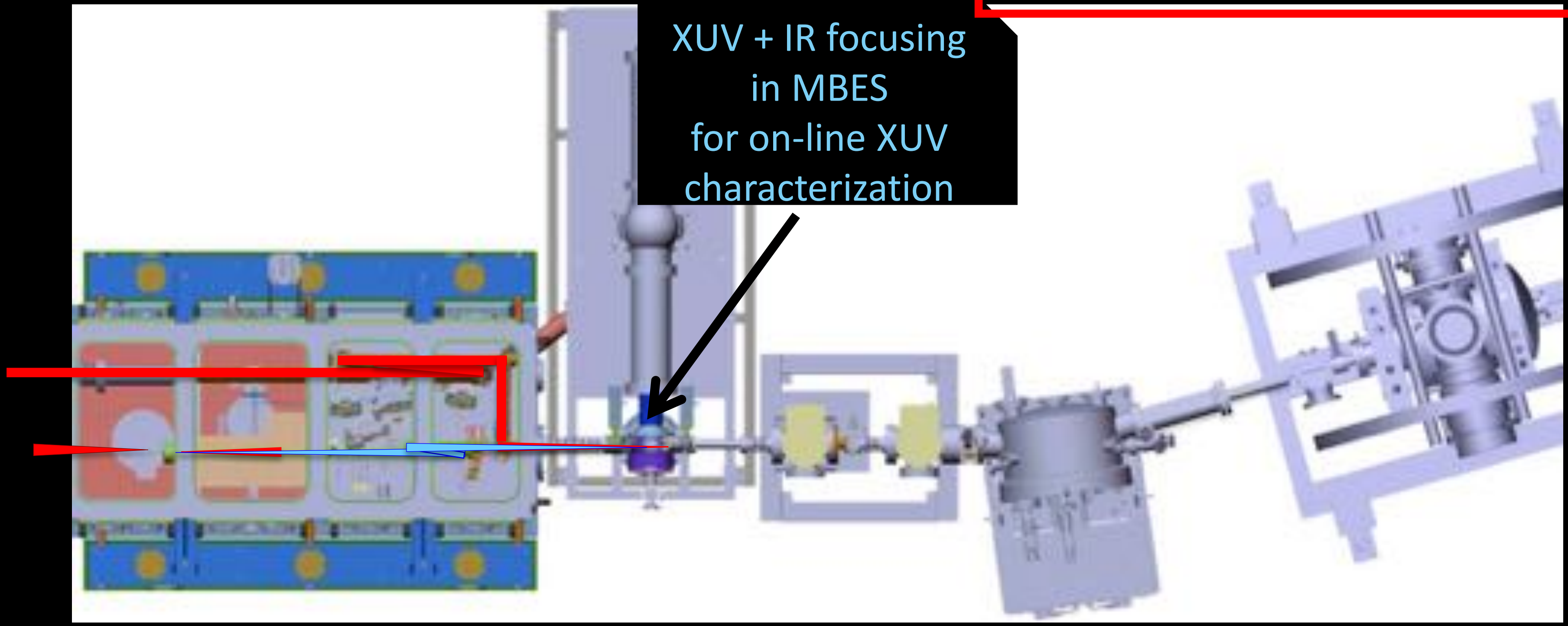
SE10 high-reprate attosecond beamline



Transport, control and characterize ultrashort (0.1 - 100 fs) XUV (10 - 100 eV) pulses and carry out time resolved pump-probe applications with sub-fs resolution

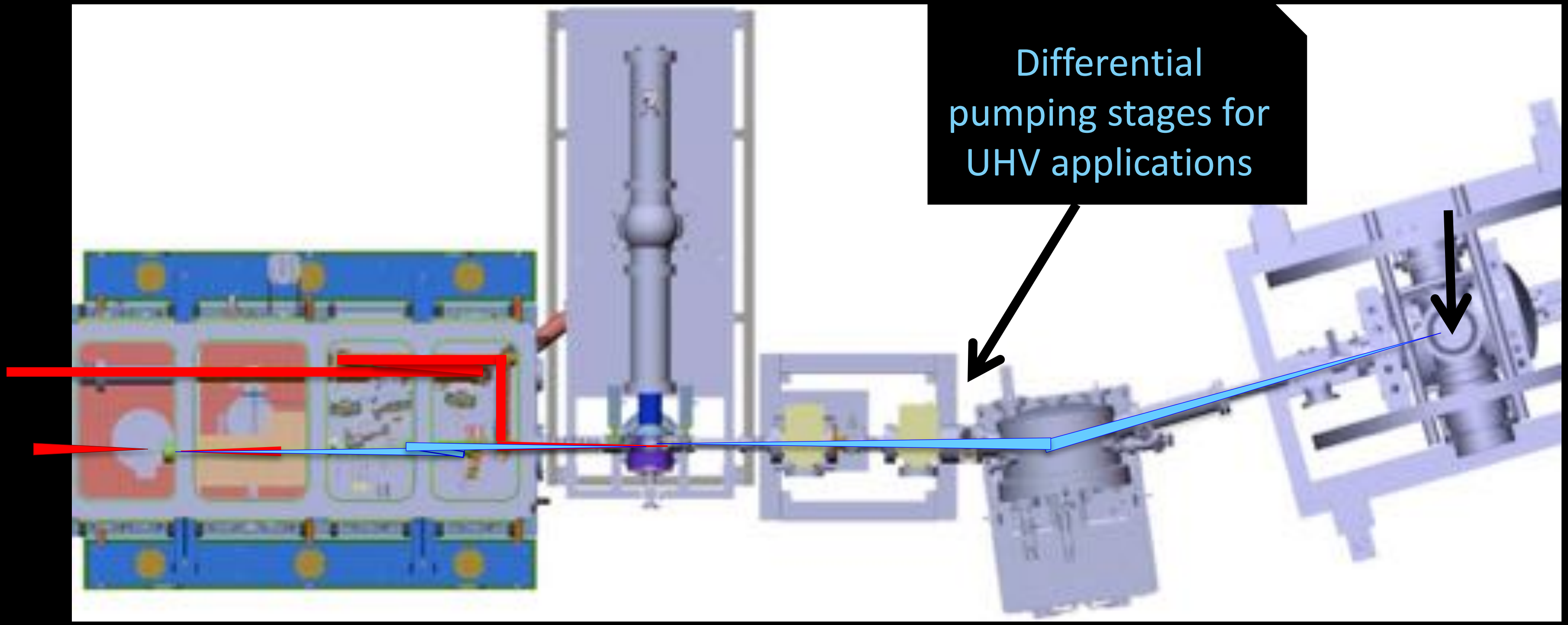
SE10 high-reprate attosecond beamline

LIZARD in-situ stabilization
Stability = 28 as RMS
M. Luttmann et al. Phys. Rev. Applied (2021)



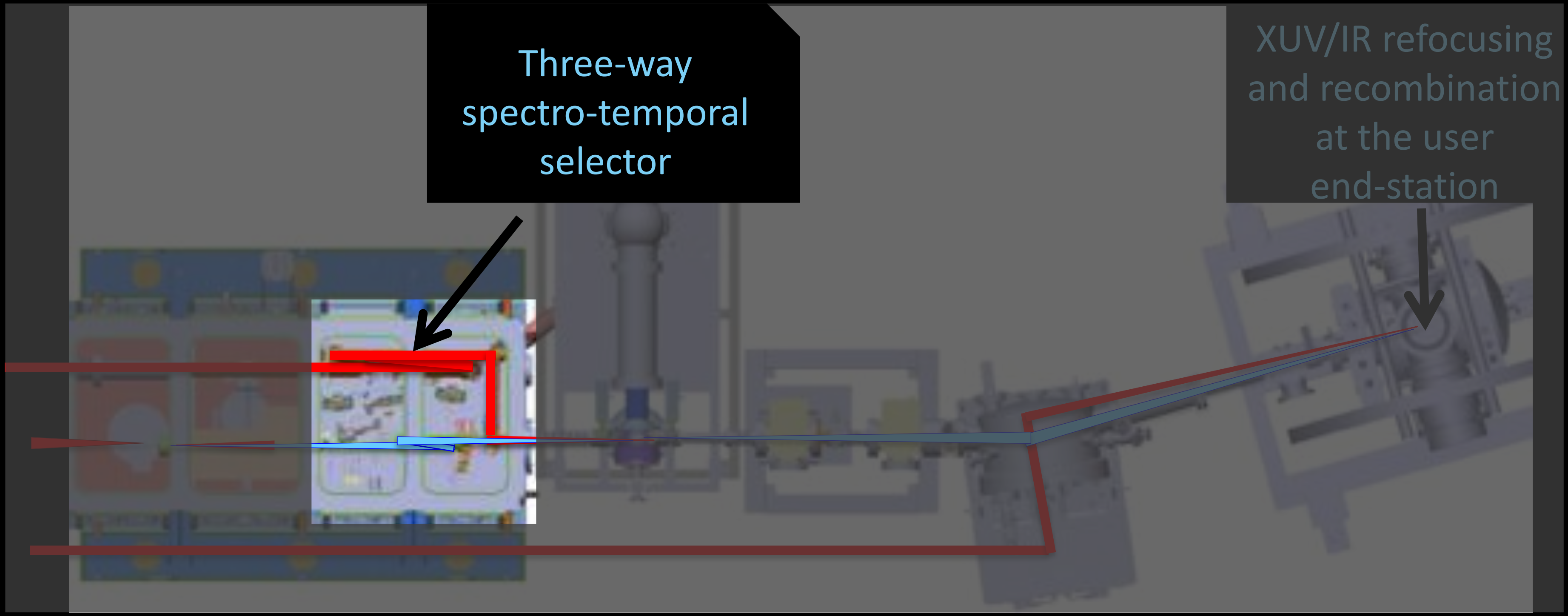
Transport, control and characterize ultrashort (0.1 - 100 fs) XUV (10 - 100 eV) pulses and carry out time resolved pump-probe applications with sub-fs resolution

SE10 high-reprate attosecond beamline



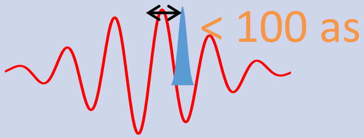
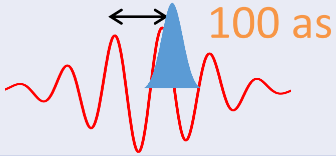
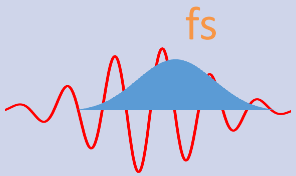
Transport, control and characterize ultrashort (0.1 - 100 fs) XUV (10 - 100 eV) pulses and carry out time resolved pump-probe applications with sub-fs resolution

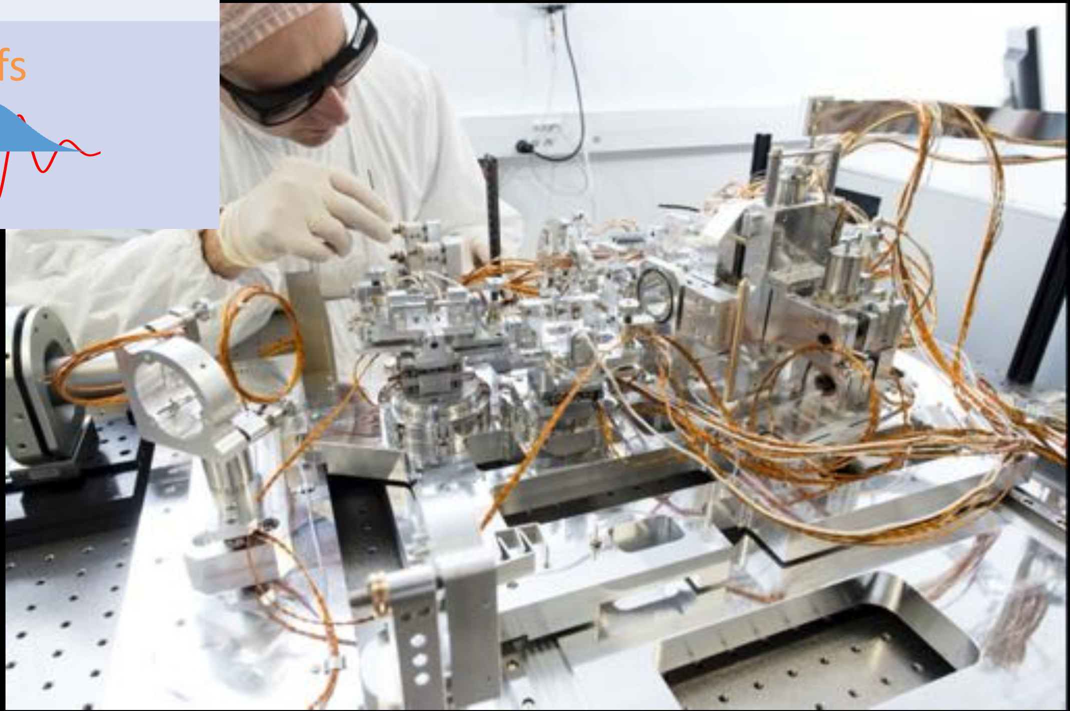
SE10 high-reprate attosecond beamline



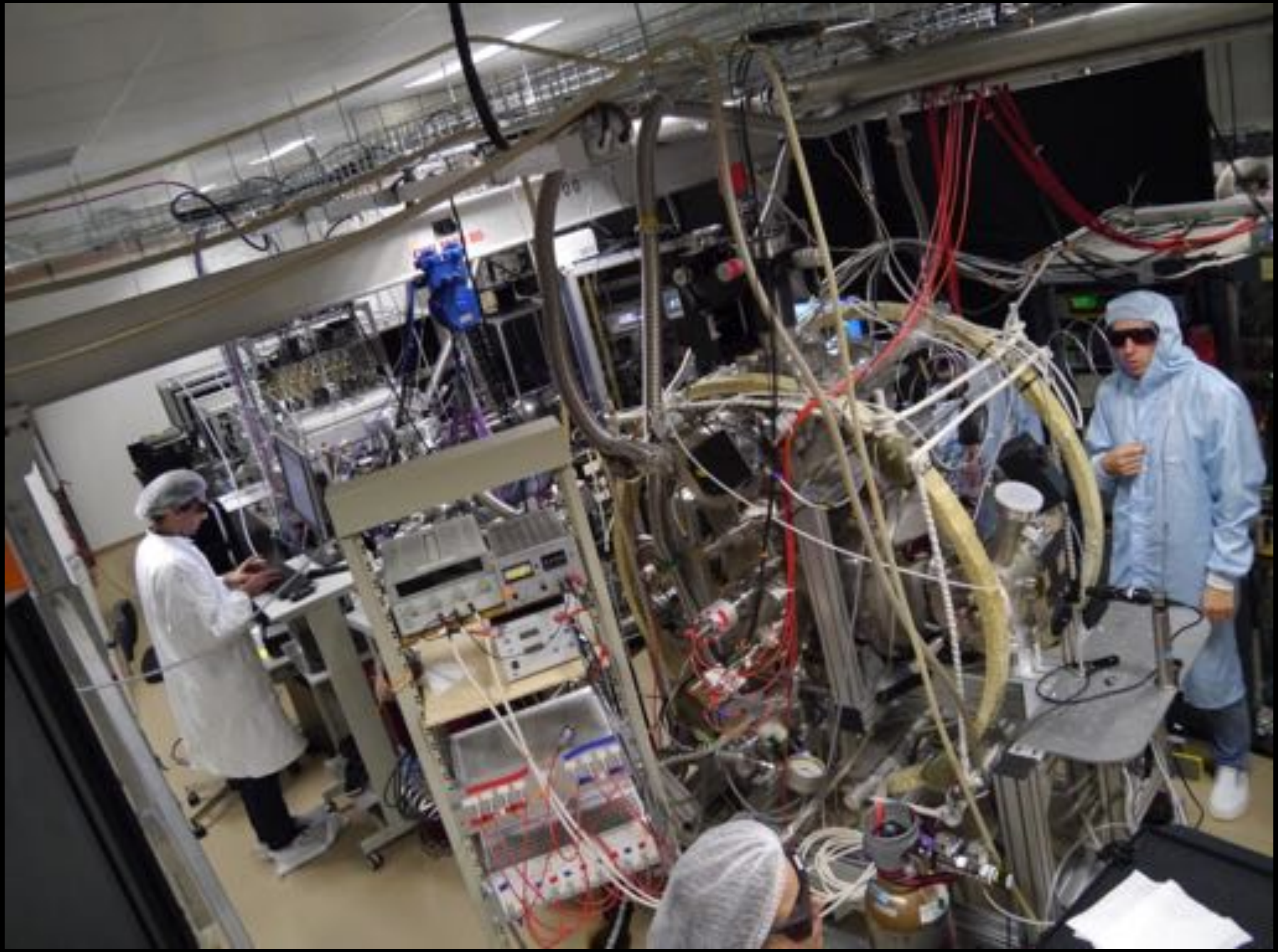
Transport, control and characterize ultrashort (0.1 - 100 fs) XUV (10 - 100 eV) pulses and carry out time resolved pump-probe applications with sub-fs resolution

XUV spectro-temporal selector

Type	XUV Δ E	XUV Δ τ	XUV / IR delay
Very broadband (VBB)	10-30 eV	100 as	
Broadband (BB)	1-5 eV	1 fs	
Narrowband (NB)	100 meV	10 fs	

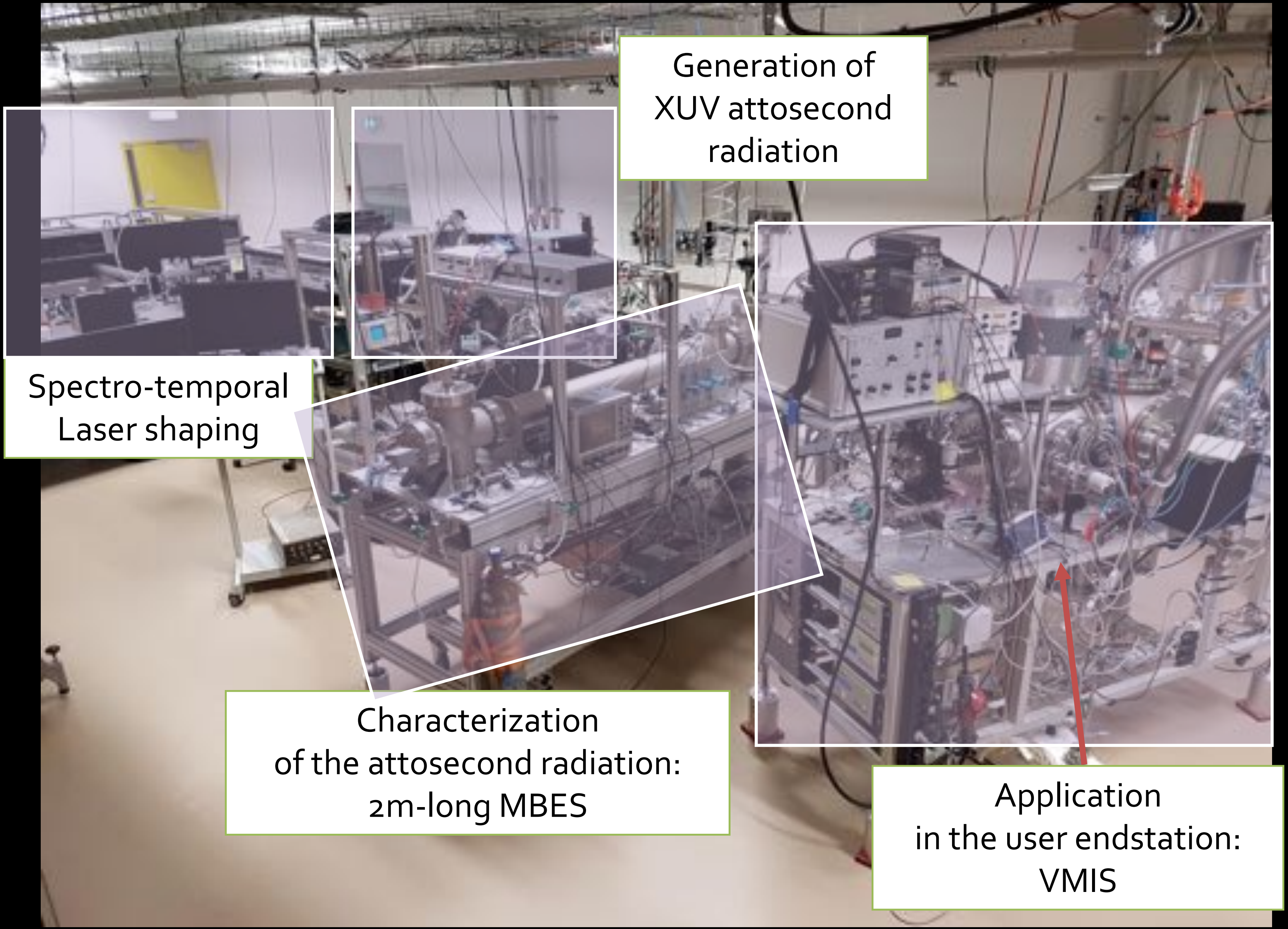


SE10 high-reprate attosecond beamline



SE1 high-energy attosecond beamline

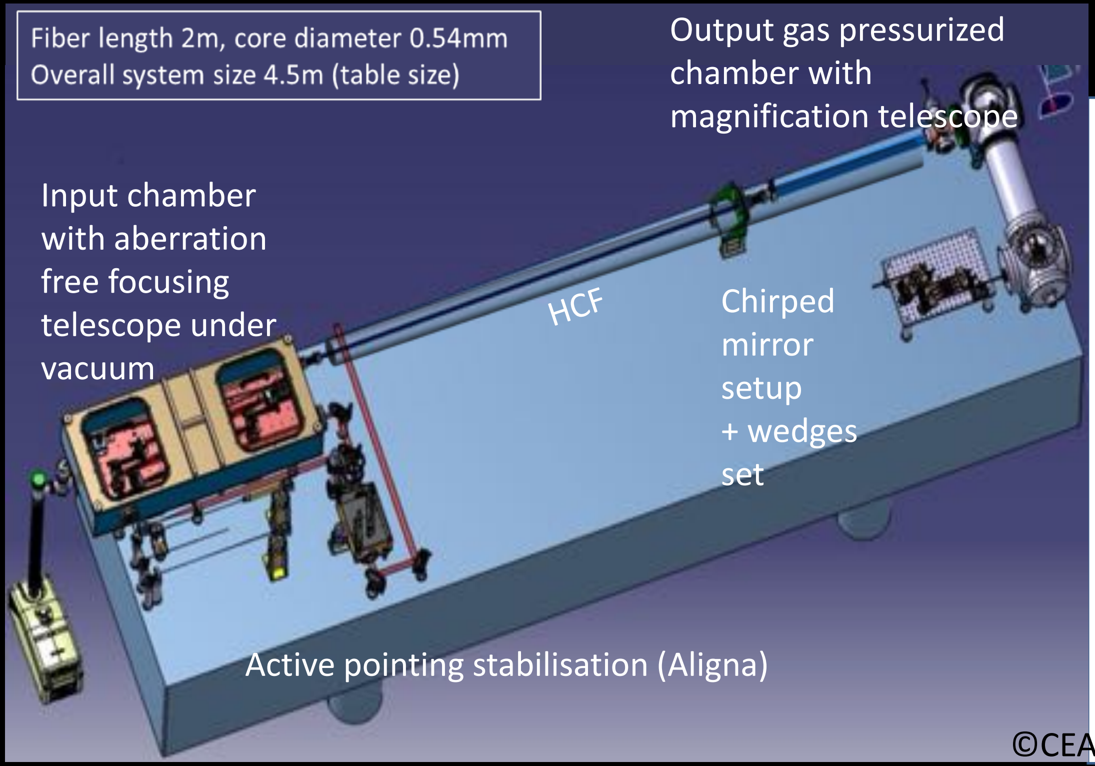
SE1 high-energy attosecond beamline



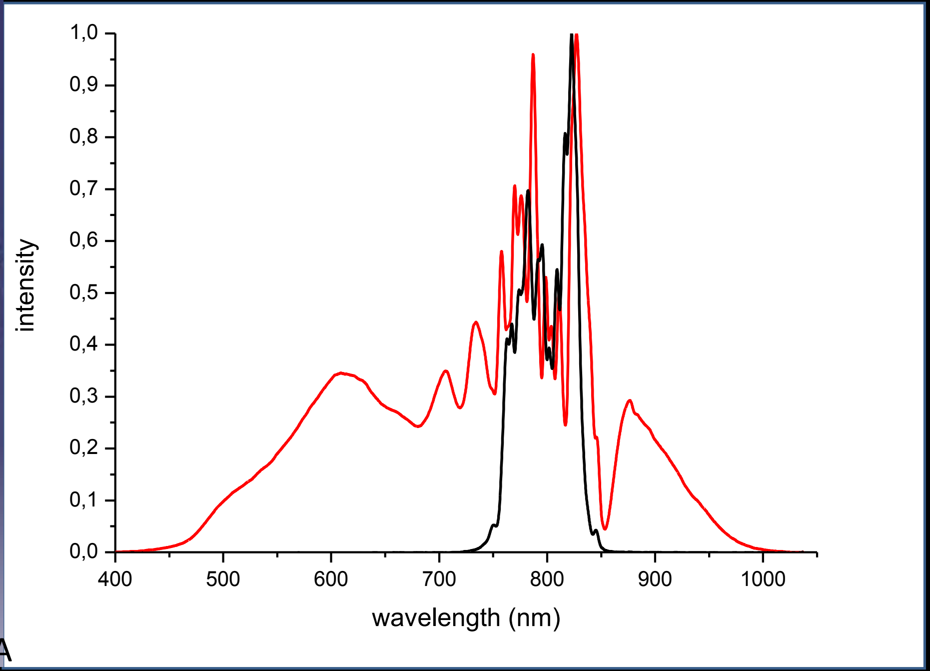
Stretched hollow-core fiber postcompression

F. Réau, F. Lepetit, O. Tcherbakoff & JF Hergott

HCF system from Laser Laboratum Gottingen P. Simon T. Nagy
Home made set-up design (discussions with) R. Lopez Martens



Input max energy:	**5mJ**
Fiber coupling (with gas):	68%
Output optics transmission including chirped mirror compressor:	79%
Max output Energy:	**2.7mJ**

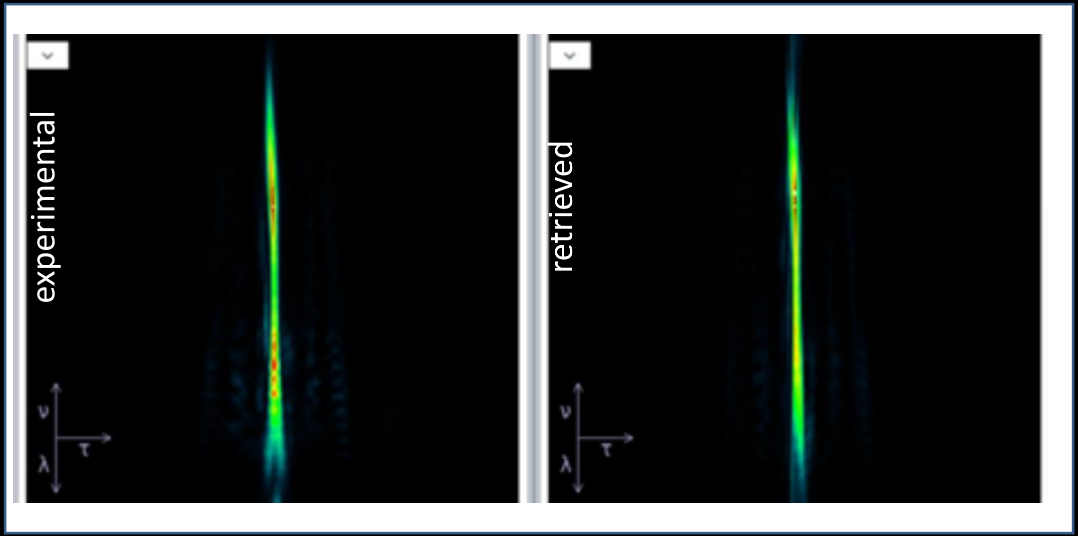


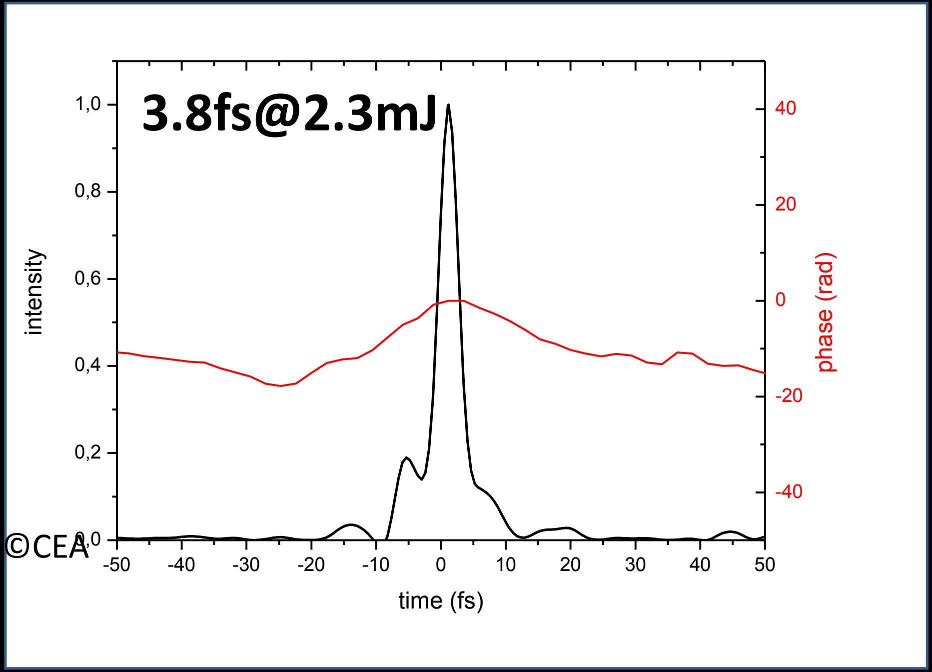
Stretched hollow-core fiber postcompression

F. Réau, F. Lepetit, O. Tcherbakoff & JF Hergott

Input max energy:	**5mJ**
Fiber coupling (with gas):	68%
Output optics transmission including chirped mirror compressor:	79%
Max output Energy:	**2.7mJ**

FEMTOEASY FROG-FC measurement





Attosecond photoionization spectroscopy

The photoelectric effect

H. Hertz, 1887

Experimental evidence

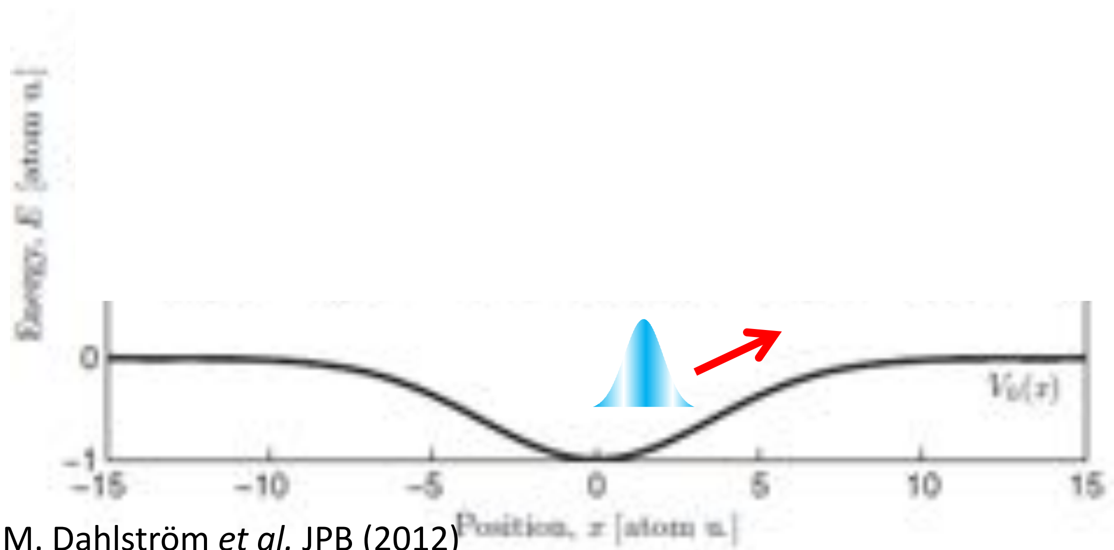
A. Einstein, 1905

Interpretation with the photon concept
Nobel prize 1921

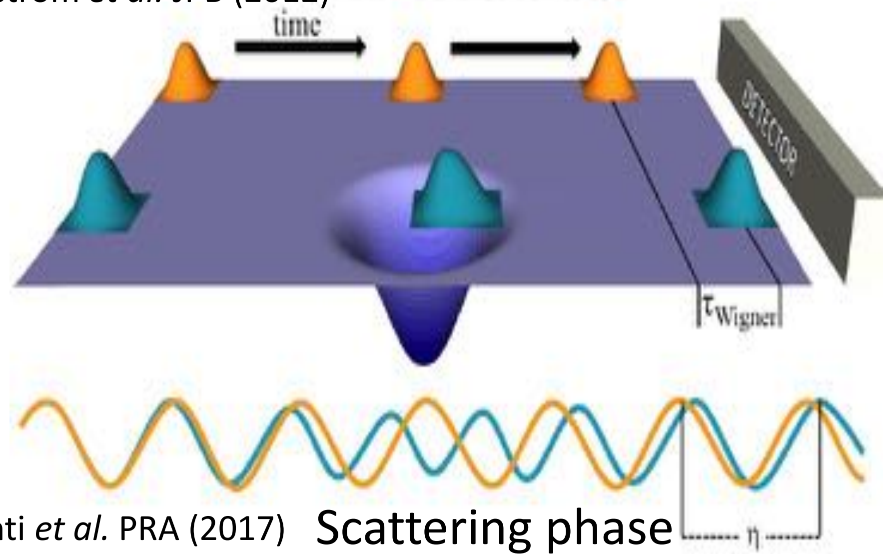
The absorption of a quantum of energy (photon) is accompanied by the quasi-simultaneous emission of an electron

Attosecond spectroscopy: accessing photoemission delays

- How long does it take to remove one electron from a system?

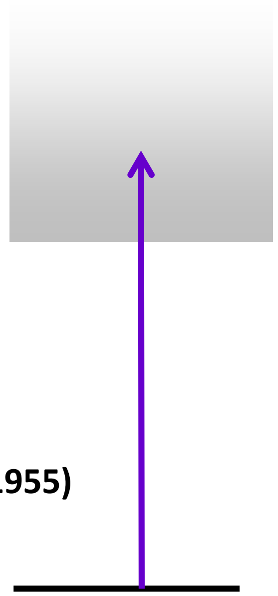


M. Dahlström *et al.* JPB (2012)



L. Argenti *et al.* PRA (2017) Scattering phase

E. P. Wigner, Phys. Rev. (1955)



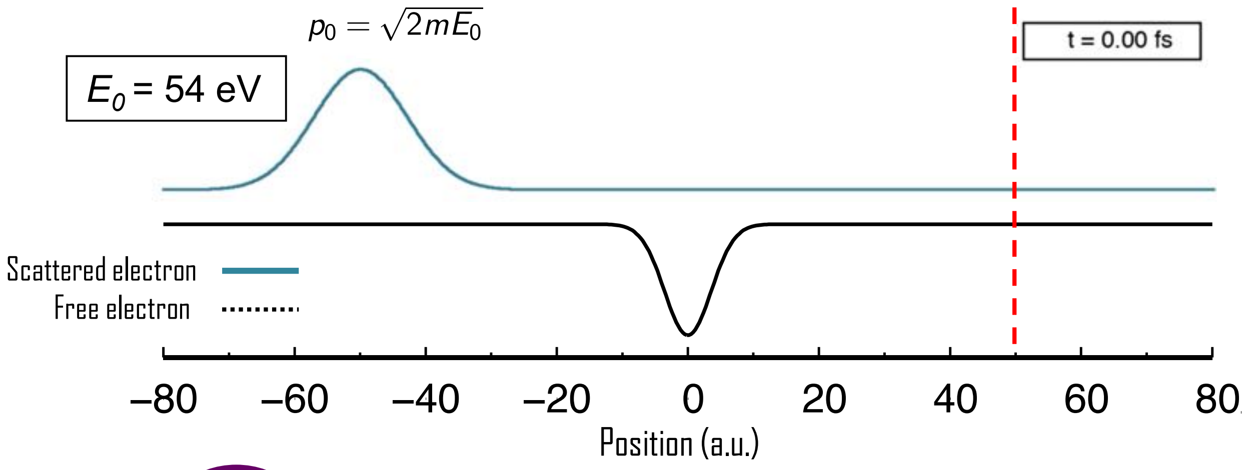
Scattering delay:
(Wigner time delay)

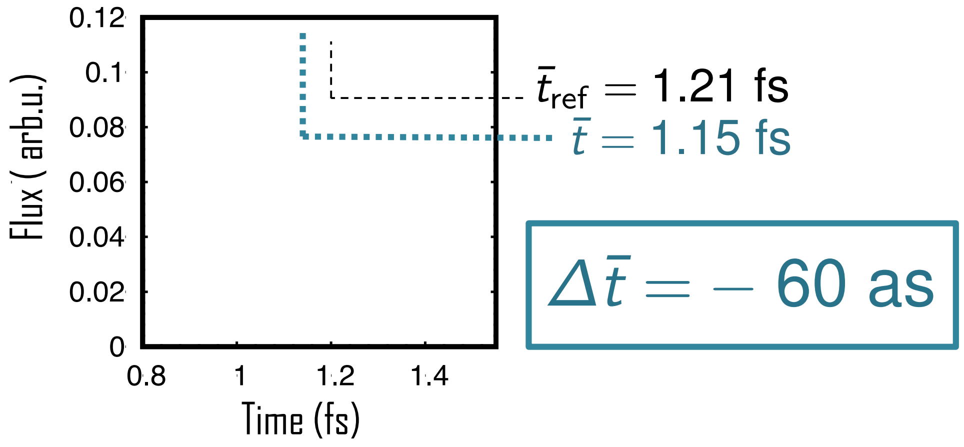
=

Ionization ė half-scattering

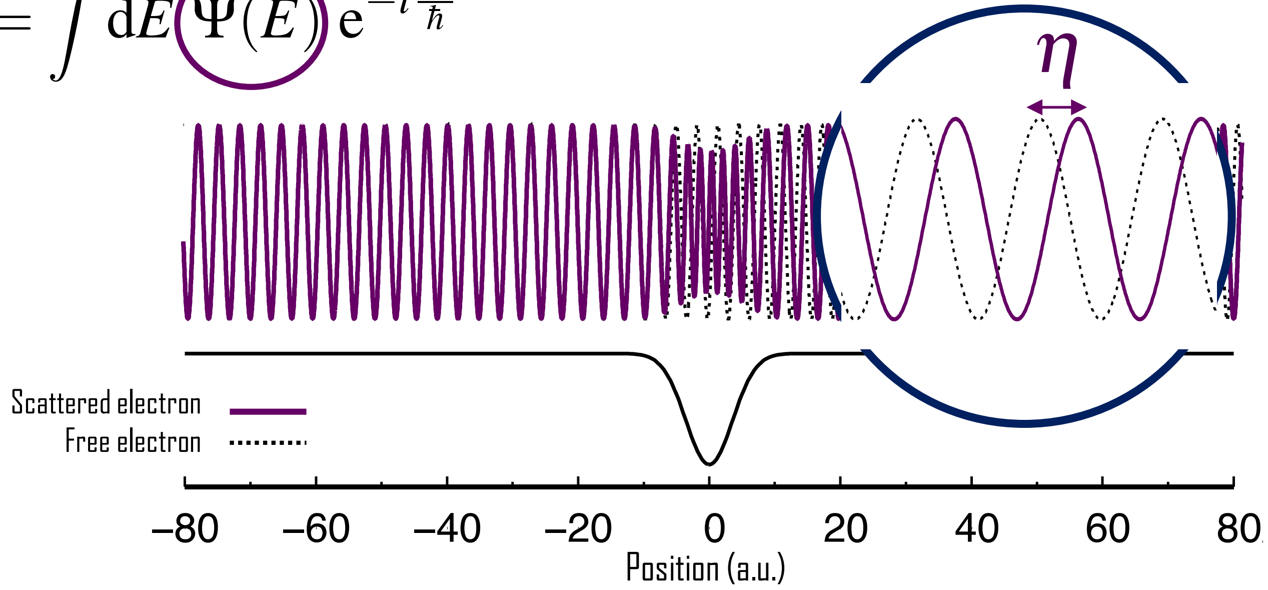
Scattering delays and phase shifts

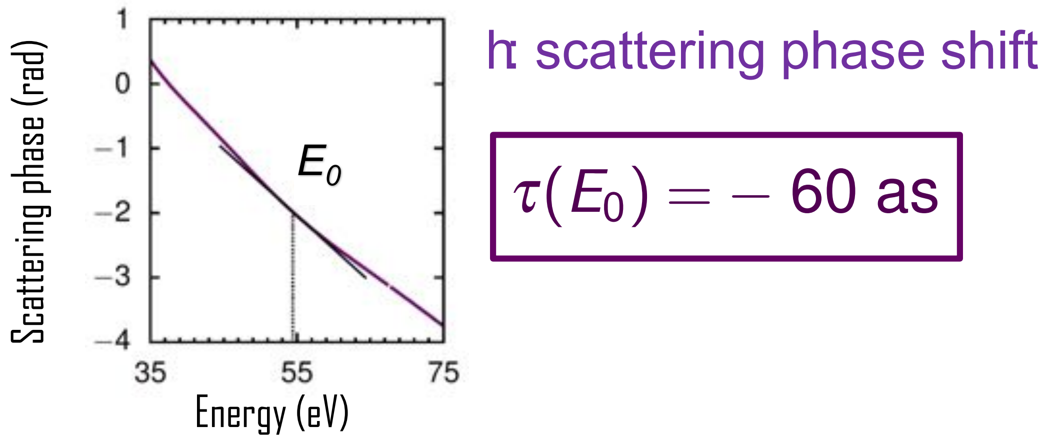
Simulation : Basile Wurmser (LCPMR)



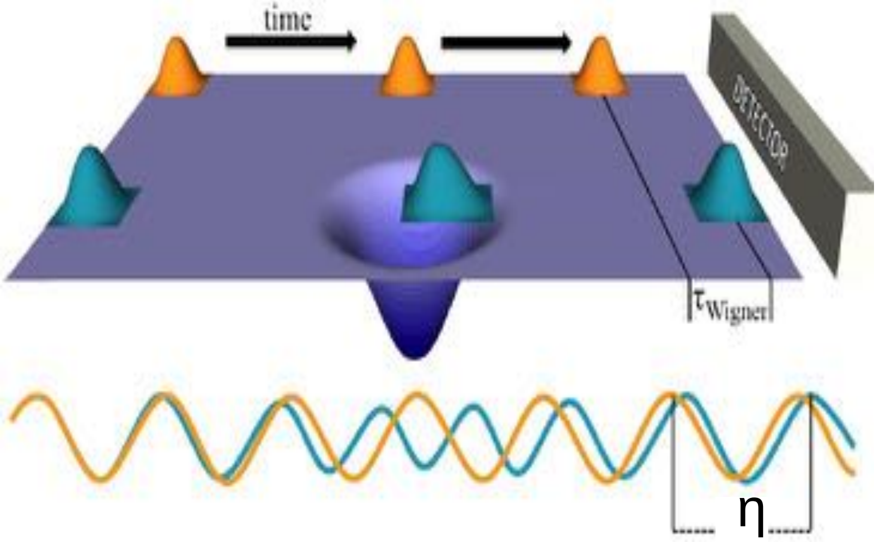


$$\psi(t) = \int \mathrm{d}E\, \Psi(E)\, \mathrm{e}^{-i\frac{Et}{\hbar}}$$





Accessing ionization delays



Scattering phase

We need:

1) a reference

Ý Difference between ionization shells, atomic species, excitation energies…

2) **An utrafast temporal technique:**

=> Direct access to

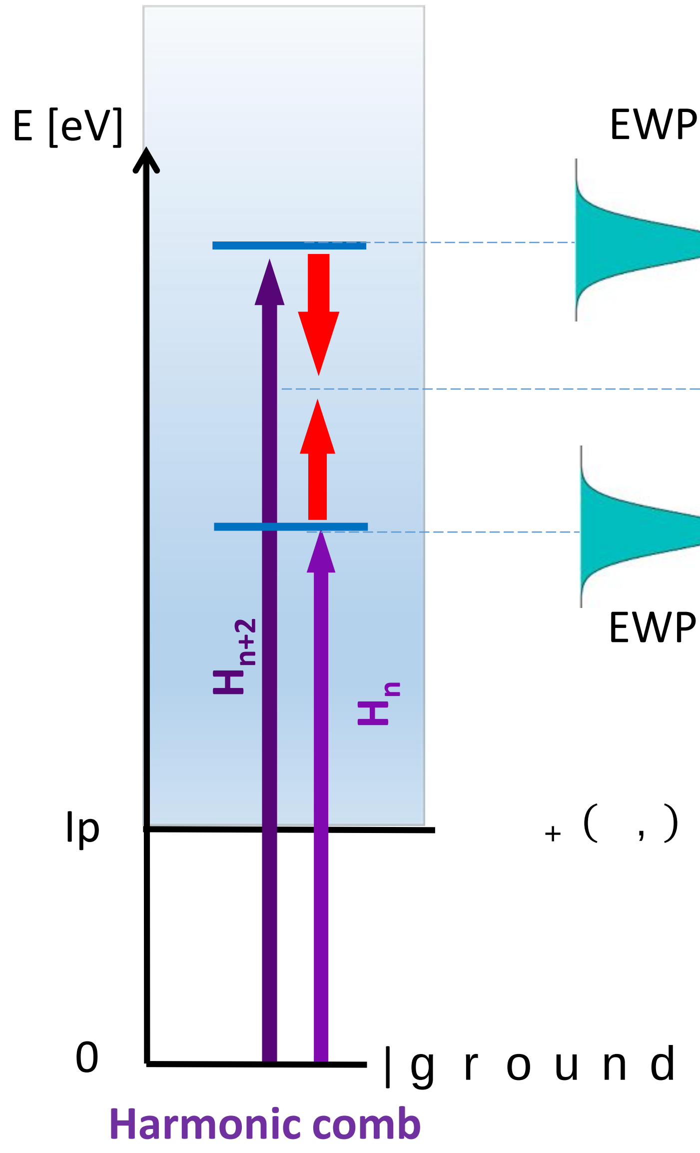
Single atto-pulse 'pump' + IR field 'probe'
(attosecond streaking)

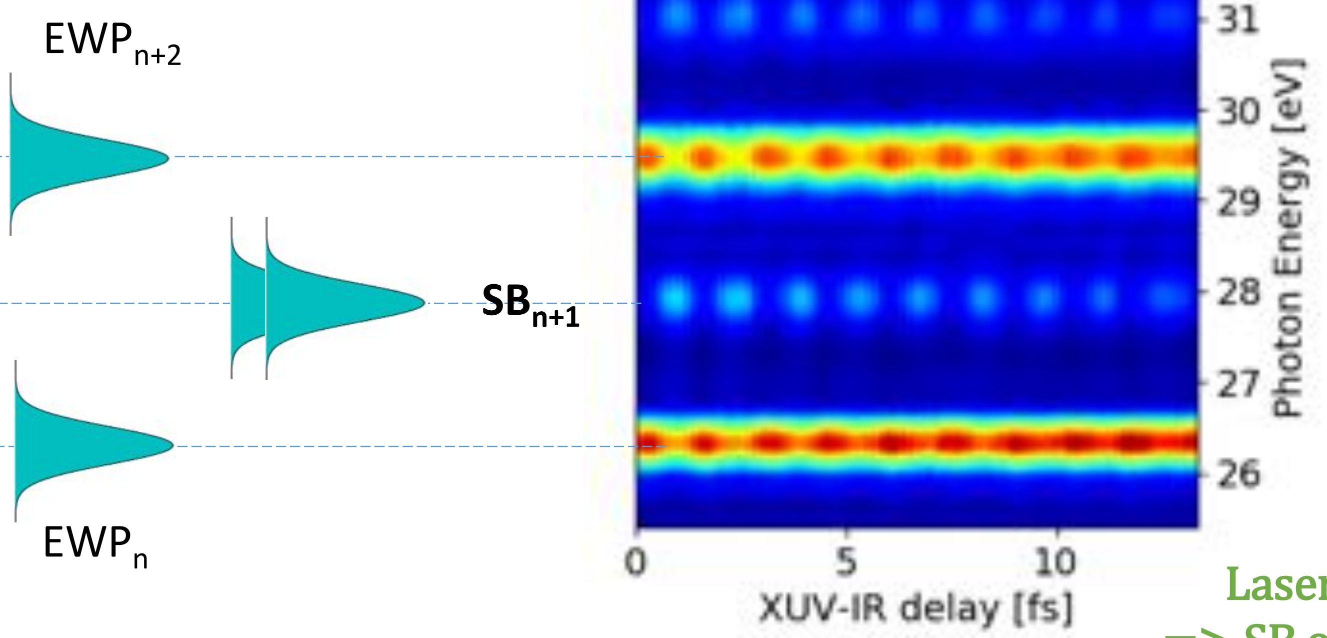
Or a broadband interferometric spectral technique:

=> Access to the spectral variation of η

Atto-pulse train + IR field (RABBIT technique)

RABBIT: Reconstruction of Attosecond Beating By Interference of two-photon Transitions





=> Attosecond metrology

Paul *et al.*, Science (2001)
Mairesse *et al.*, Science (2003)

Harmonic phase

+ ( ) − ( )

Laser phase
=> SB oscillations

Atomic phase

+ (,) = | ( ) ( )+ ( )| + | ( ) (+)− ( )| + | ( ) ( )+ ( )|| ( ) (+)− ( )| + + ( ) + ( )

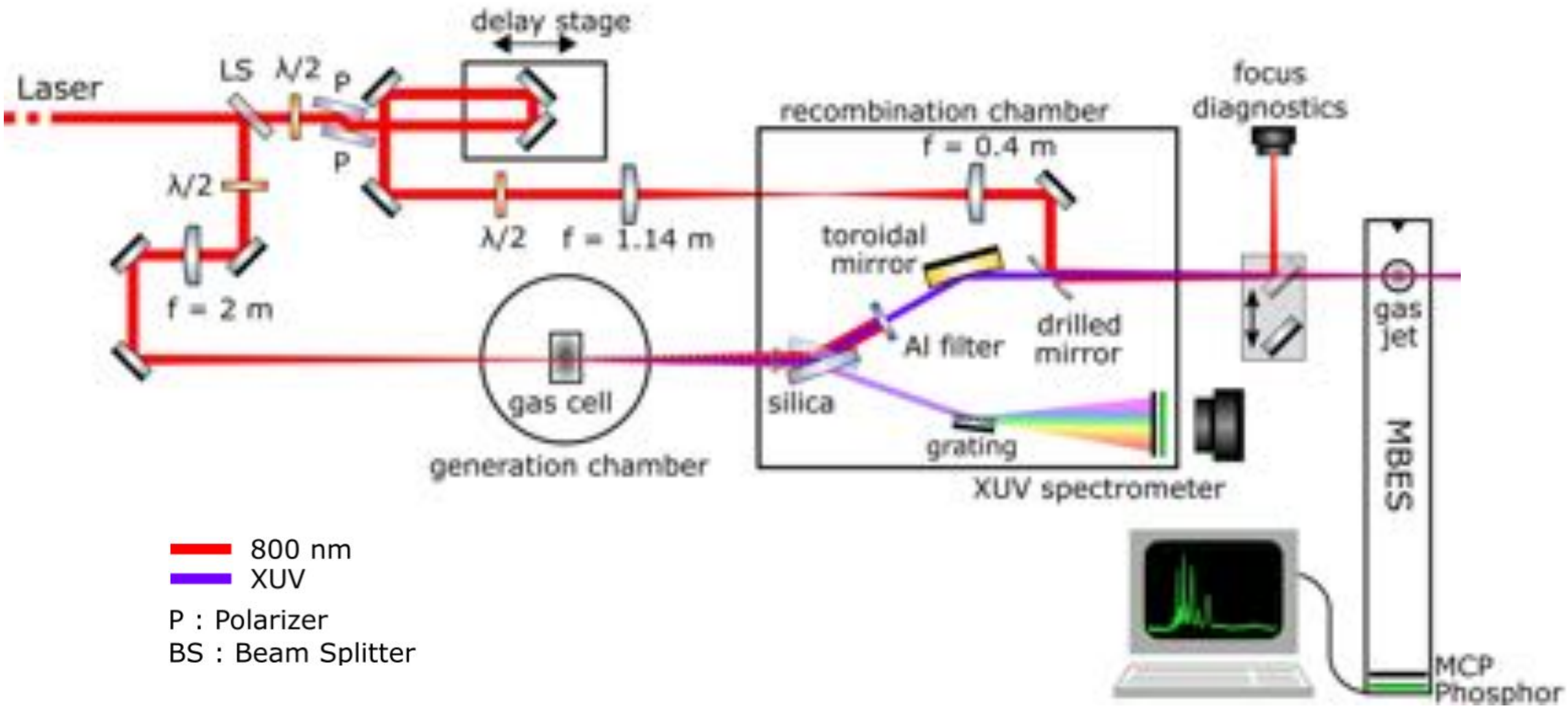
=> Attosecond spectroscopy

( ) ≈ ( ) + ( )

Direct access to the ionization delays:

τ ( ) ≈ τ ( ) + τ ( )

Experimental setup

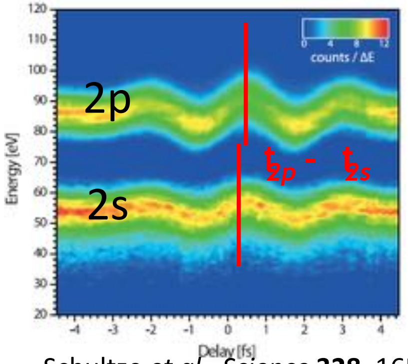


Angularly-integrated RABBIT spectrogram

Photo-ionization time delays

Atoms

Schultze *et al.*, **Science** (2010)
Klünder *et al.*, **PRL** (2011)
Dahlström *et al.*, **JPB** (2012)
Guénot *et al.*, **PRA** (2012)
Dahlström *et al.*, **Chem. Phys.** (2013)
Guénot *et al.*, **JPB** (2014)
Palatchi *et al.*, **JPB** (2014)
Sabbar *et al.*, **PRL** (2015)
Dahlström *et al.*, **JPB** (2016)
Kotur *et al.*, **Nat. Comm.** (2016)
Gruson *et al.*, **Science** (2016)
Heuser *et al.*, **PRA** (2016)
Ossiander *et al.*, **Nat. Phys.** (2017)
Jordan *et al.*, **PRA** (2017)
Isinger *et al.*, **Science** (2017)
Cirelli *et al.* **Nat. Comm.** (2018)
Joseph *et al.*, **JPB** (2020)
Turconi *et al.*, **JPB** (2020)

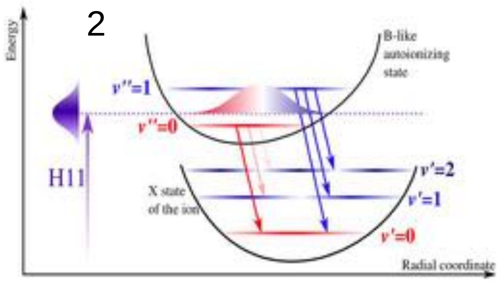


Neon

Schultze *et al.*, *Science* **328**, 1659 (2010)

Molecules

Haessler *et al.*, **PRA** (2009)
Caillat *et al.*, **PRL** (2011)
Hockett *et al.*, **JPB** (2016)
Huppert *et al.*, **PRL** (2016)
Beaulieu *et al.*, **Science** (2017)
Cattaneo *et al.*, **Nat. Phys.** (2018)
Vos *et al.* **Science** (2018)
Biswas *et al.*, **Nat. Phys.** (2020)
Loriot *et al.*, **J. Phys. Phot.** (2020)
Nandi *et al.*, **Sci. Adv.** (2020)

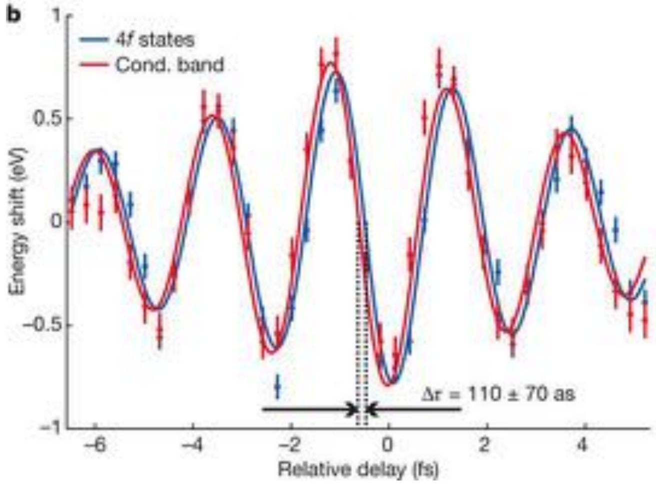


Haessler *et al.*, PRA (2009)

Solids

Cavalieri *et al.*, **Nature** (2007)
Neppl *et al.*, **PRL** (2012)
Neppl *et al.*, **Nature** (2015)
Tao *et al.*, **Science** (2016)
Ossiander *et al.*, **Nature** (2018)

Tungsten t_{CB} - t_{core}



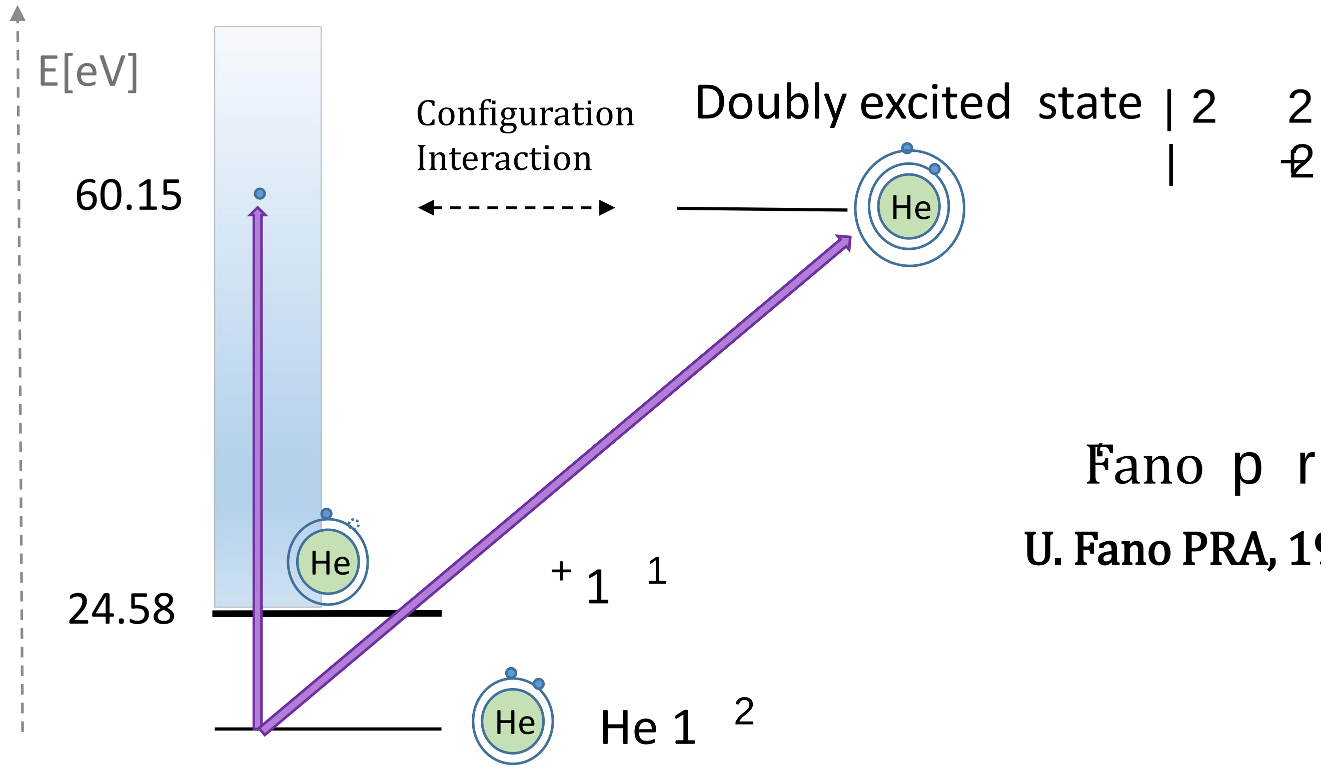
Cavalieri *et al.* Nature (2007)

- Ionization dynamics close to resonances?

=> Extremely sensitive to fine details of the potential, electron correlation…

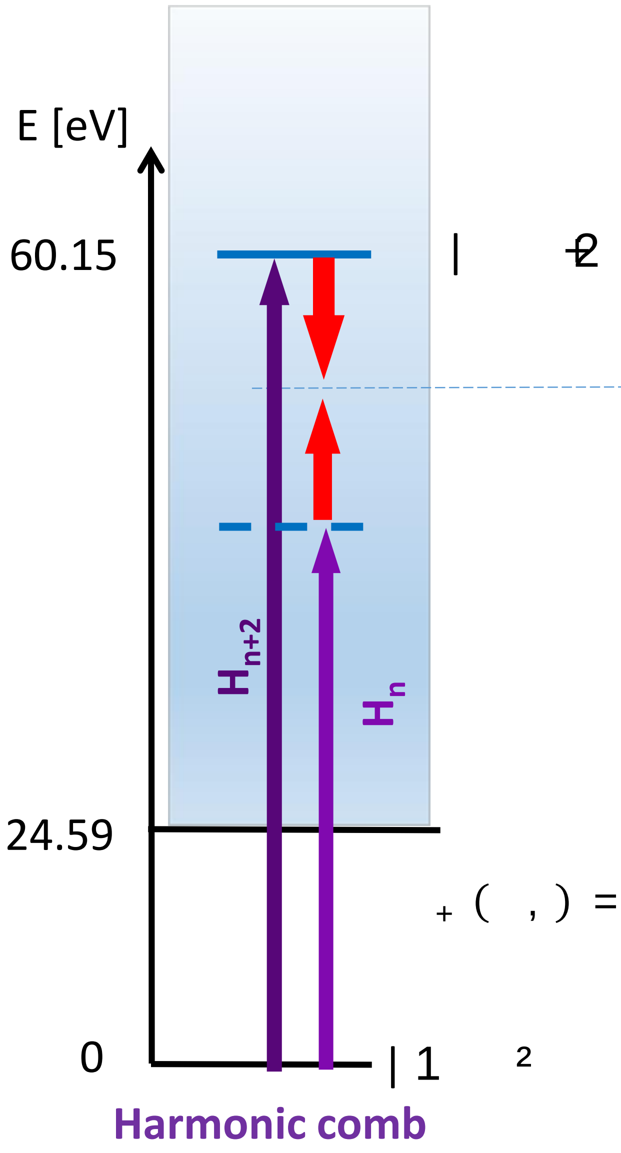
=> Fast phase variation: dynamics cannot be described any more by a simple group delay

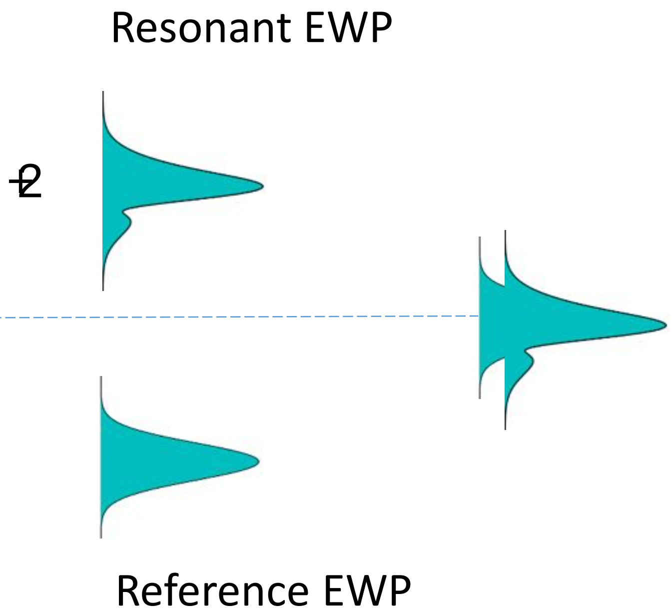
Autoionization in Helium

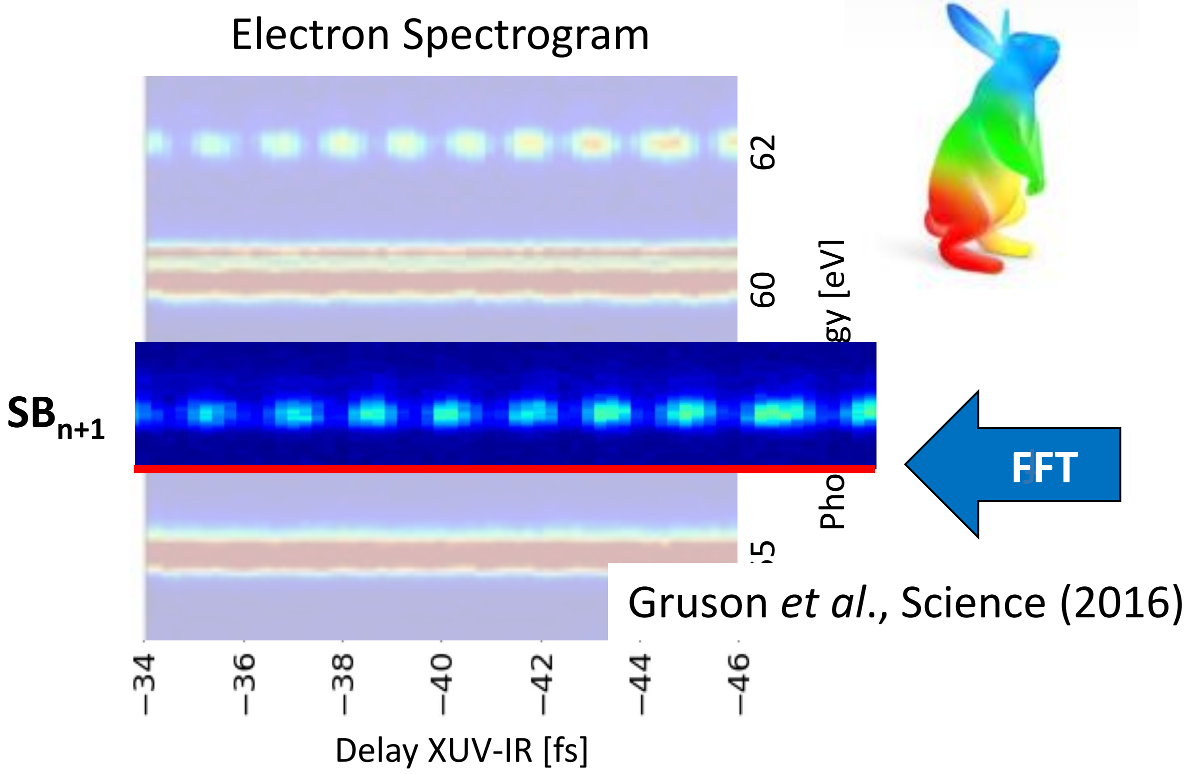


Spectrally resolved electron interferometry

RAINBOW RABBIT







Gruson *et al.*, Science (2016)

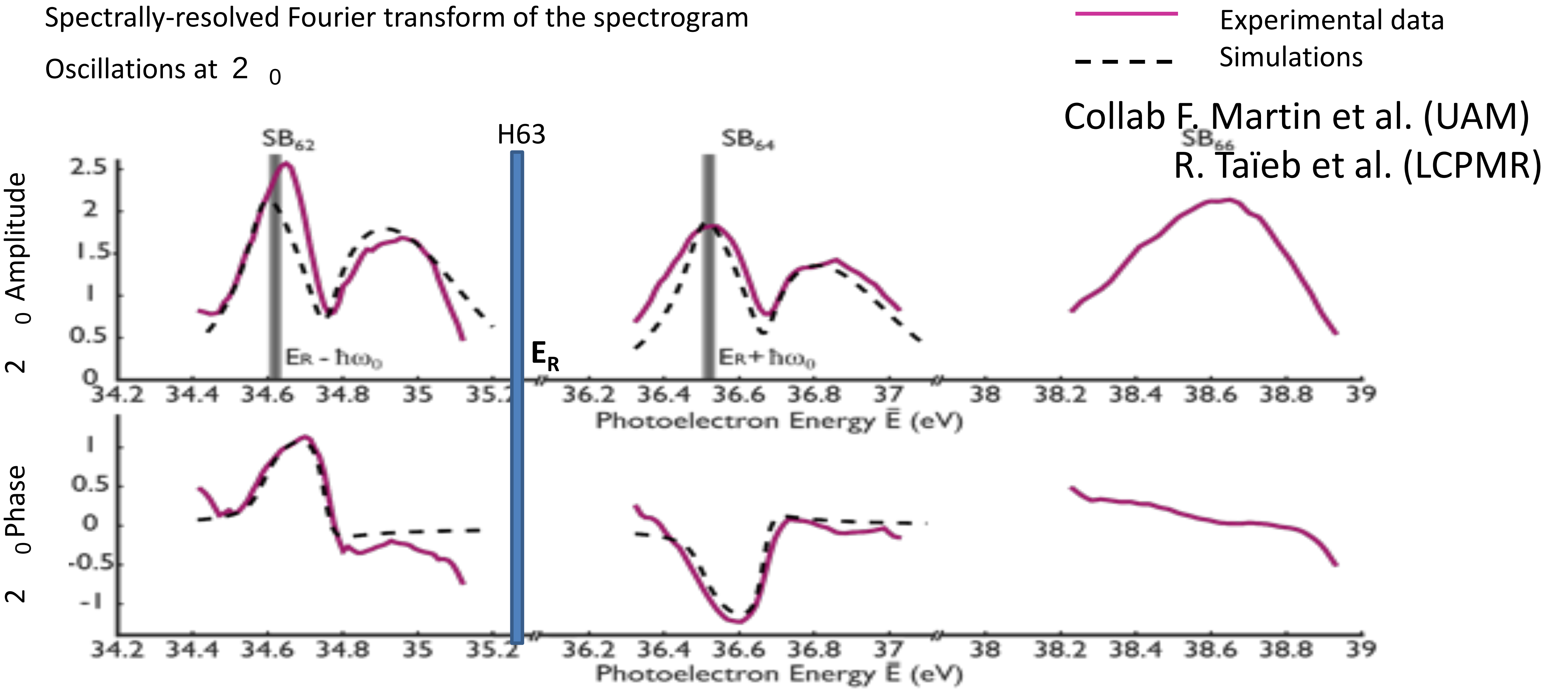
RABBIT+Scan the laser frequency:

Kotur *et al.*, Nat. Comm. (2016)

Spectral amplitude and phase measurements

Spectrally-resolved Fourier transform of the spectrogram

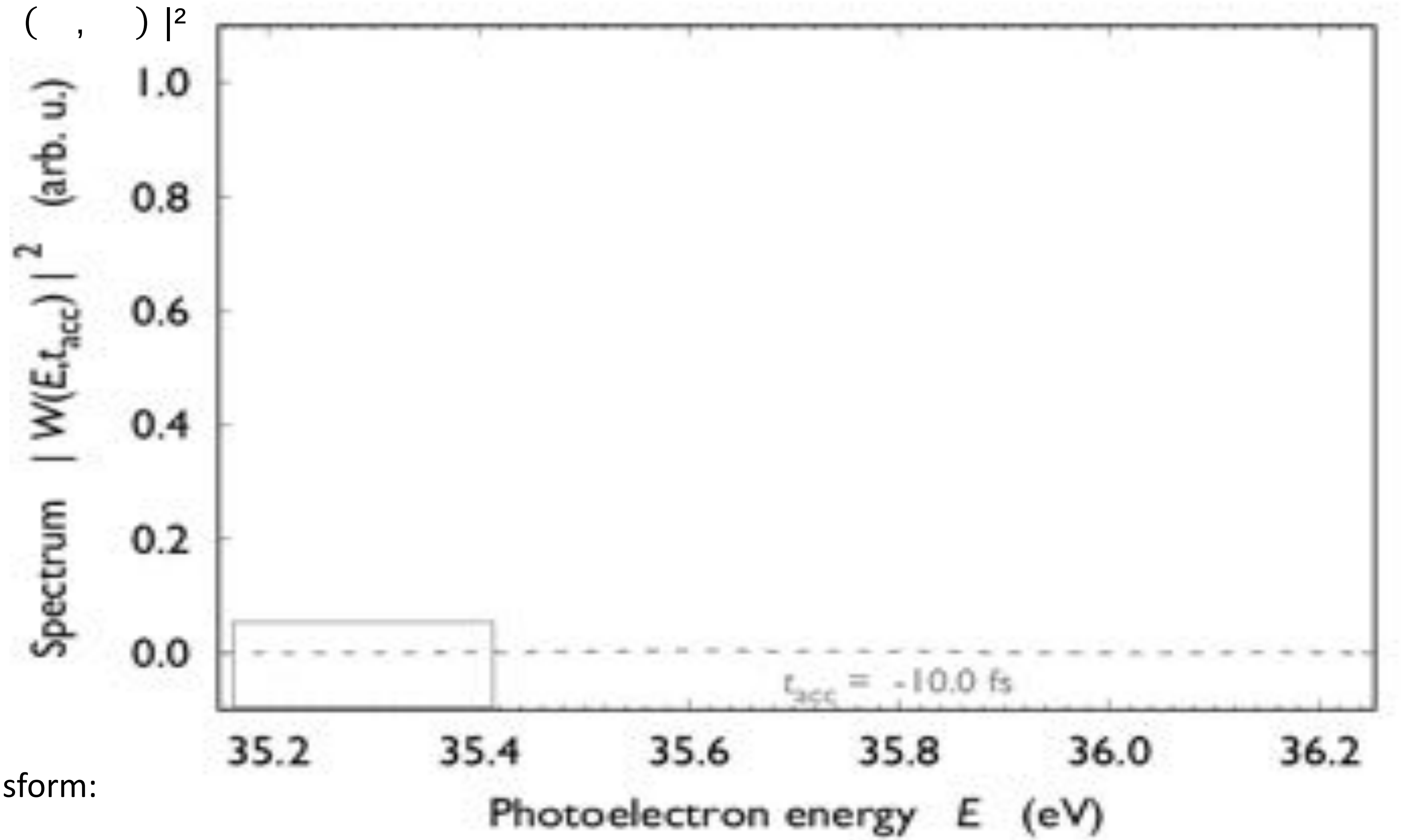
Oscillations at 2 0



=> Allows the reconstruction of the resonant electron wavepacket in the time domain: $(\ ) = \frac{1}{2} \int_{-\infty}^{+\infty} |\ (\ )|\ (\ )$

Reconstruction of the buildup of the photoelectron WP

| (,) |²



Time-limited Fourier transform:

(,) = ( ) + T

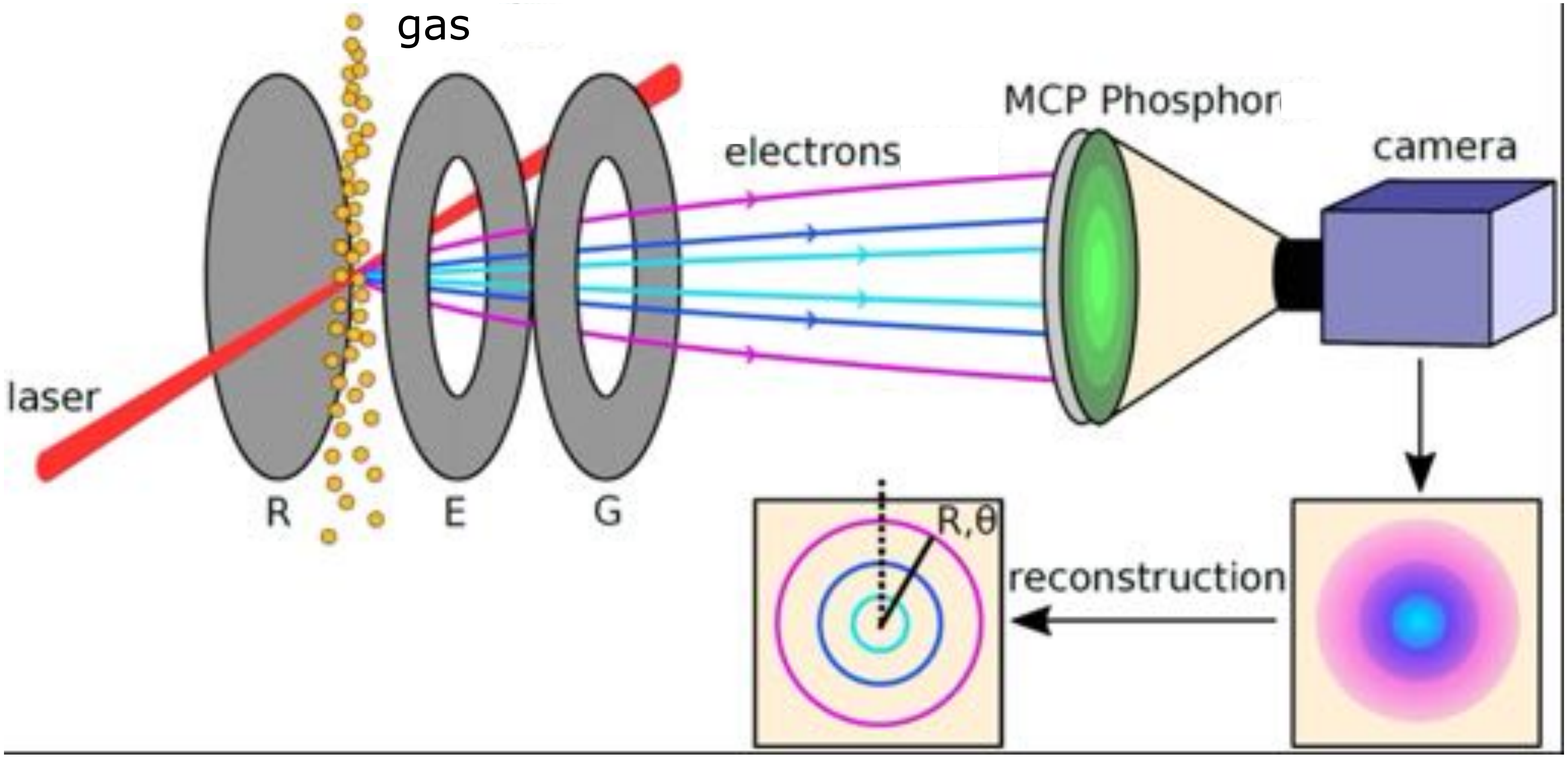
−∞

V. Gruson, L. Barreau et al., Science 354, 734 (2016)

Angularly-resolved photoemission dynamics

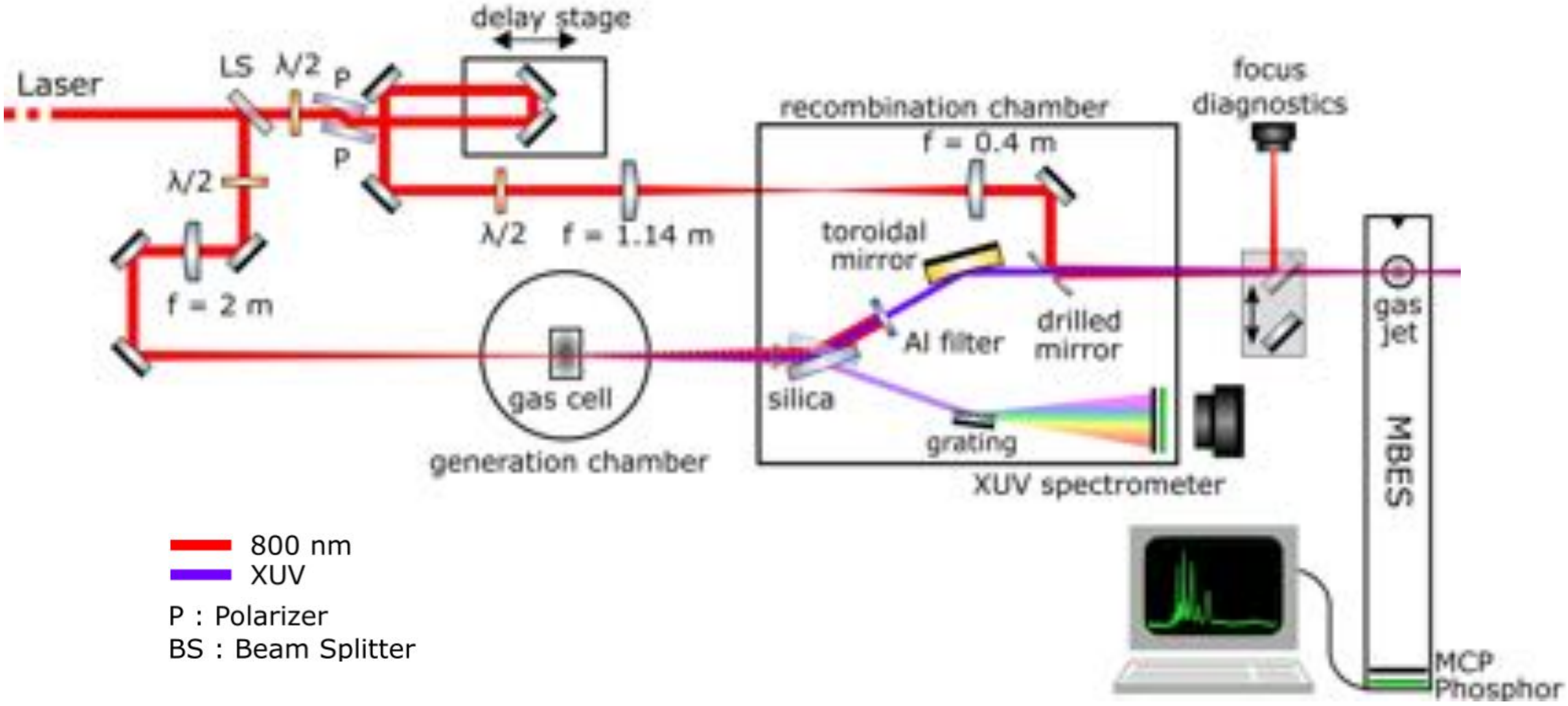
=> Combining phase spectroscopy with momentum imaging

Velocity Map Imaging Spectrometer (VMIS)



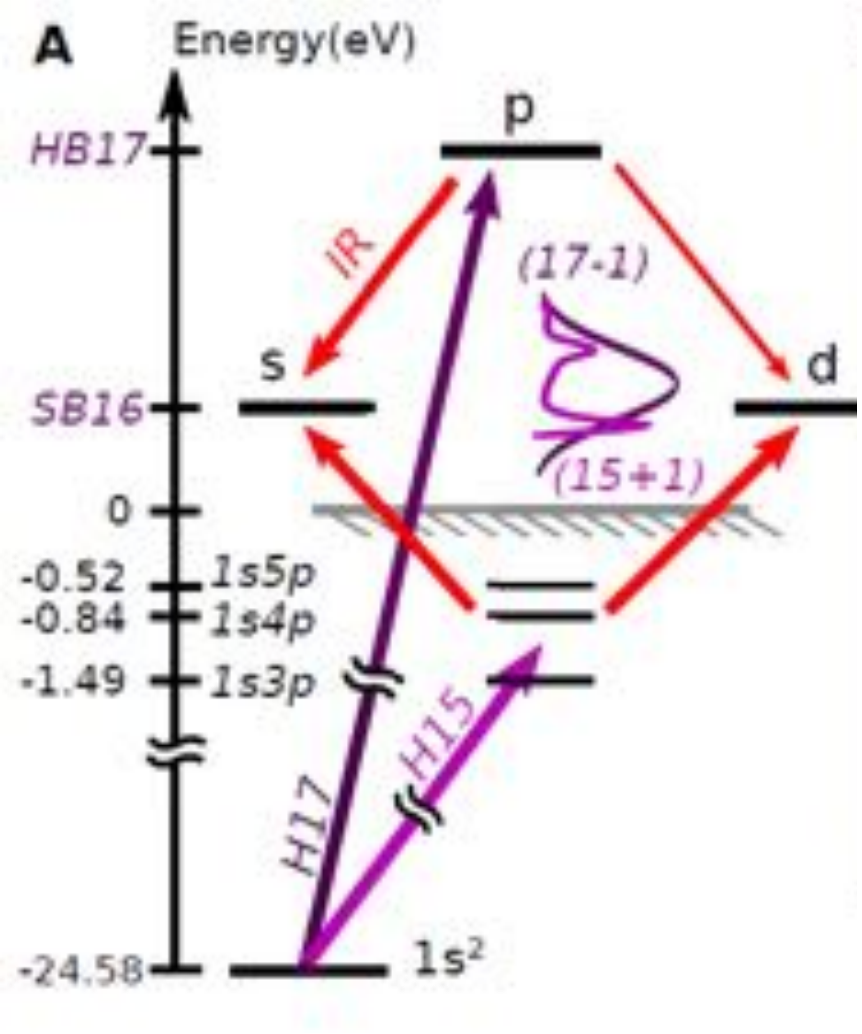
Experimental setup

Angularly-resolved RABBIT spectrogram



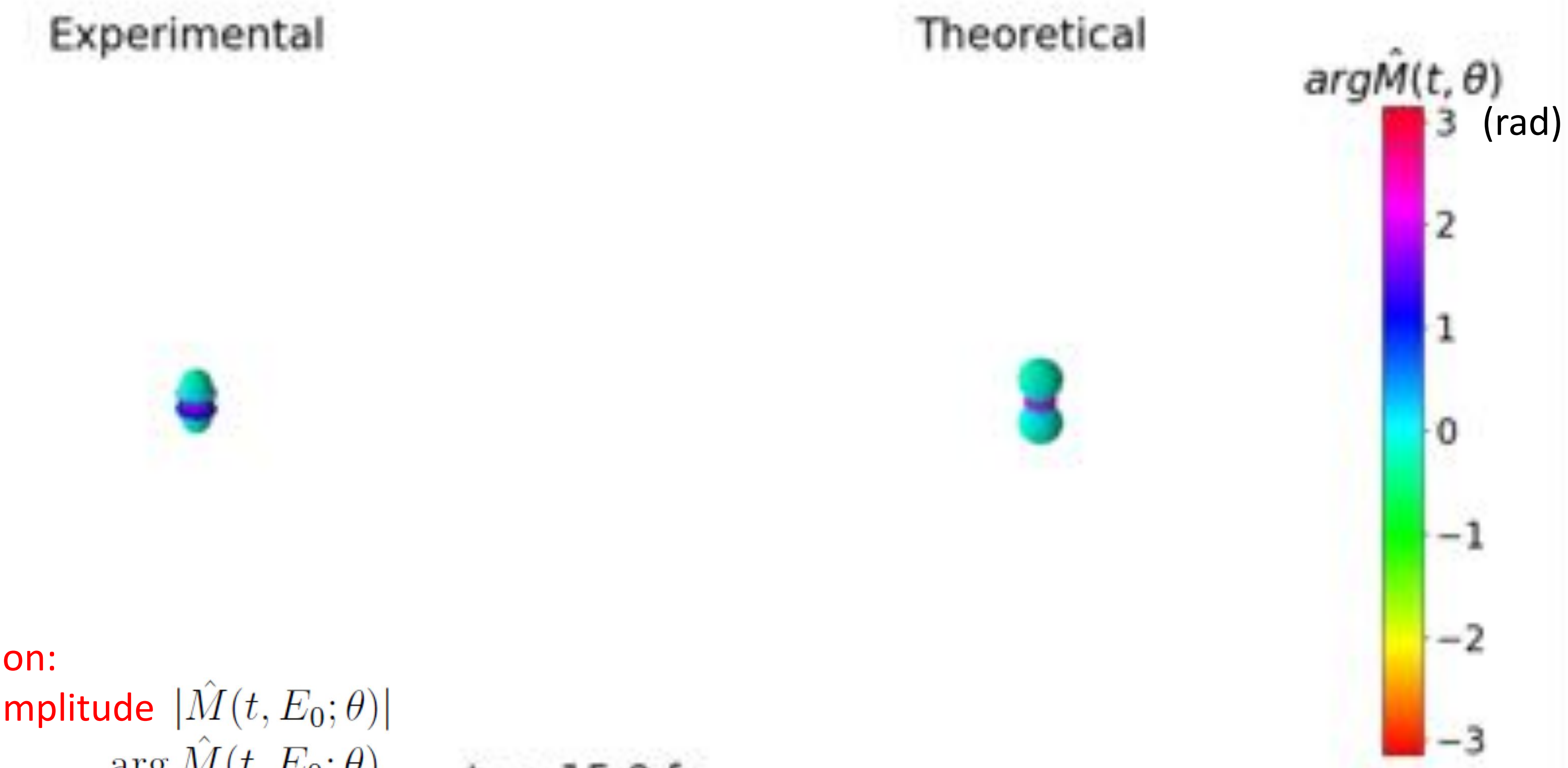
Angularly-integrated RABBIT spectrogram

Two-photon resonant ionization of Helium



Momentum map of Sideband 16

3D movie of resonant two-photon ionization



Polar representation:

(r, θ) : ionization amplitude $|\hat{M}(t, E_0; \theta)|$

(color) : phase $\arg \hat{M}(t, E_0; \theta)$

t = -15.0 fs

Conclusions and outlook

- Attosecond science is now 20 years old and was mostly based on gas high harmonic sources

  2 emerging sources: plasma harmonic sources and XFELs

  => Opens a large variety of applications in all states of matter

- ATTOLab-Orme: an attosecond facility combining advanced sources/beamlines with state-of-the-art endstations

- Attosecond spectroscopy is able to resolve in space and time photoemission dynamics
  - ü Time-resolved studies of correlated electron dynamics (electron rearrangements , ..
  - ü Study of decoherence effects in photoionization

A. Autuori, D. Platzer, M. Lejmann, G. Gallician, L. Maëder, A. Covolo, L. Bosse, M. Dalui, D. Bresteau, L. Barreau, C. Alexandridi, M. Turconi, V. Gruson, H.J.B. Marroux, J.-F. Hergott, O. Tcherbakoff, F. Lepetit, P. Salières

B. Wurmser, A. Desrier, F. Risoud, A. Maquet, J. Caillat, R. Taïeb

UNIVERSIDAD AUTONOMA DE MADRID

Á. Jiménez Galán, L. Petersson, M.Klinker, C. Marante, J. Gonzalez-Vazquez, F. Martín

INSTITUT LUMIÈRE MATIÈRE

V. Loriot, F. Lépine

L. Poisson

UNIVERSITY OF CENTRAL FLORIDA 1963

L. Argenti