

Attosecond spectroscopy on solids

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X-Ray Science in the 21st Century
KITP
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Overview

- Needs for generating **isolated** attosecond **x-ray** pulses via HHG:
Phase-stable few-cycle driver pulses at **long wavelengths**
- attosecond measurement technique:
streaking
- What to do with attosecond pulses?
- Pulse duration measurement at LCLS

The Laser System

**Hollow Fiber/
Chirped Mirror
Pulse Compressor**

<3.5 fs pulse duration
400 μ J energy

Experiment

Booster: 4 kHz, 4 mJ, 28 fs

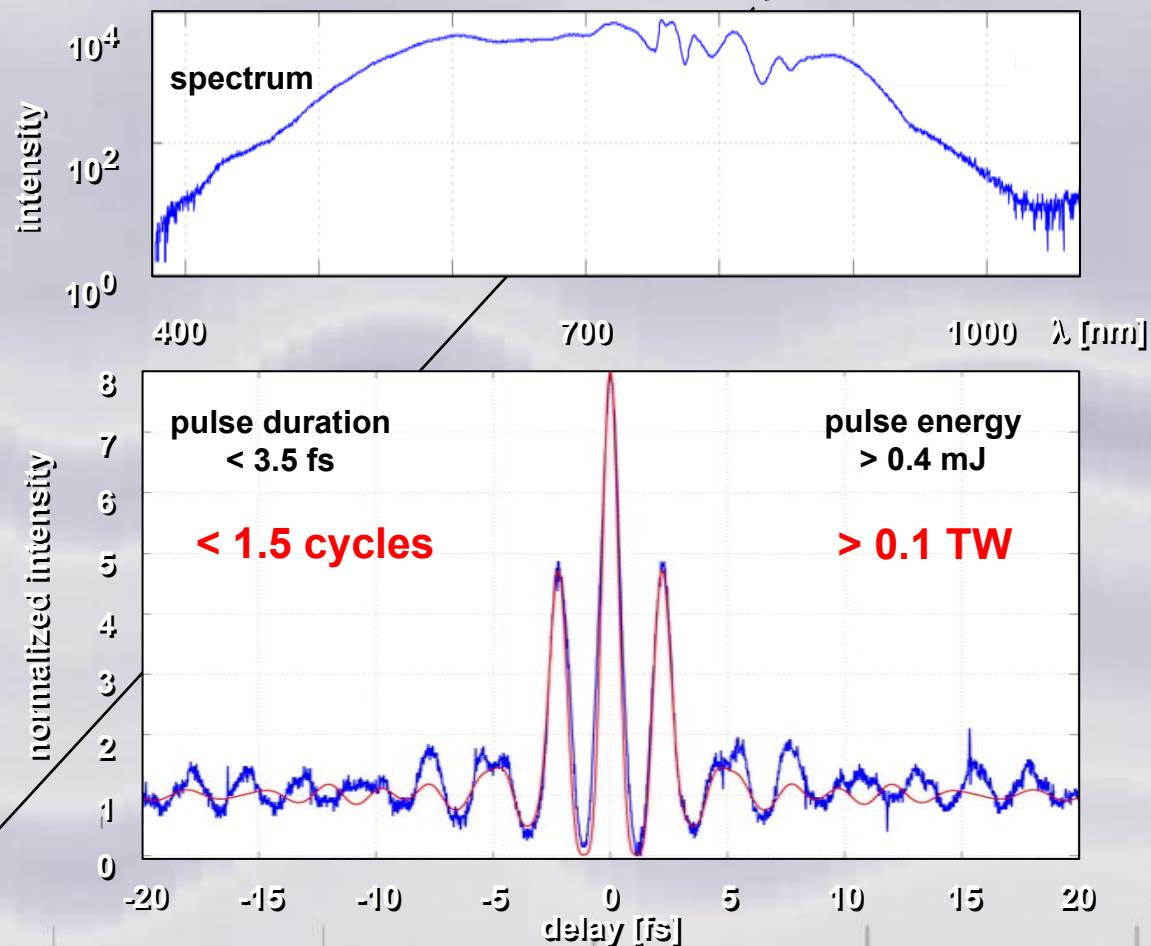
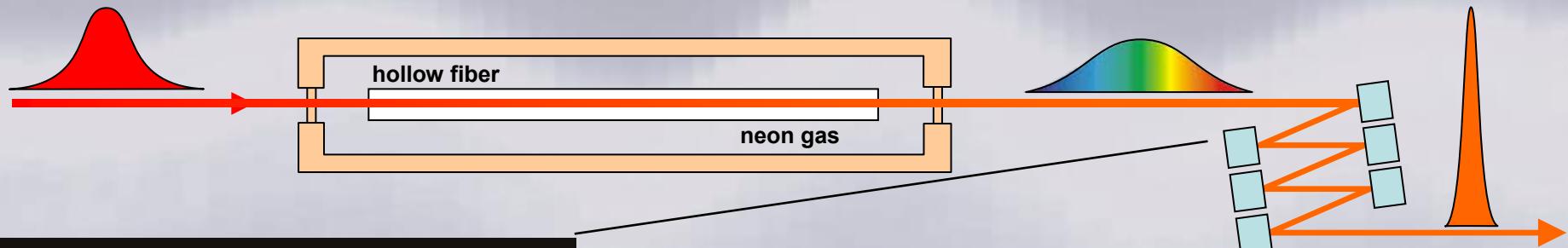
CPA Amplifier

20 fs pulse duration
1.3 mJ energy
3 kHz repetition rate

Ti:Sapphire Oscillator

sub-10 fs pulse duration
4.6 nJ energy
82 MHz repetition rate

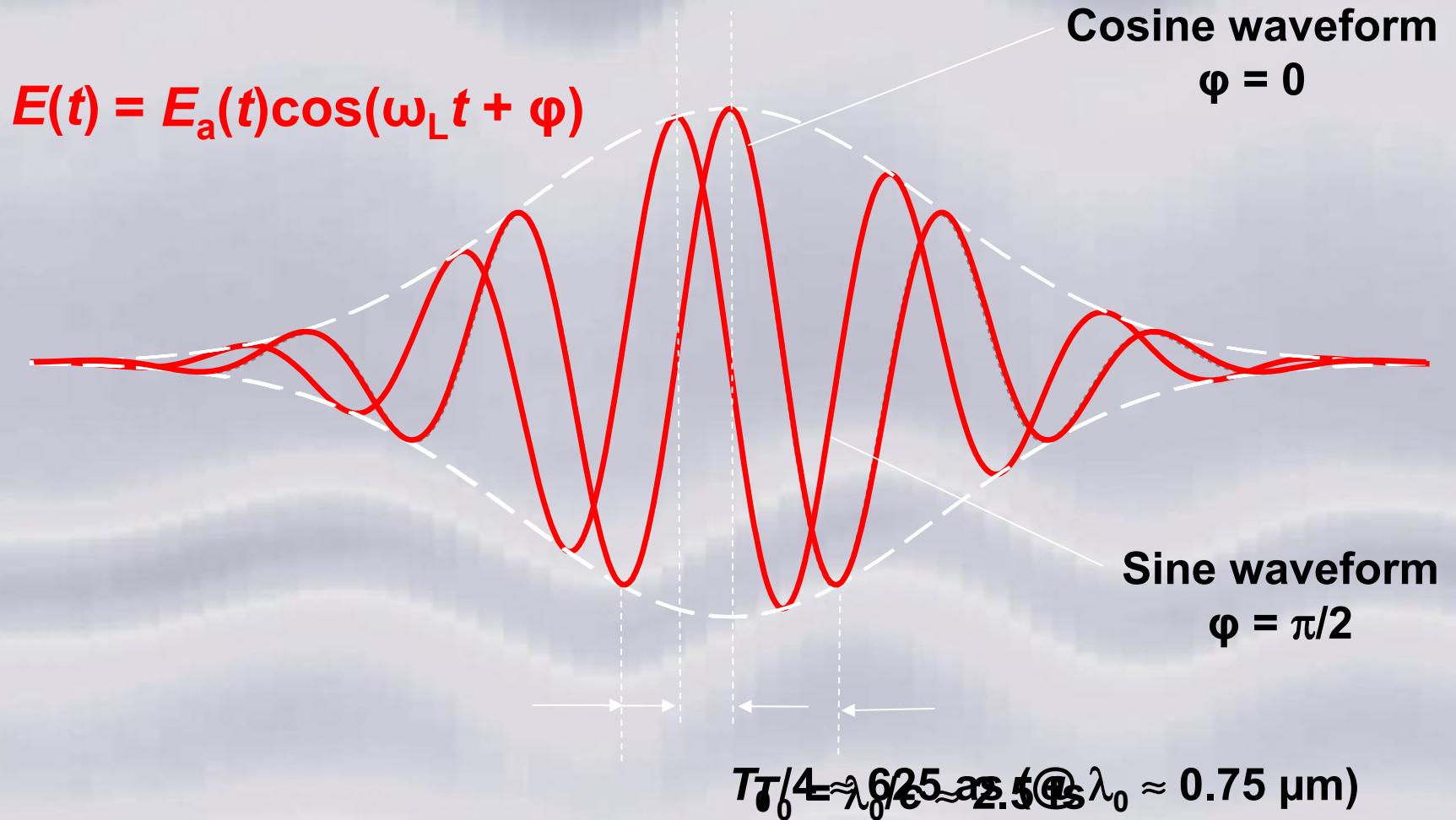
key tools of attosecond technology: **synthesized few-cycle wave & synchronized sub-fs xuv pulse**



A. L. Cavalieri et al, *New J. Phys.* 9, 242 (2007);

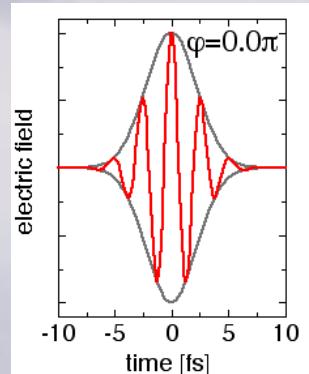
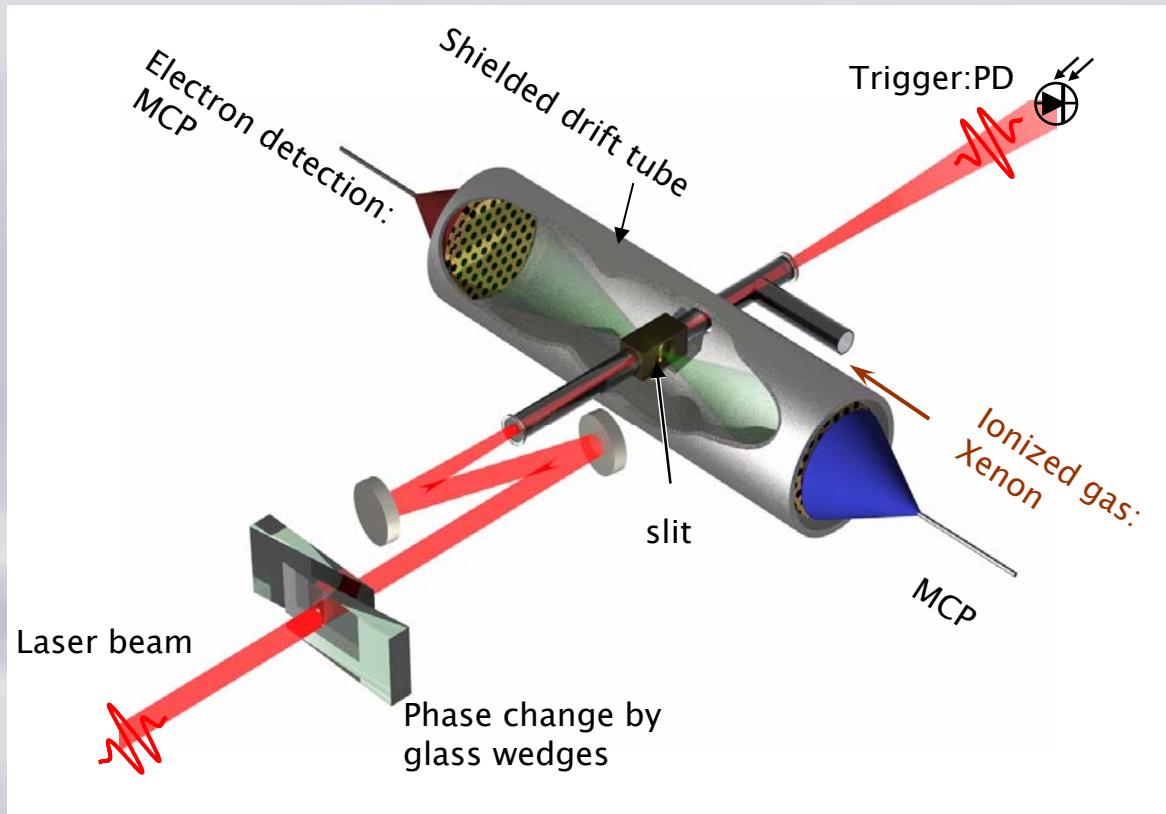
E. Goulielmakis et al, *Science* 317, 769 (2007)

Control of the waveforms

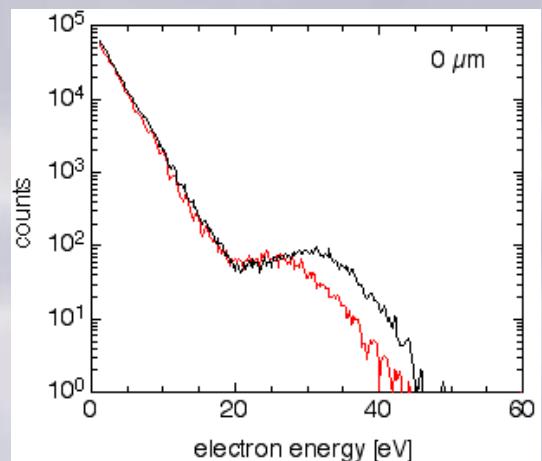


Requires measurement & control of φ

Detection of the absolute phase



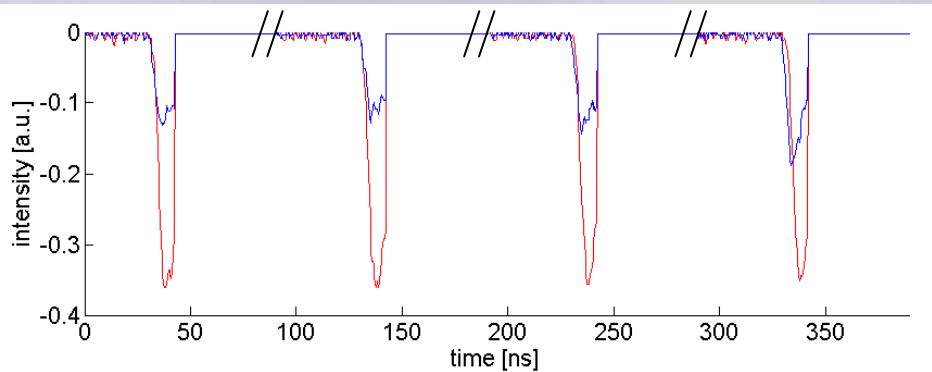
Left-right ATI spectra



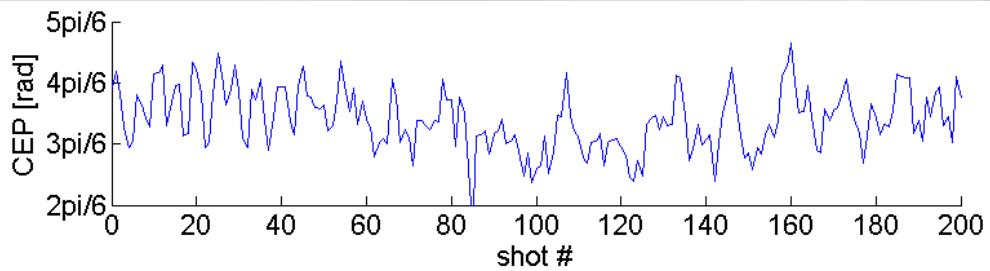
Previous results:
average of
10000 shots

CEP of consecutive phase-stabilized pulses

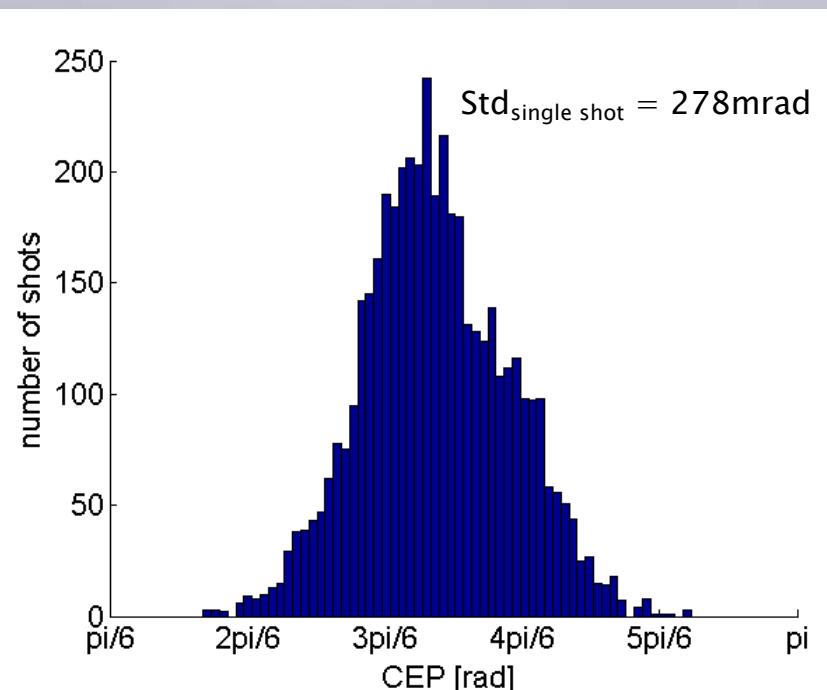
consecutive time-of-flight ATI spectra of rescattered electrons (laser rep. rate = 3kHz)



shot-to-shot evolution of the CEP

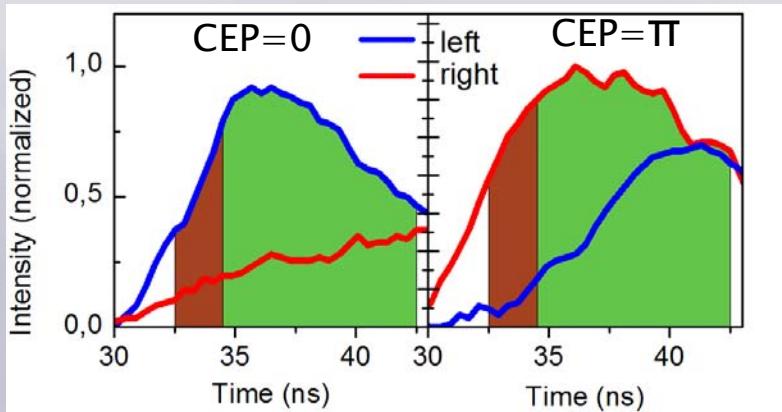


phase distribution of CEP-stabilized shots
(4500 shots; 3kHz)

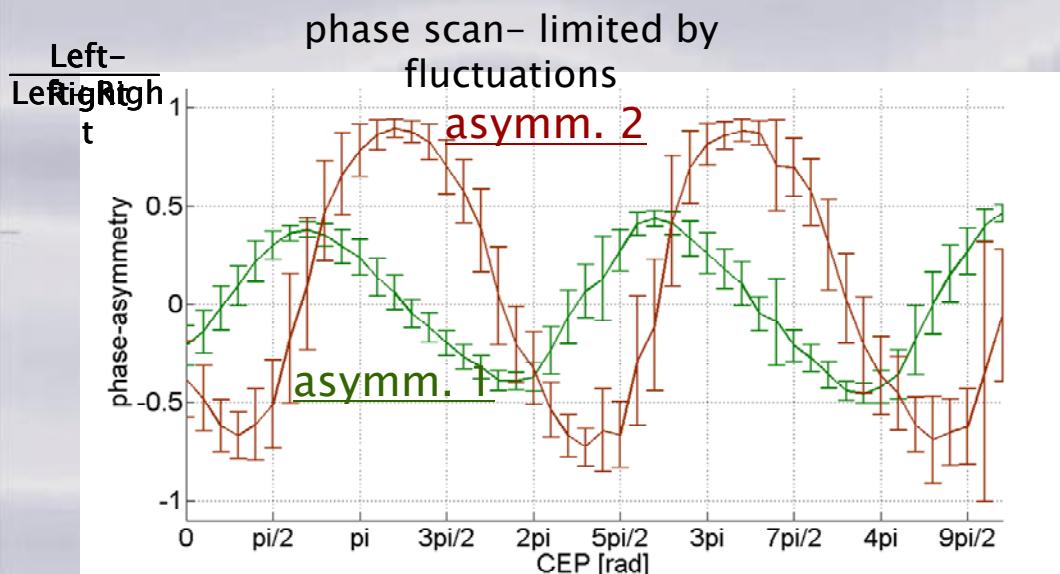
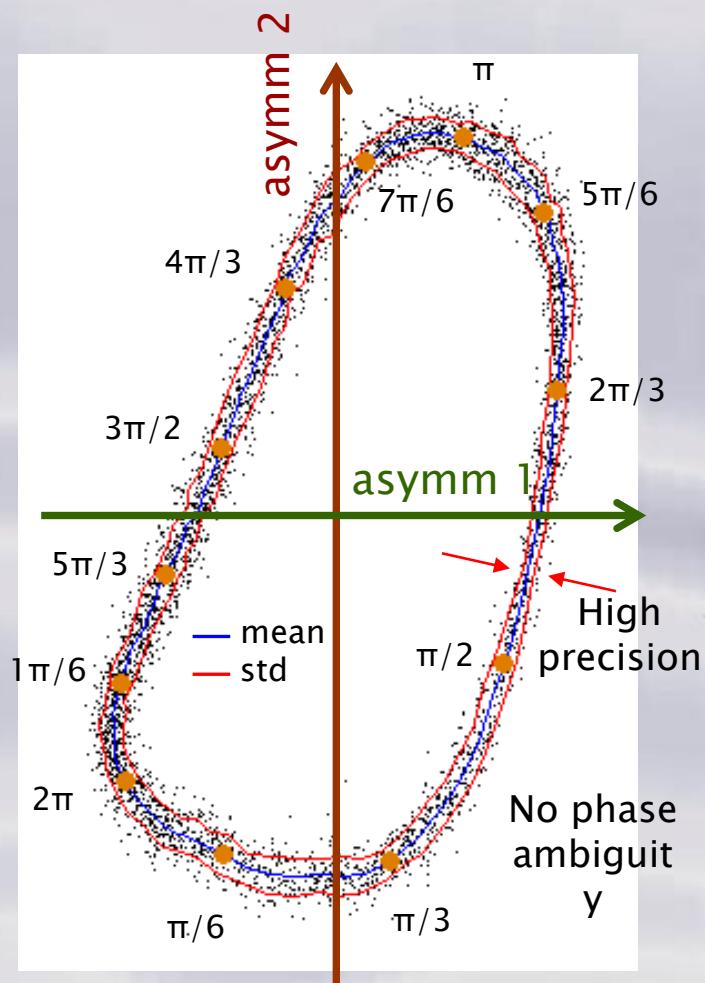


Parametric (Lissajous-like) representation

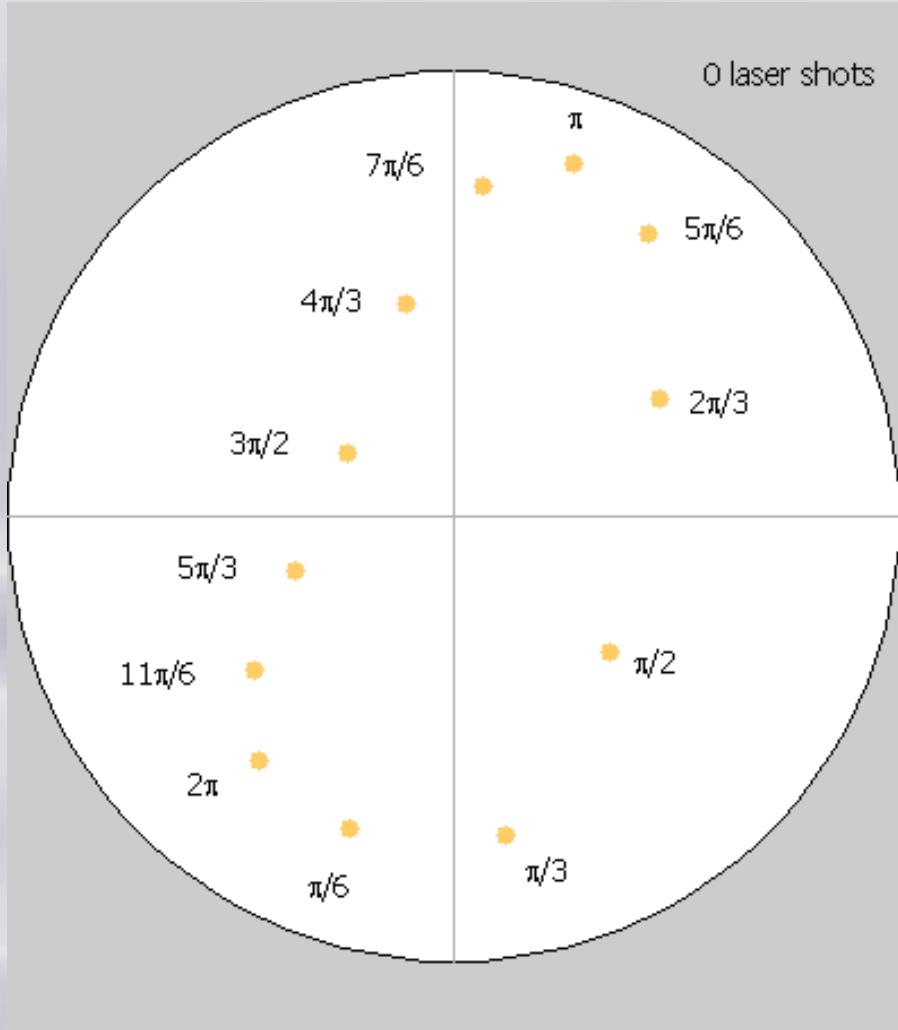
single shot left-right TOF spectra



Non-phase-stabilized single shots- random distribution



Non-phase-stabilized laser shots as they're arriving...



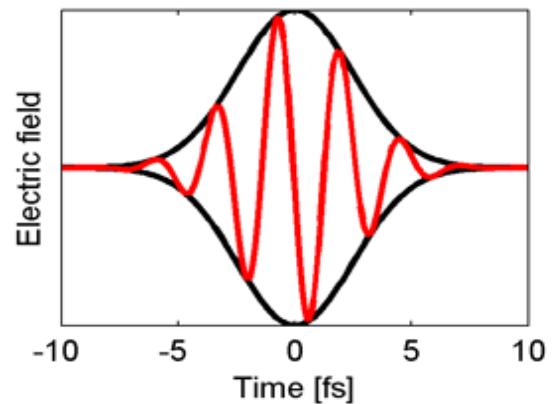
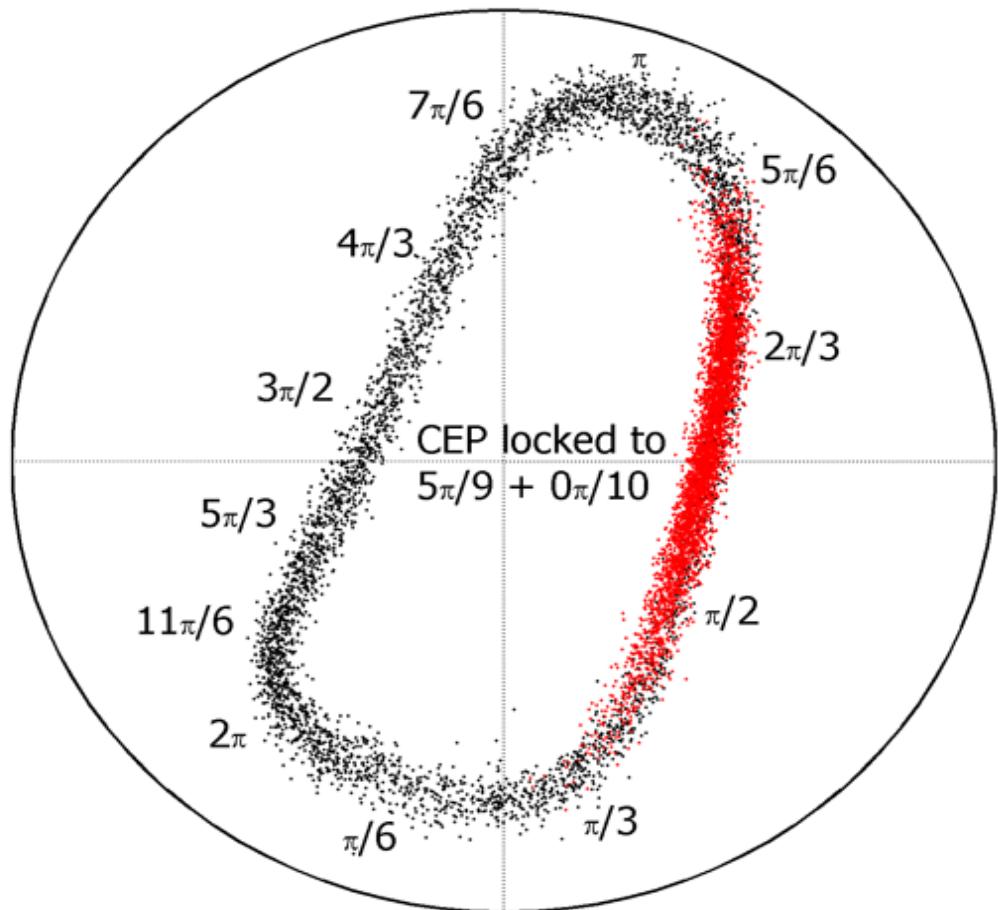
non-stabilized shots has random phase distribution



first principle calibration:
phase difference between
shots is immediately given

real CEP retrieved by
TDSE

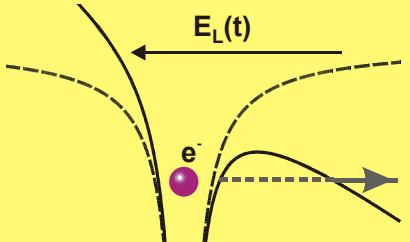
Phase-scan with a stabilized laser



High-order Harmonic Generation in the gas phase

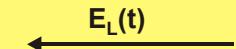
Step 1

Optical field ionization



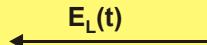
Step 2

e^- Acceleration



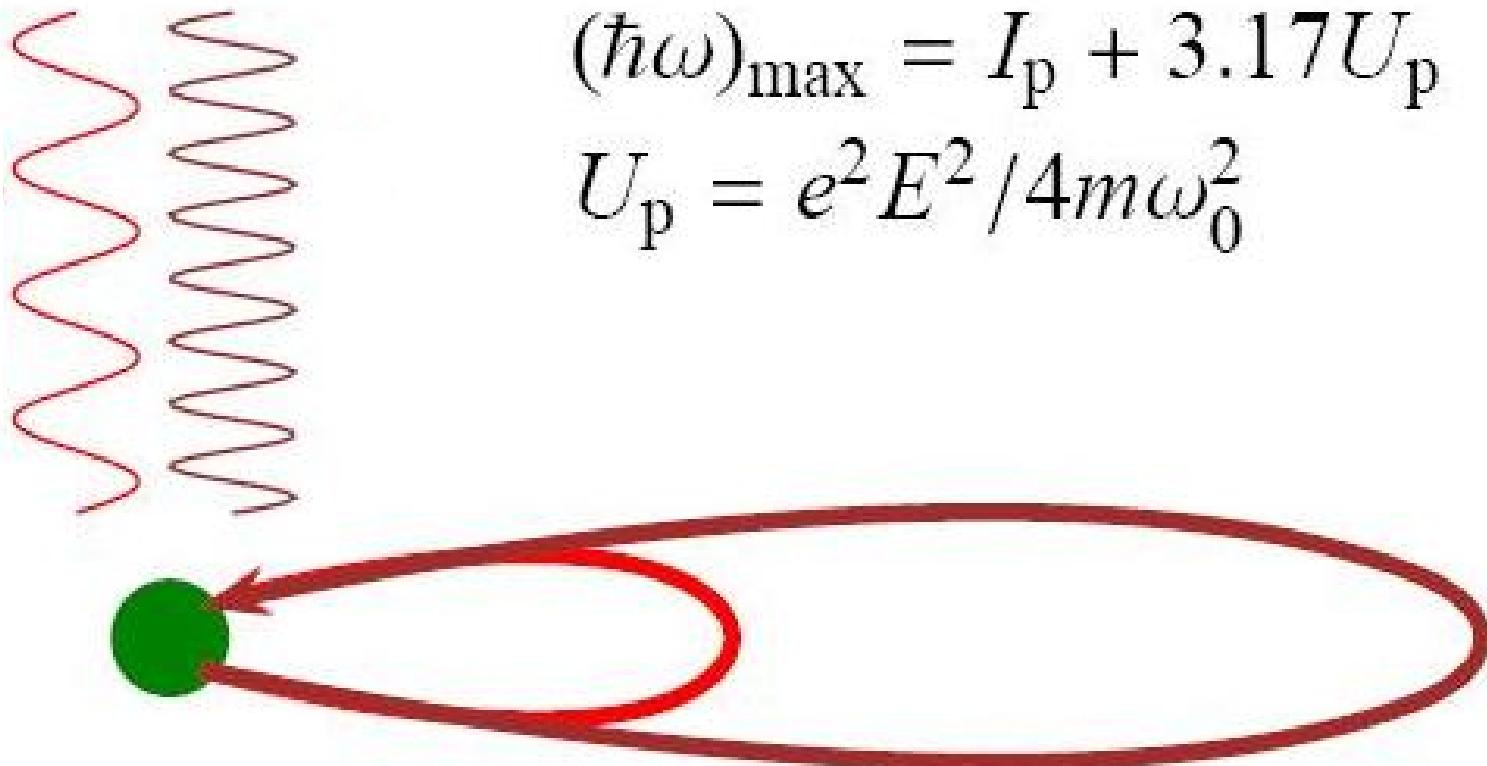
Step 3

XUV emission on recollision



P.B. Corkum, PRL 71 (1993)

Extending the cutoff



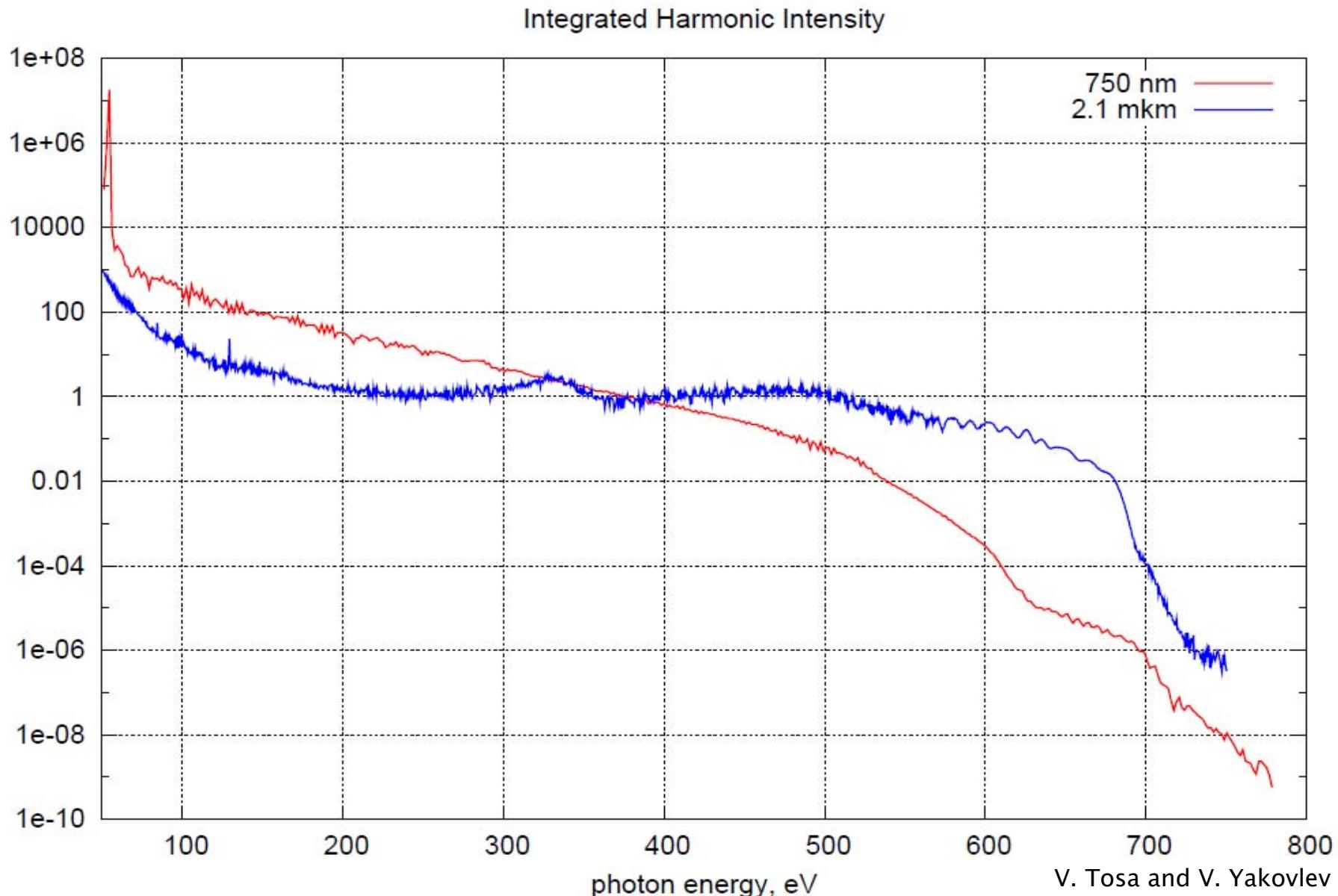
$$(\hbar\omega)_{\max} = I_p + 3.17U_p$$

$$U_p = e^2 E^2 / 4m\omega_0^2$$

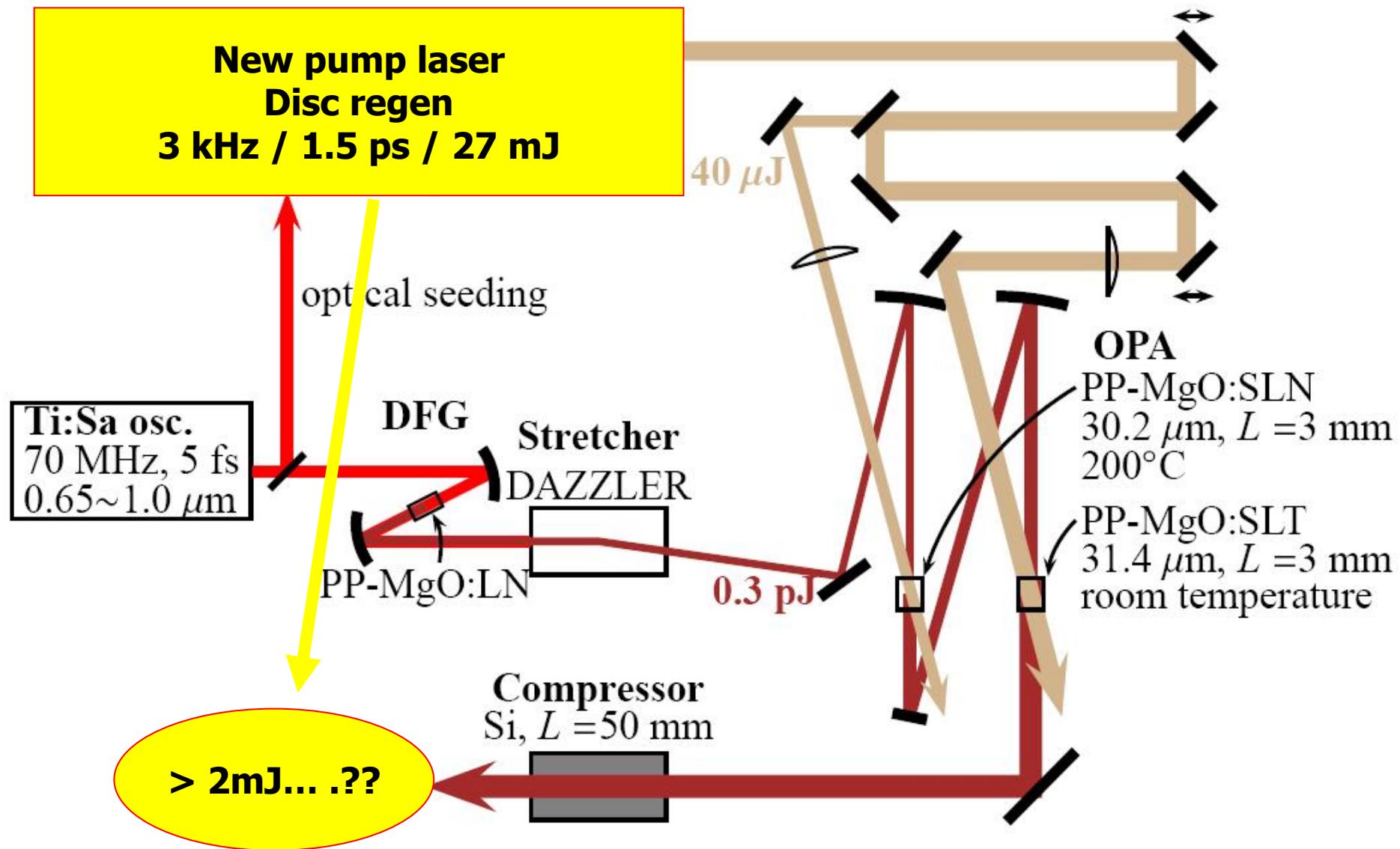
using an IR driving field

- ω : frequency of HHG
- I_p : atomic ionization potential
- U_p : kinetic energy of the electron
- E : amplitude of electric field
- ω_0 : laser frequency
- m : mass of the electron
- e : charge of the electron

Comparison VIS / IR driver



2.1 μm few-cycle OPCPA system

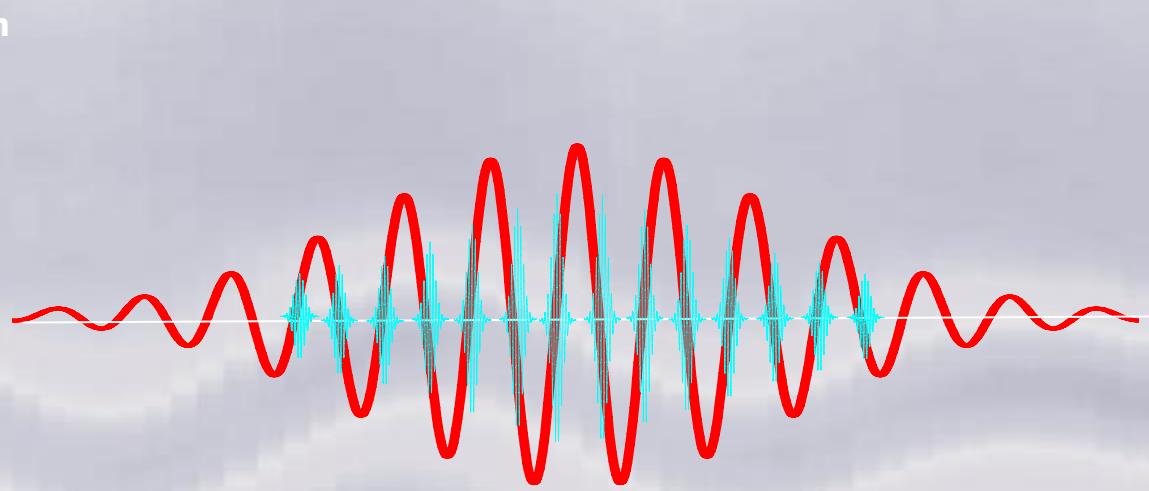
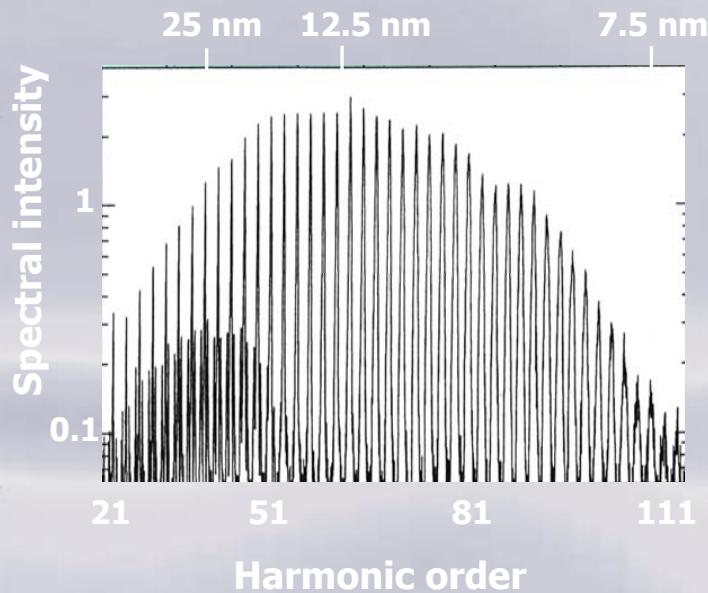


IR – driven HHG

Cut-off ~1.6 keV

Multi-cycle driver pulse : $\tau_p \gg T_o$

High-order odd harmonics of the driver laser

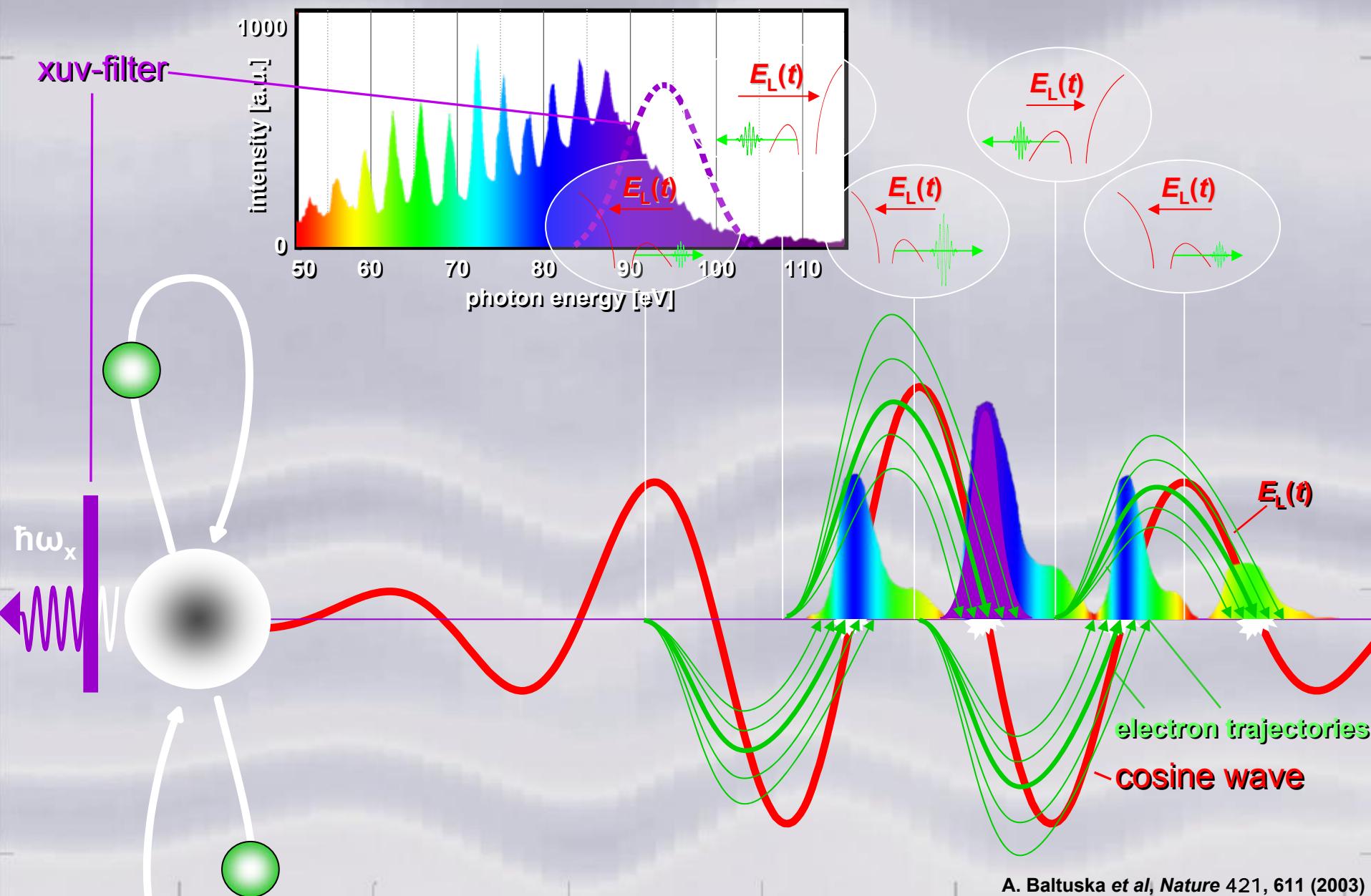


Cut-off harmonics: train of attosecond bursts

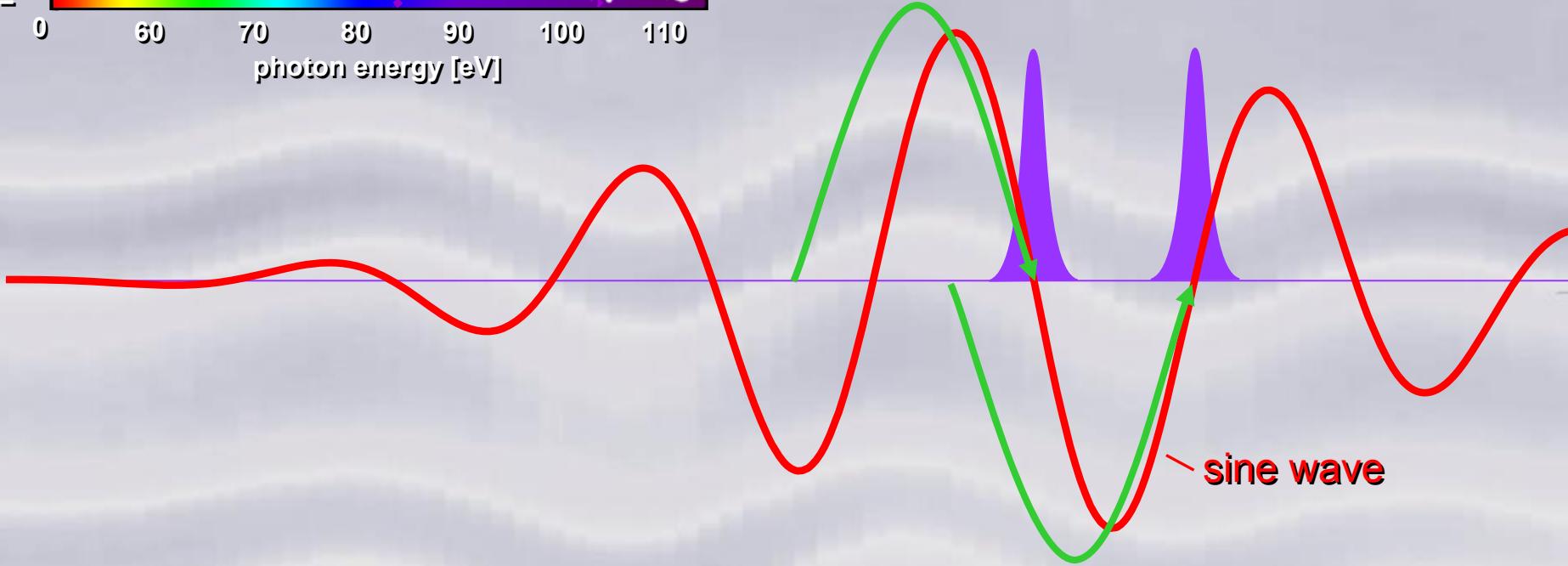
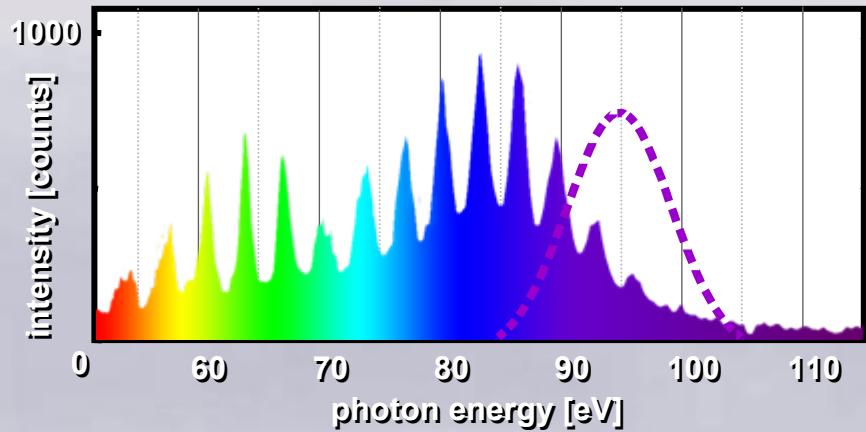
L'Huillier, Balcou, 1993, *PRL* 70, 774
Macklin *et al*, 1993, *PRL* 70, 766

Paul *et al*, *Science* 292, 1689 (2001)
Tsakiris, Charalambidis *et al*, 2003

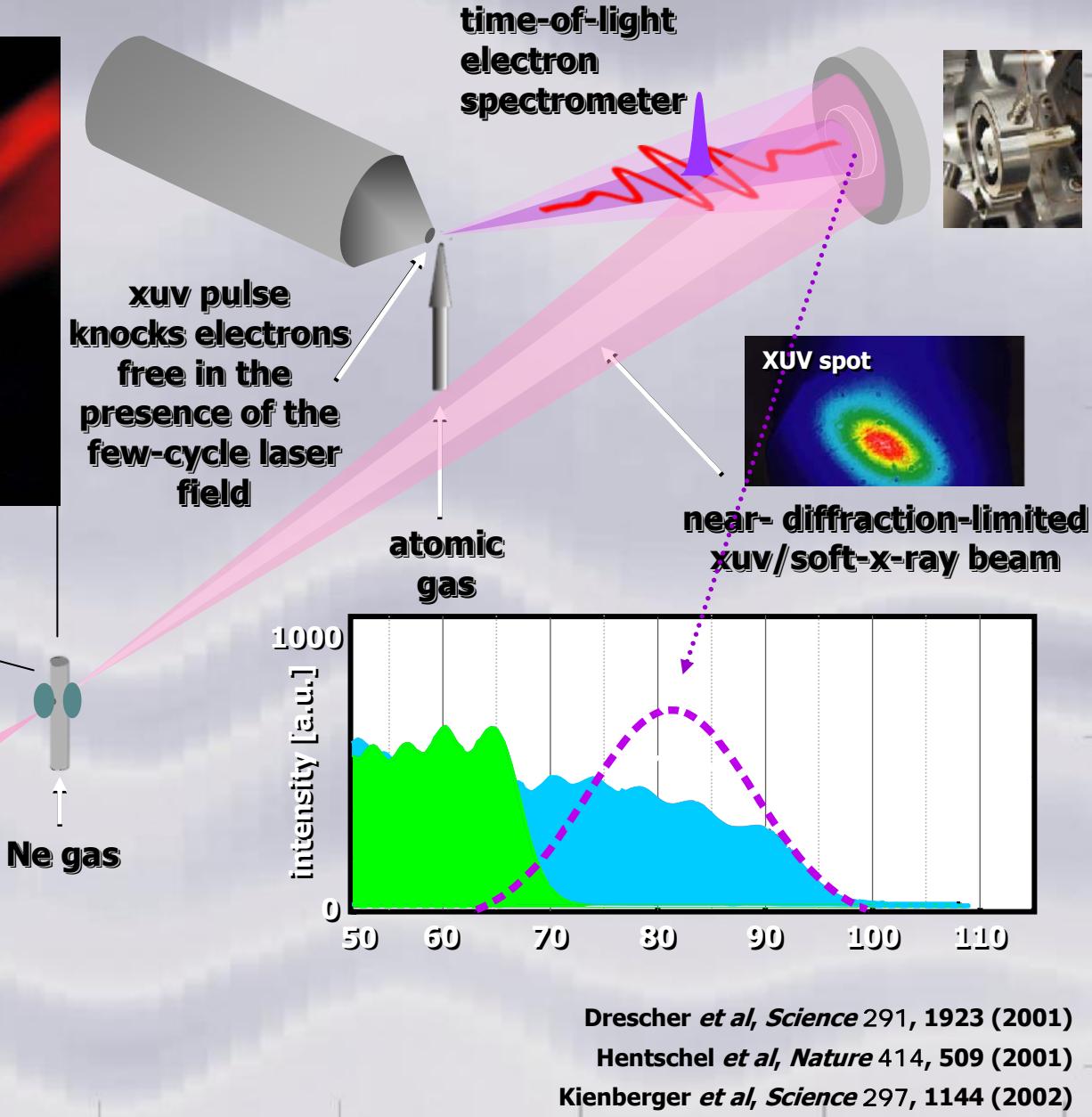
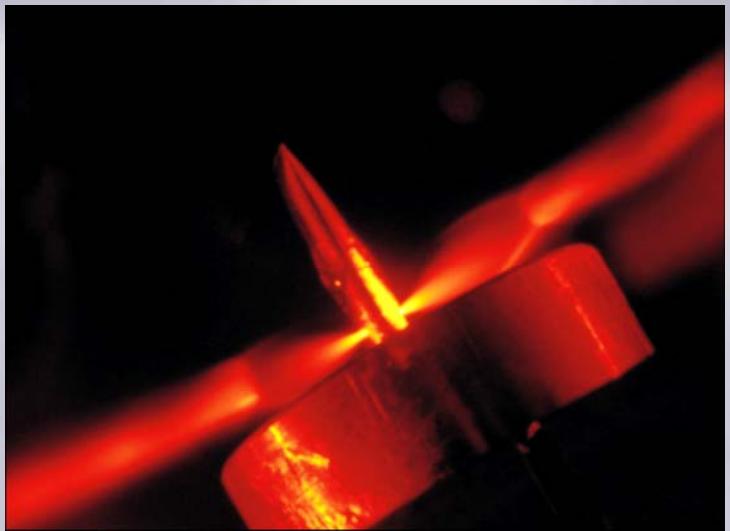
steering bound electrons with controlled light fields: the birth of an attosecond pulse



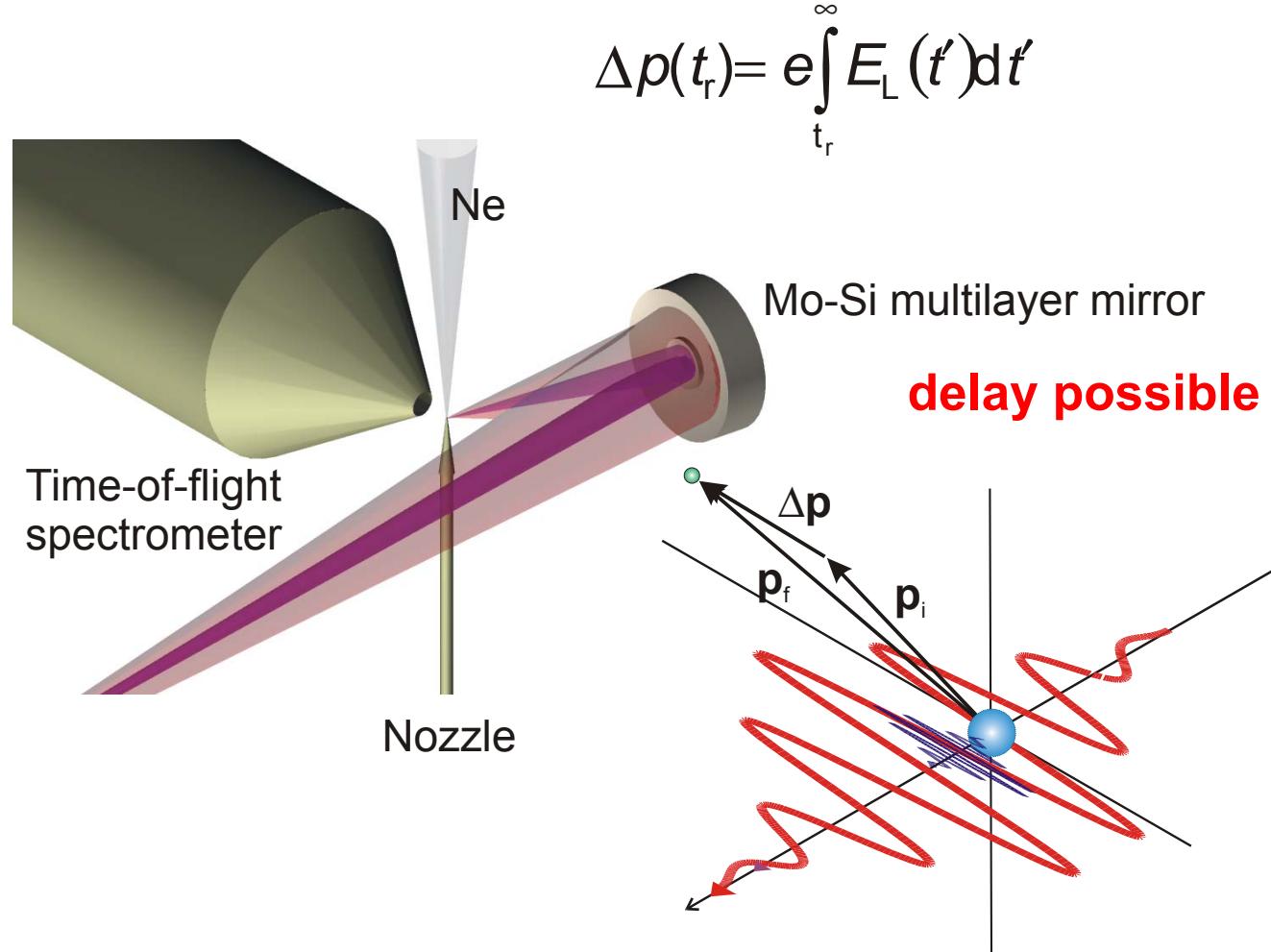
attosecond xuv/x-ray pulse generation



attosecond pulse generation and measurement



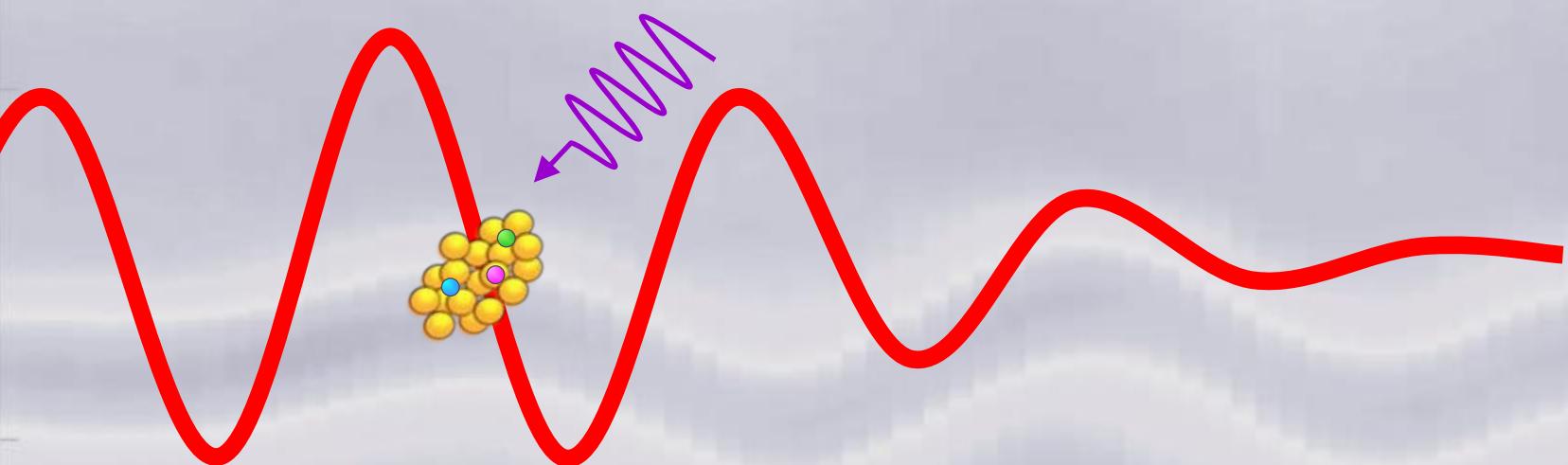
Ionization with an Isolated Attosecond Pulse



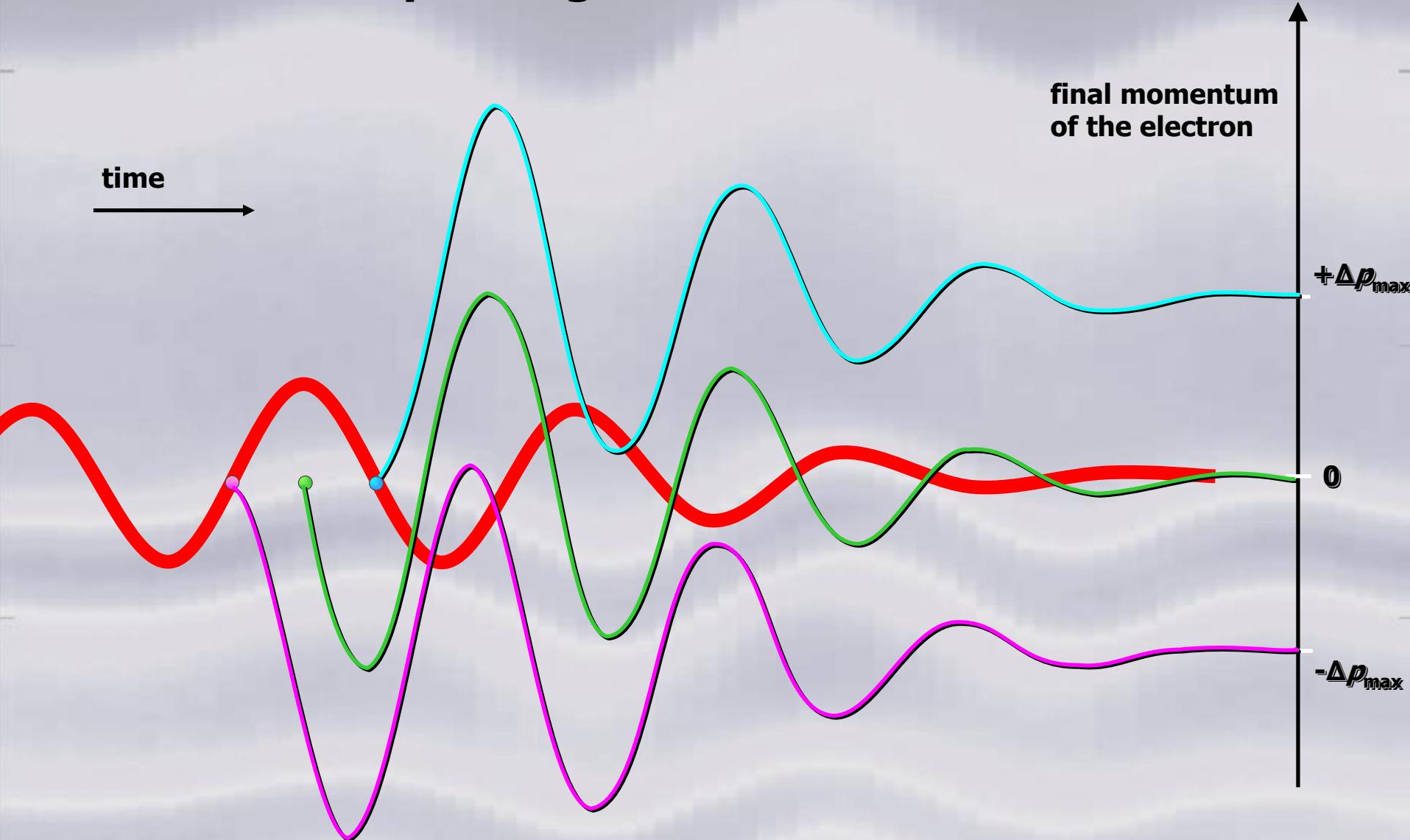
Detection as in:
Kienberger *et al.*, Science **297**, 1144 (2002)

XUV cut-off energy: ~100 eV
Mirror reflectivity bandwidth up to: 30 eV (FWHM)

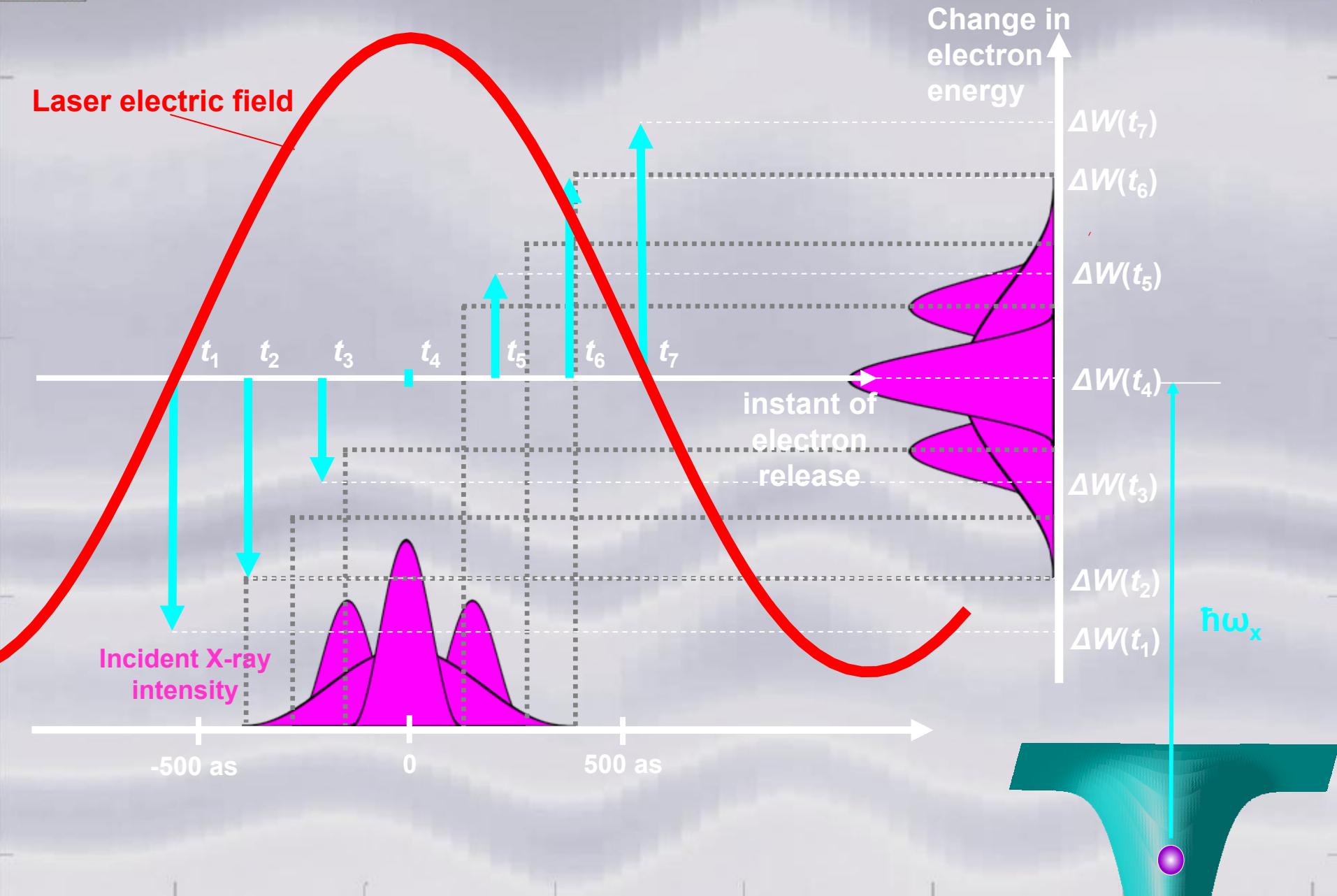
Ionization at different instants of time



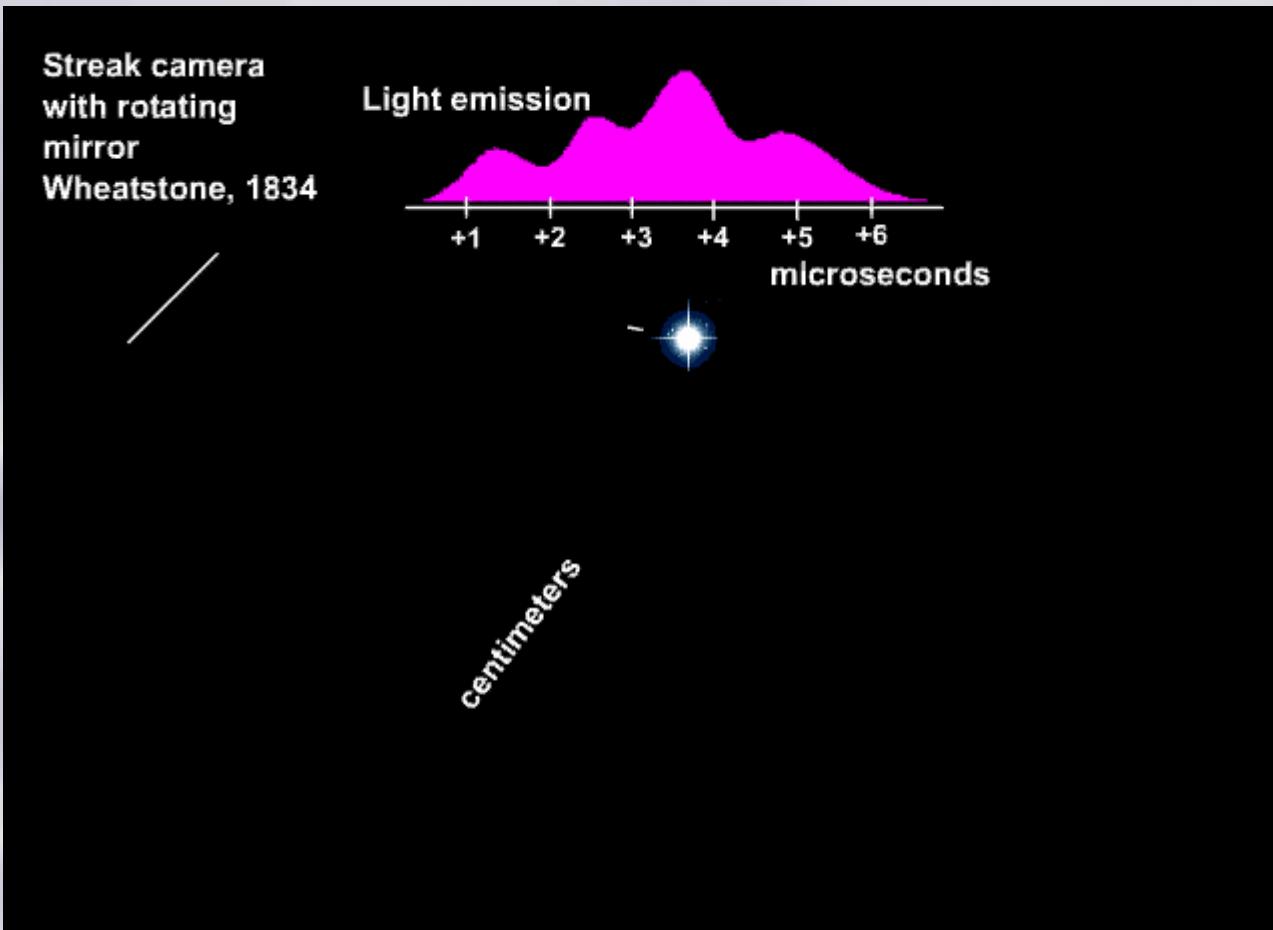
final momentum of photoelectrons depending on the release time



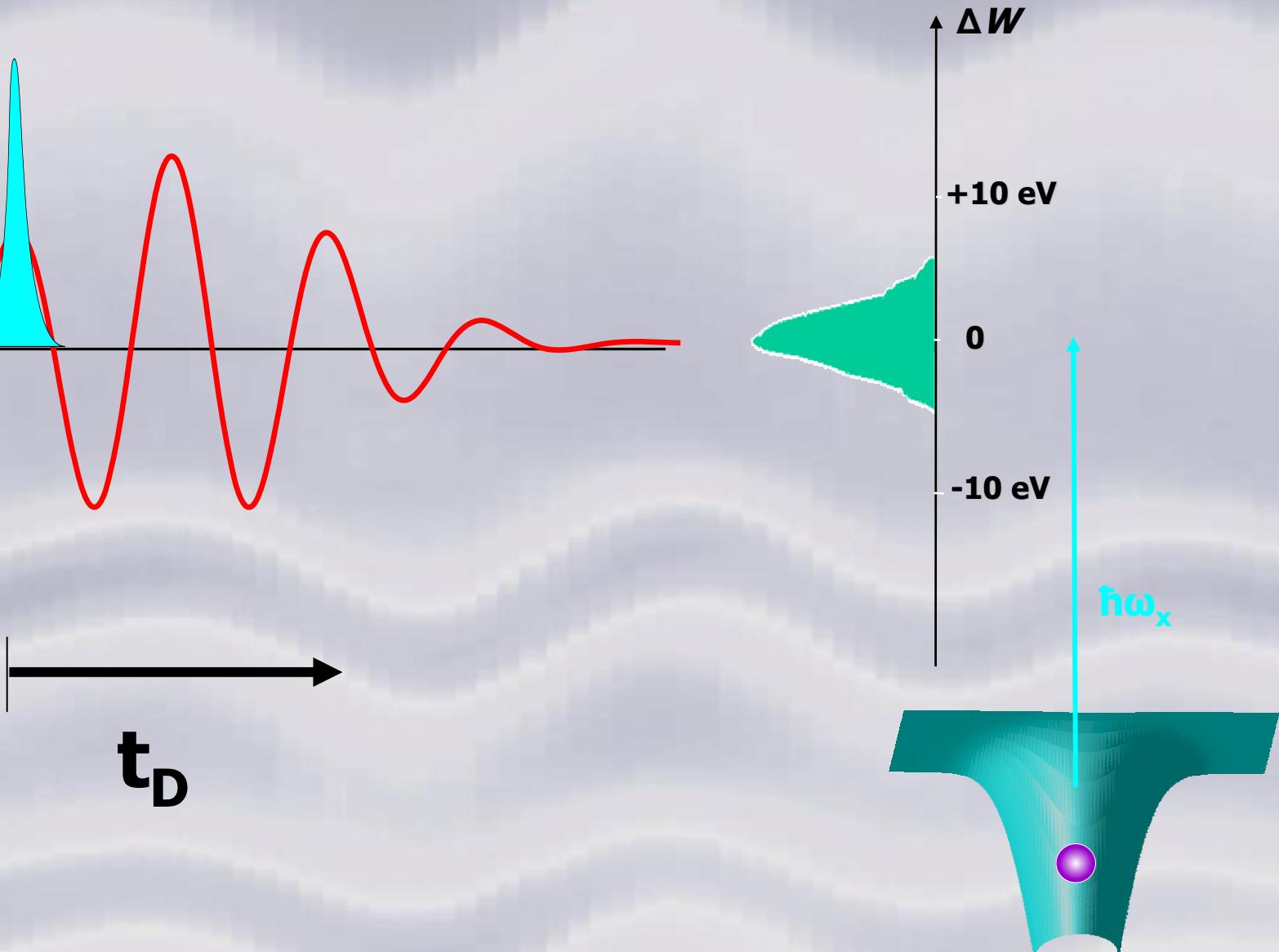
Mapping Time to Energy



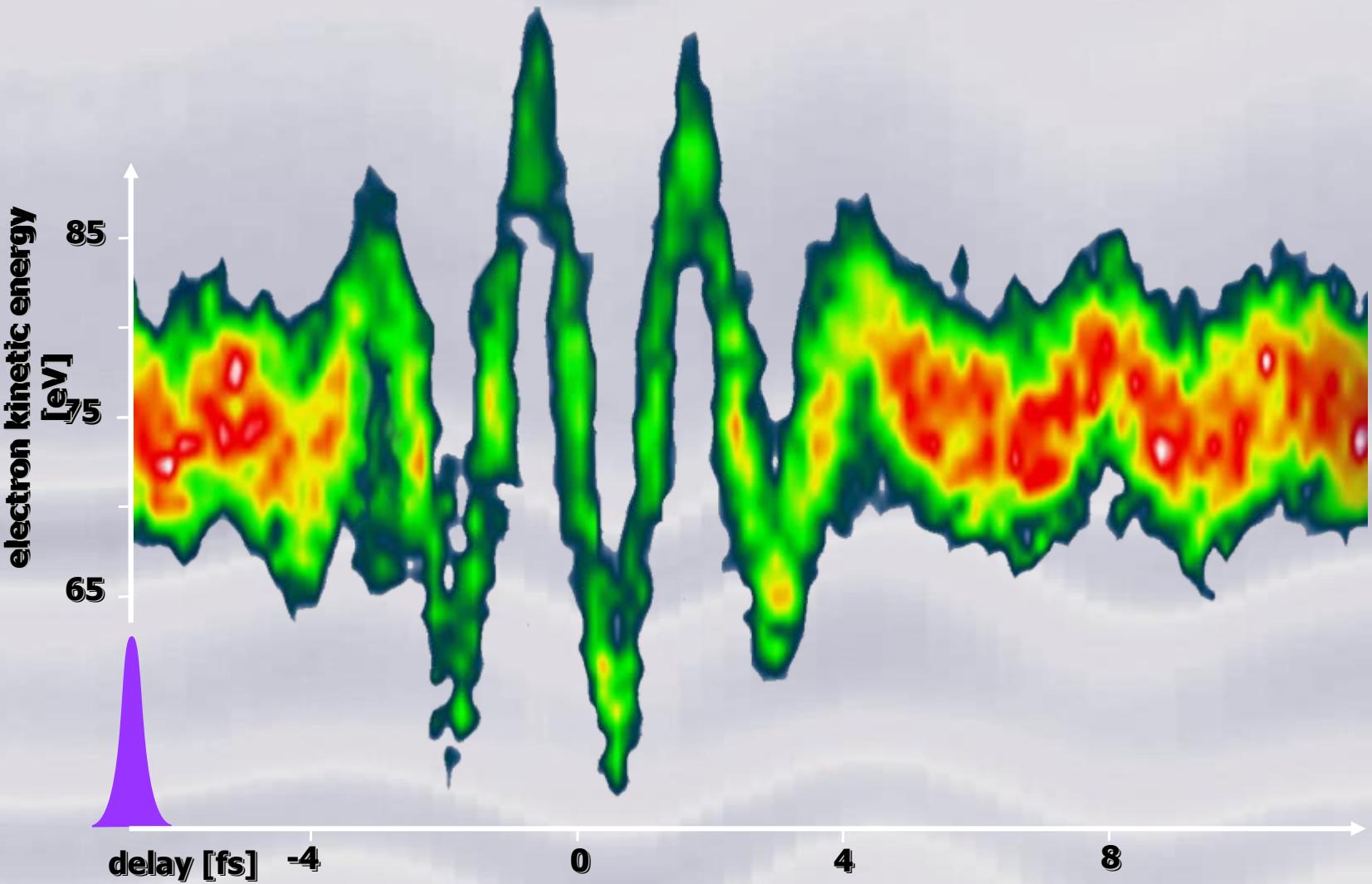
Optical Streak Camera, 1834



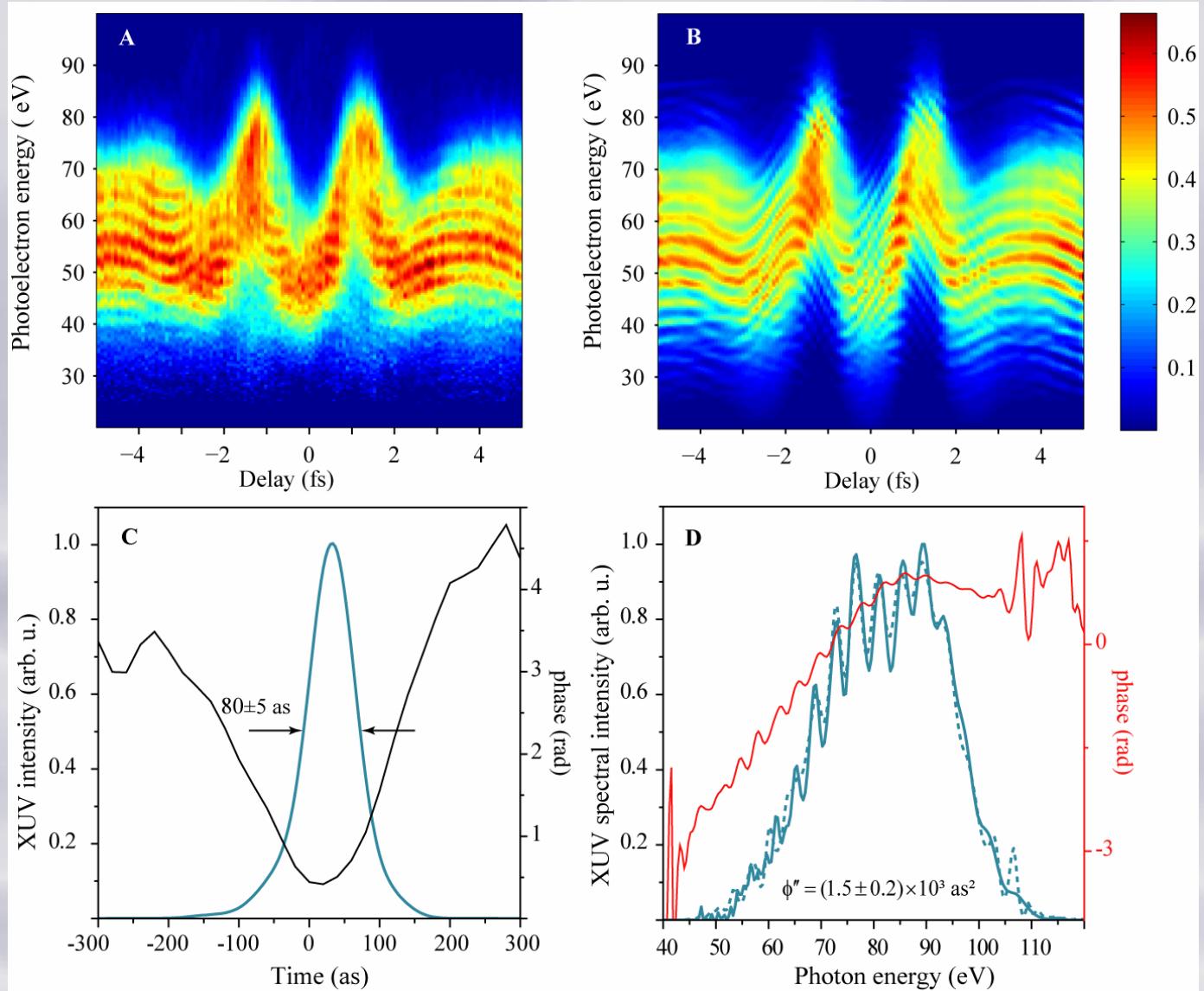
sampling field oscillations



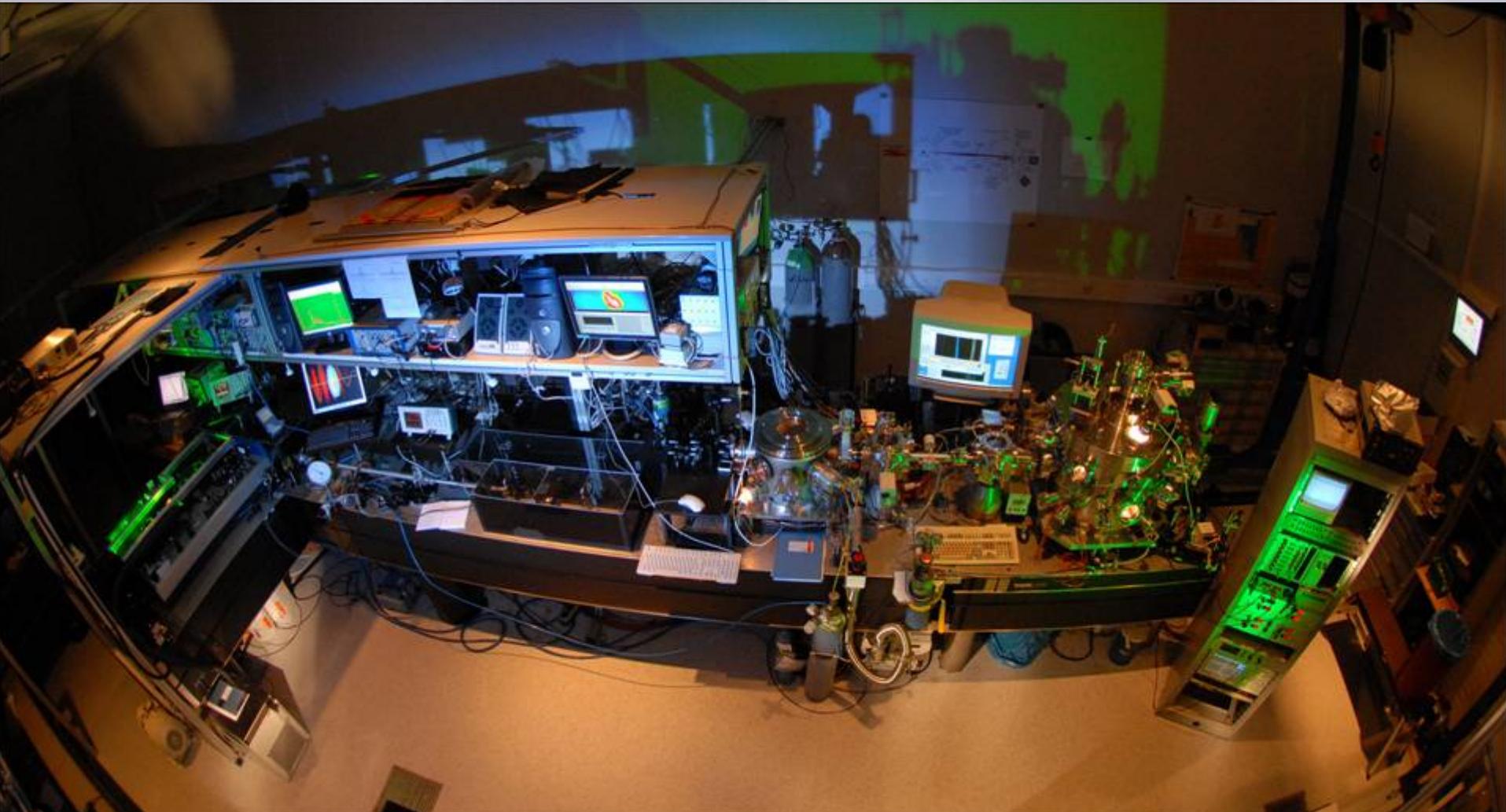
a streaking trace



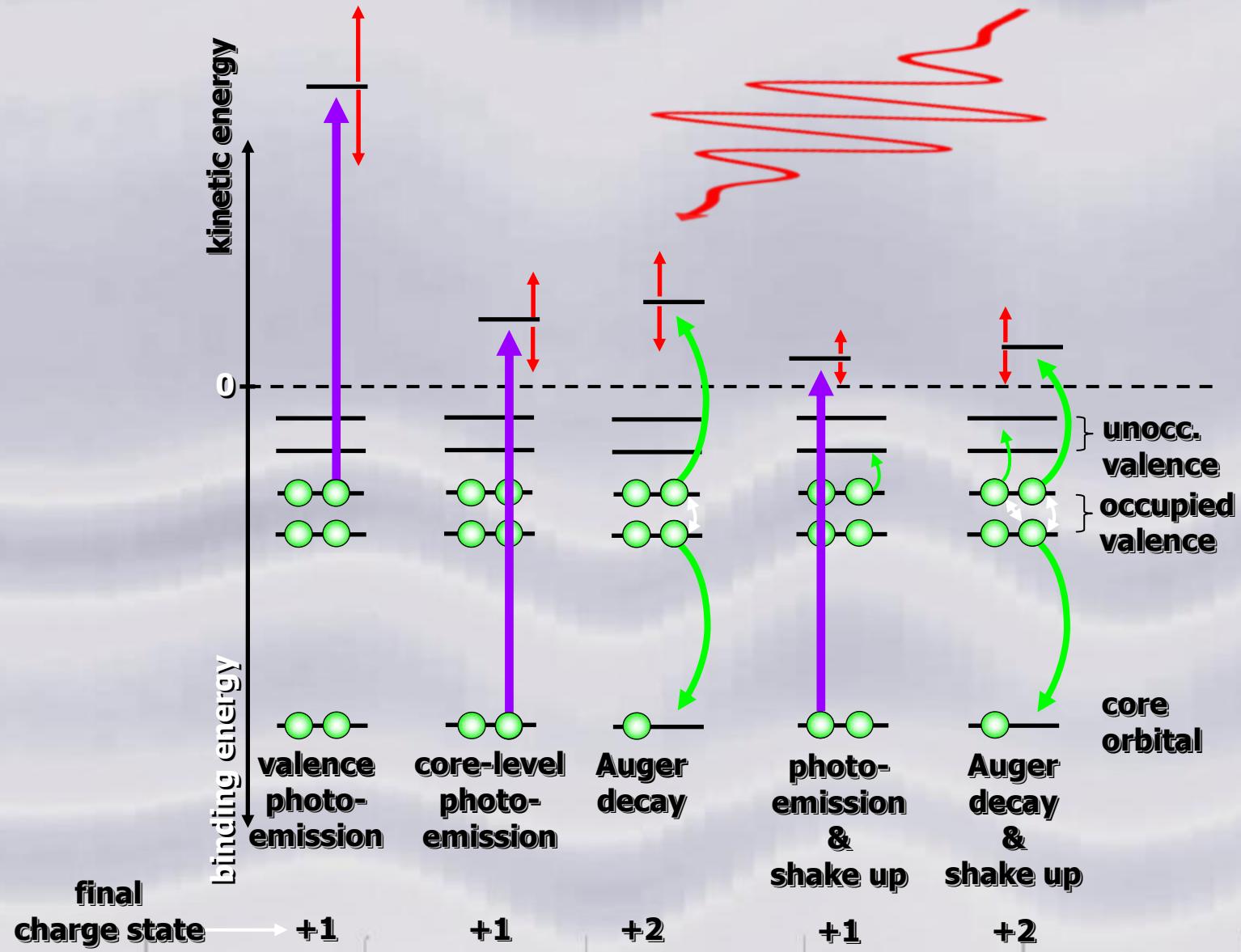
isolated sub-100-as pulses



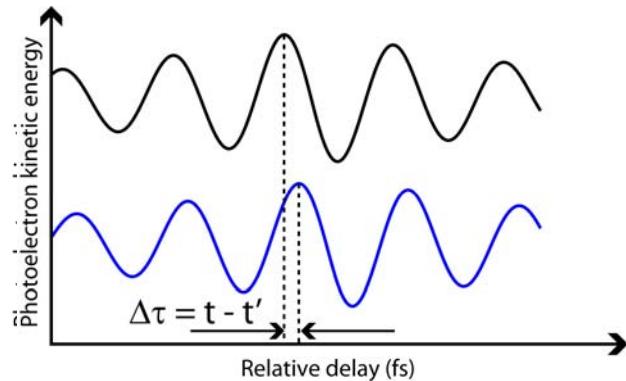
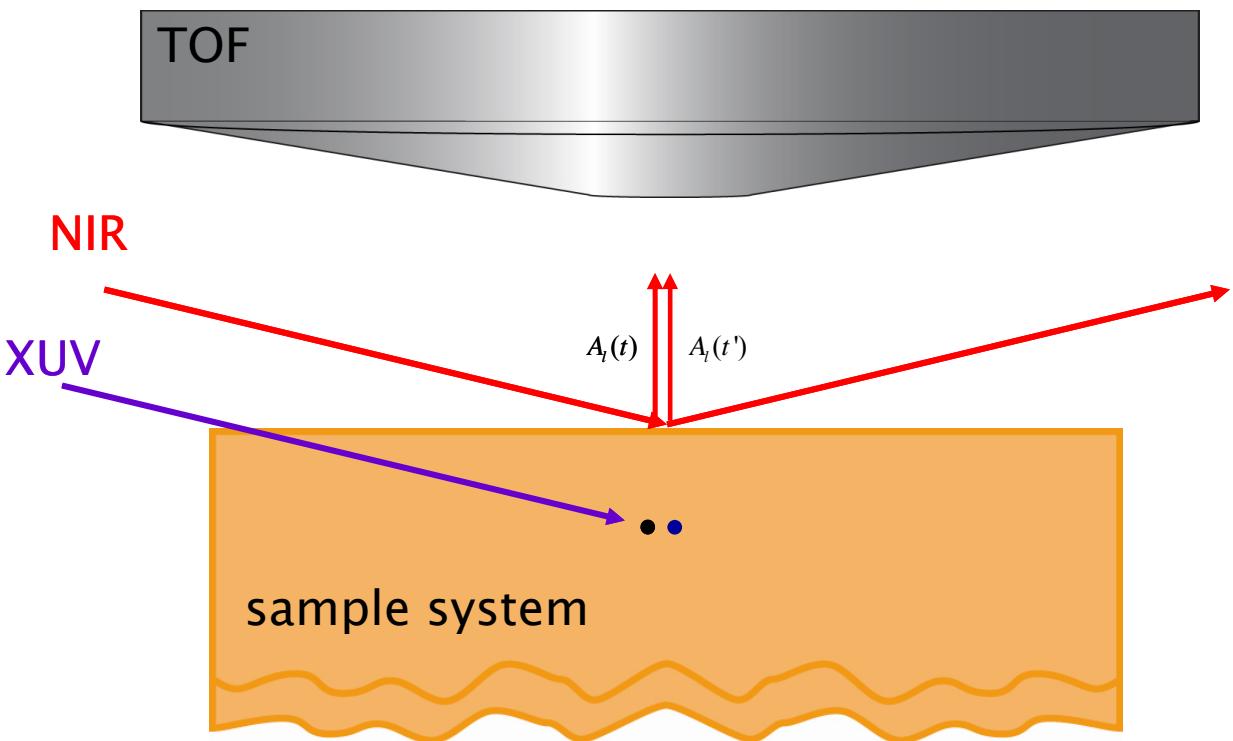
AS beamline



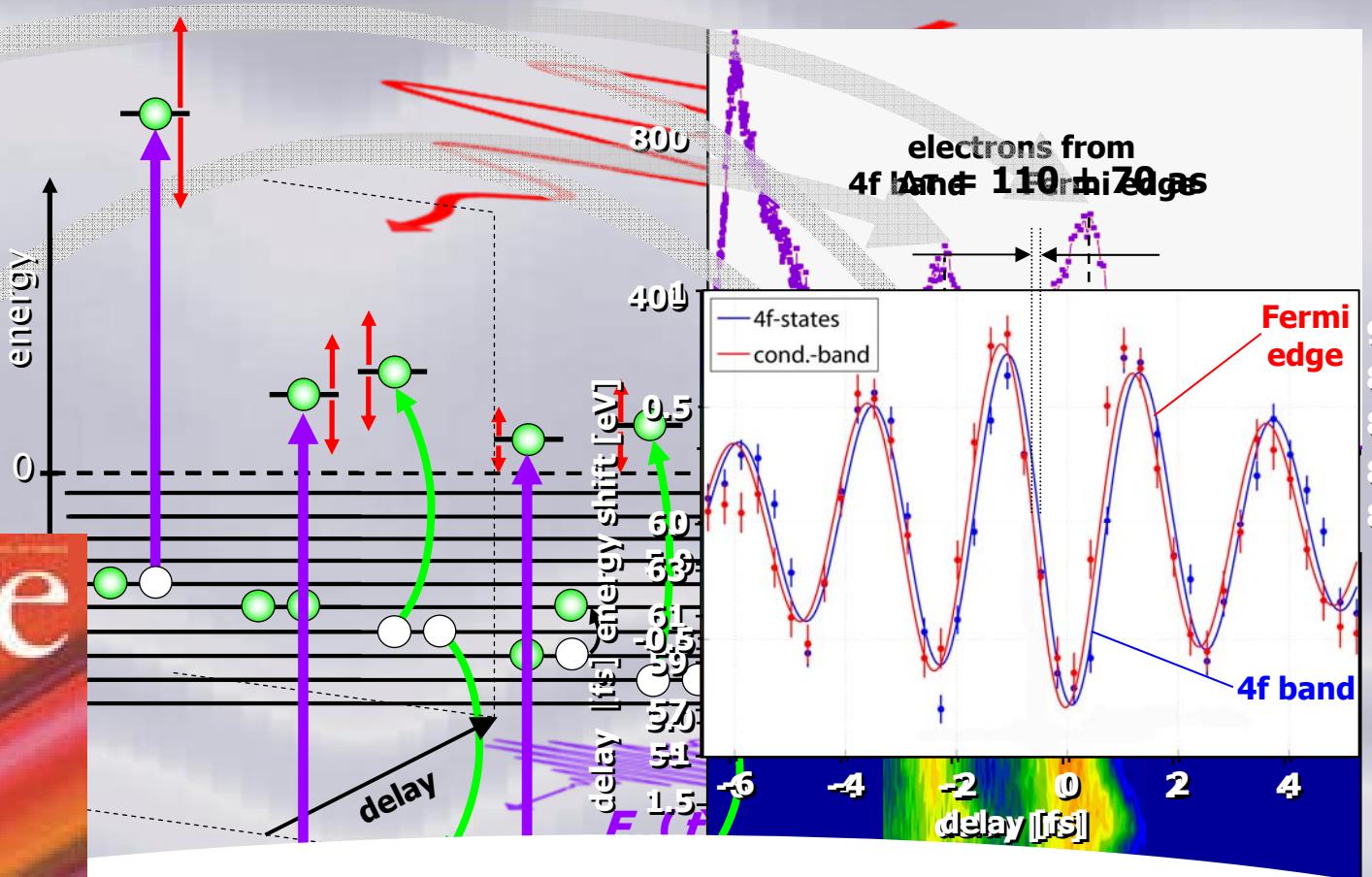
by means of strong-field-induced free-free transitions: **streaking**



Ionization of electrons in a solid sample



proof of principle: attosecond streaking of core-level (4f) & conduction-band (Fermi-edge) electrons in tungsten



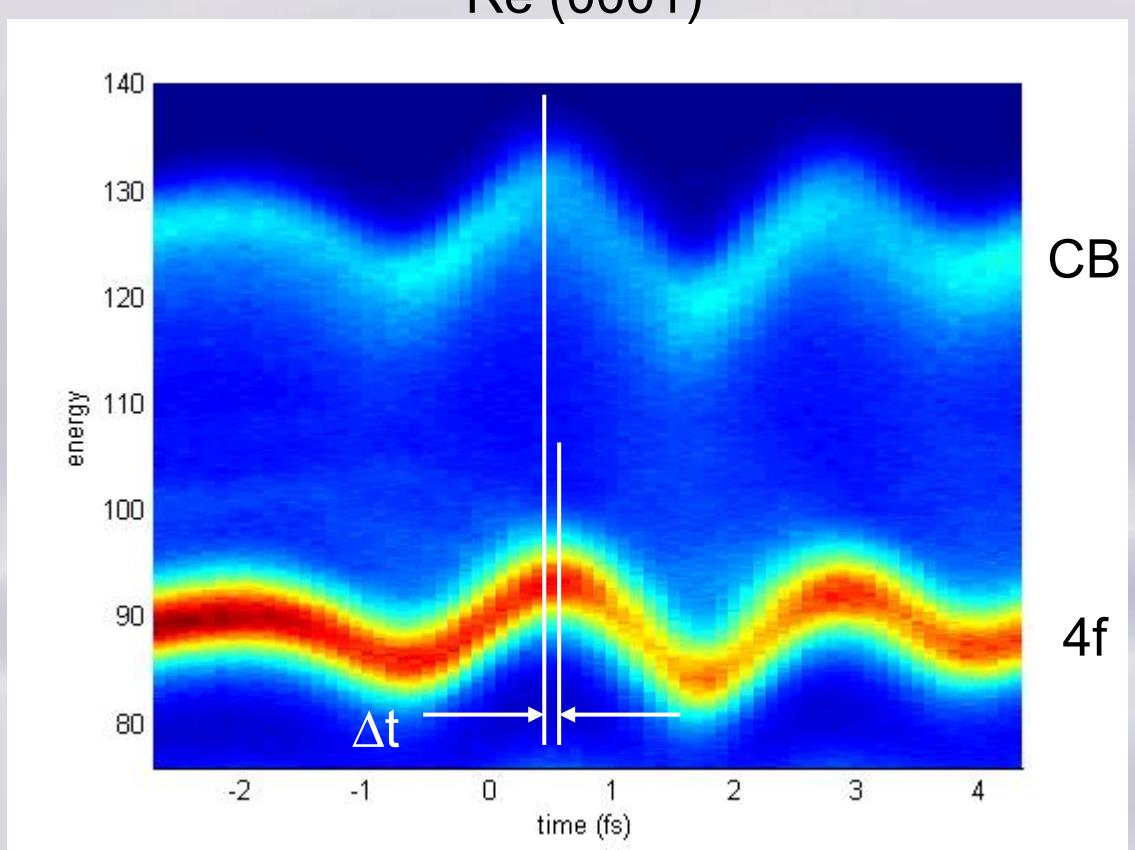
electrons from the 4f band reach the surface
 ~ 110 as later than those from the Fermi-edge

as spectroscopy of clean metal surface

Samples:
Re(0001) and W(110)

Photon energies:
90, 120, 130 eV

Core level electrons
delayed by 55 – 90 as.



Spectrogram analysis:
retrieval algorithm based on
analytical solution of TDSE

Delay of 4f core levels relative to CB:
 64 ± 10 as

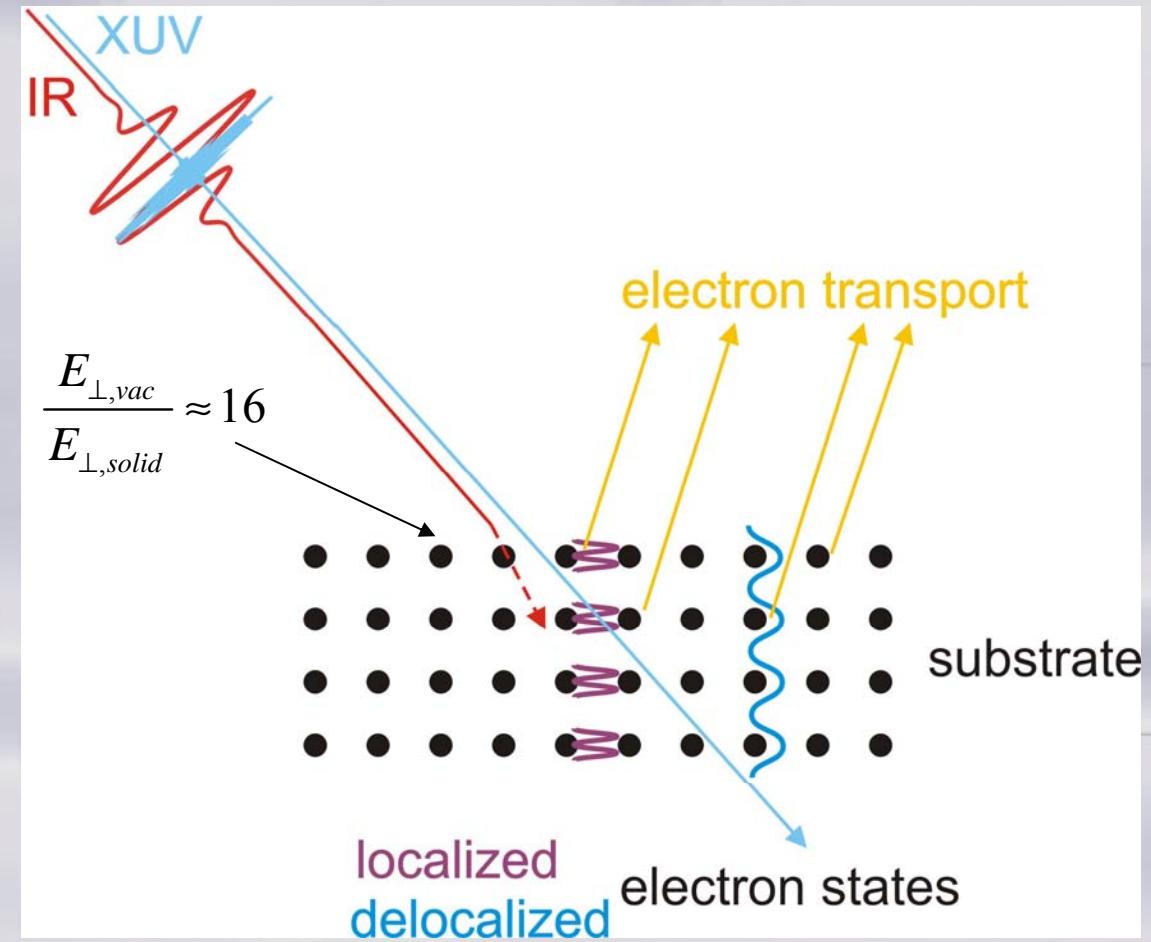
Origin of delay?

Propagation effect

Assumption:
streaking acts outside solid

- Different inelastic mean free path → different depth profile
- Different group velocity
(final-state effect?)

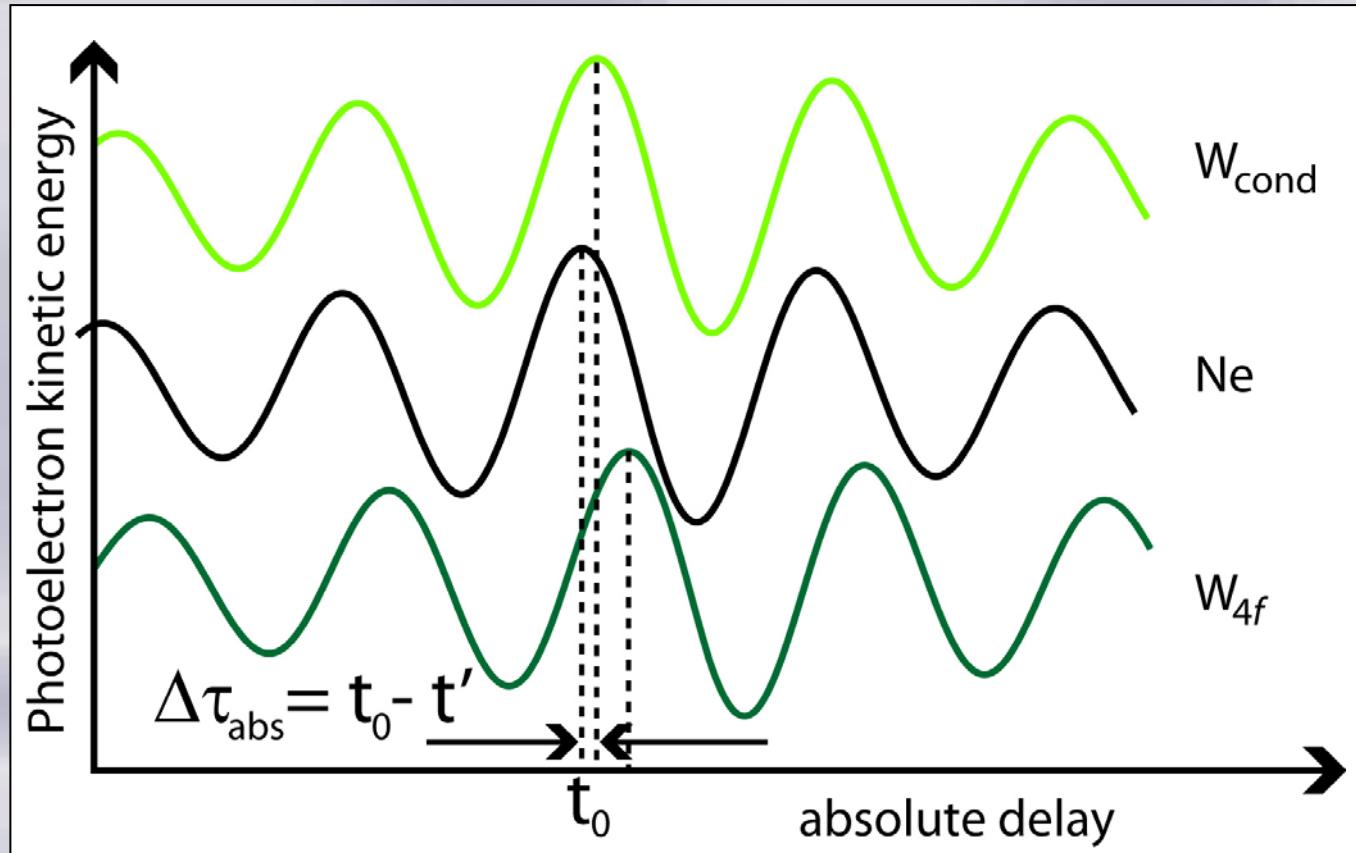
→ 4f emitted after CB



Theory:

- A. Kazansky et al., PRL 102, 177401 (2009).
C. Lemell et al., PRA 79, 62901 (2009).
C.-H. Zhang et al., PRL 102, 123601 (2009).
J.C. Baggesen et al., PRA 78, 32905 (2008).

Current Effort: Absolute Emission Time



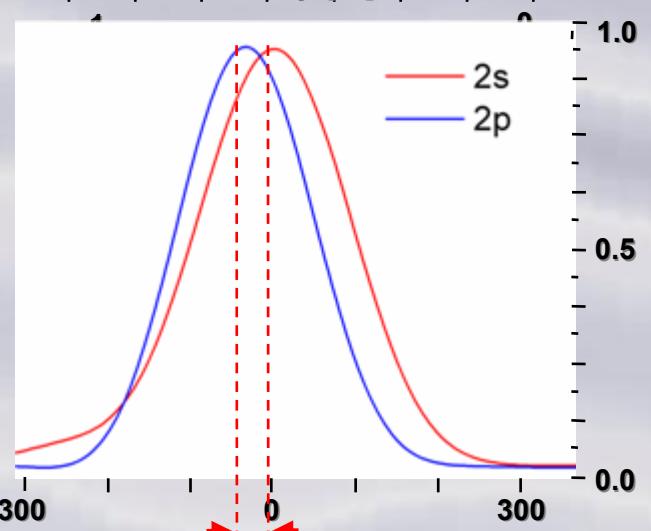
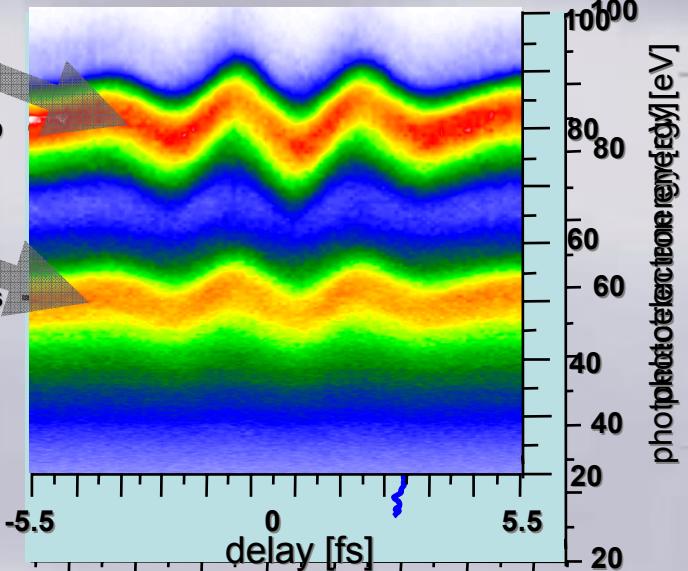
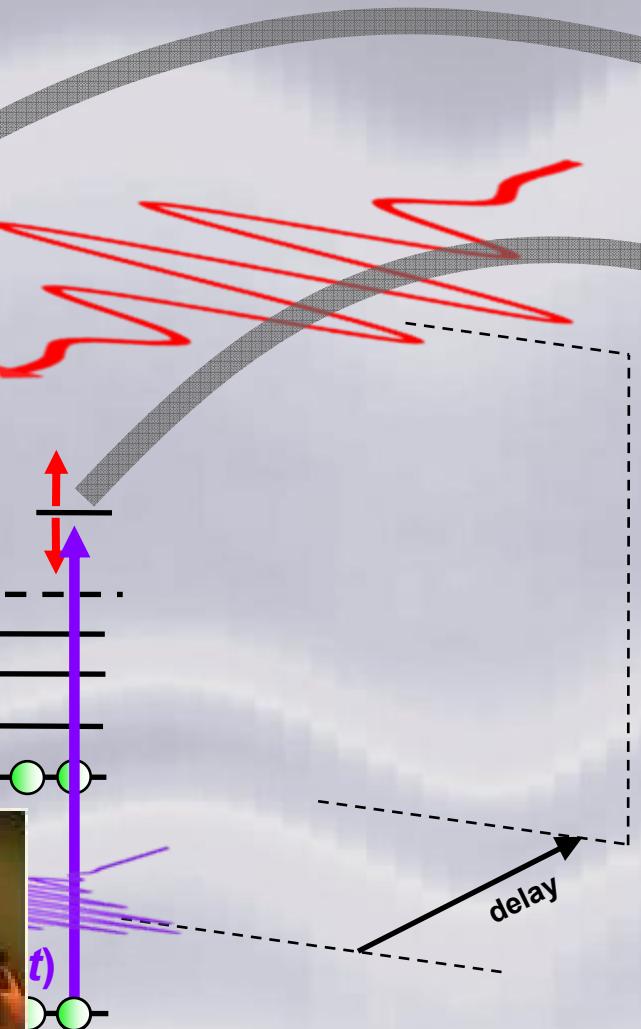
.....blow gas on the sample.....

geometric effects....

What about the reference? Ne 2s / 2p gas phase

kinetic energy

0



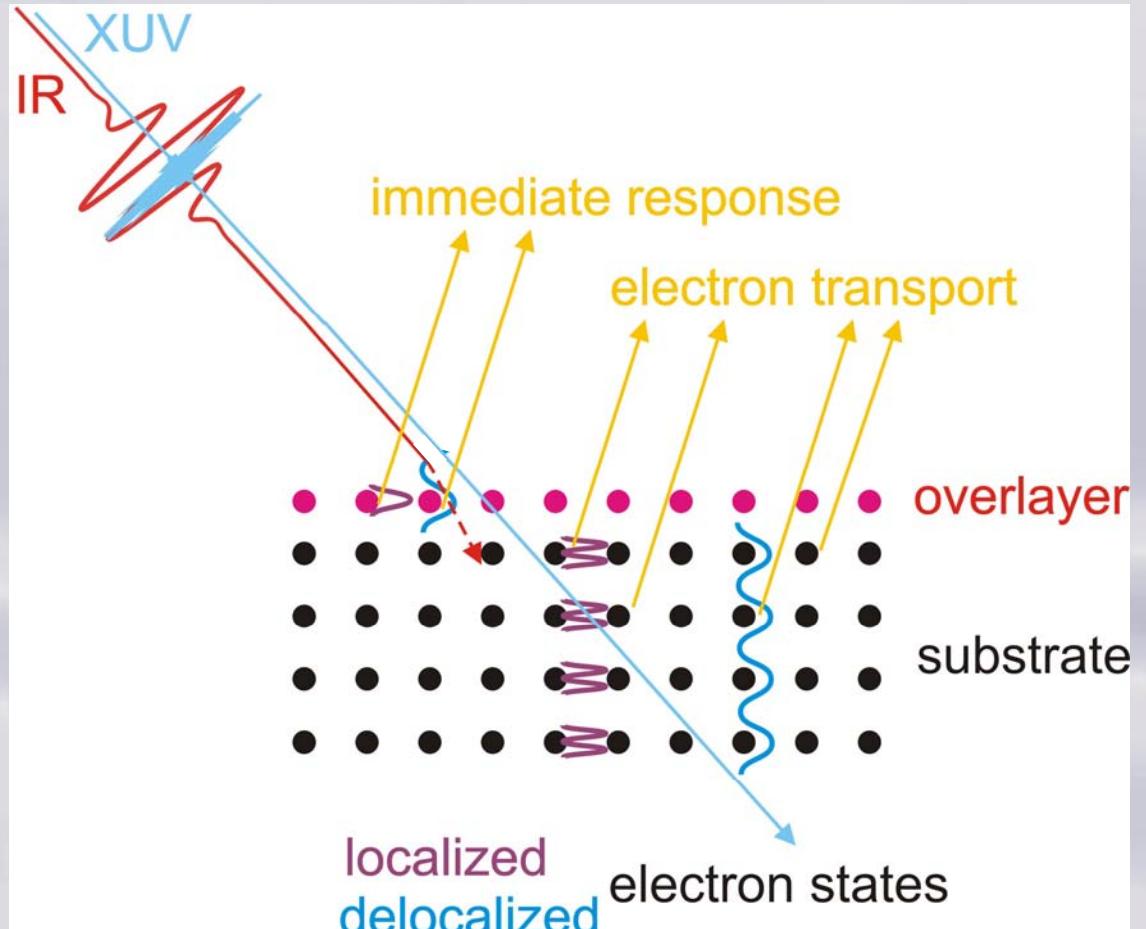
emission of low-energy photoelectron is delayed by about 20 as

Duration of photoemission?

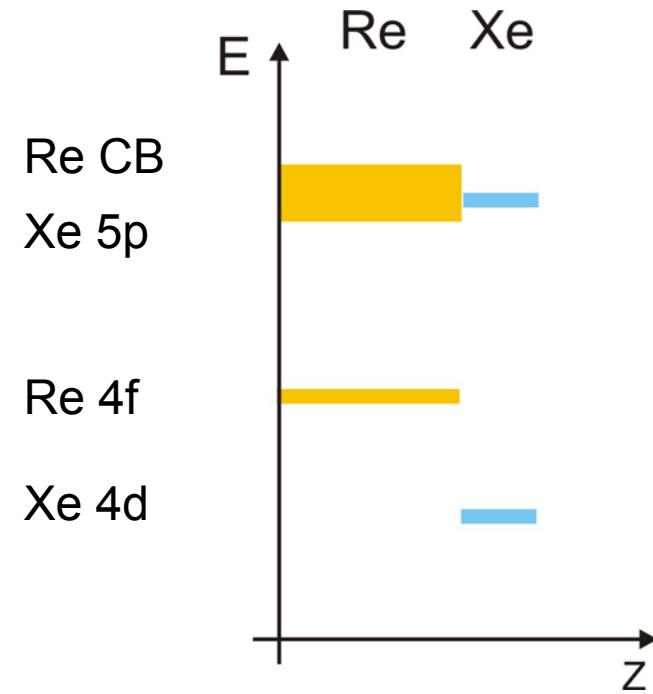
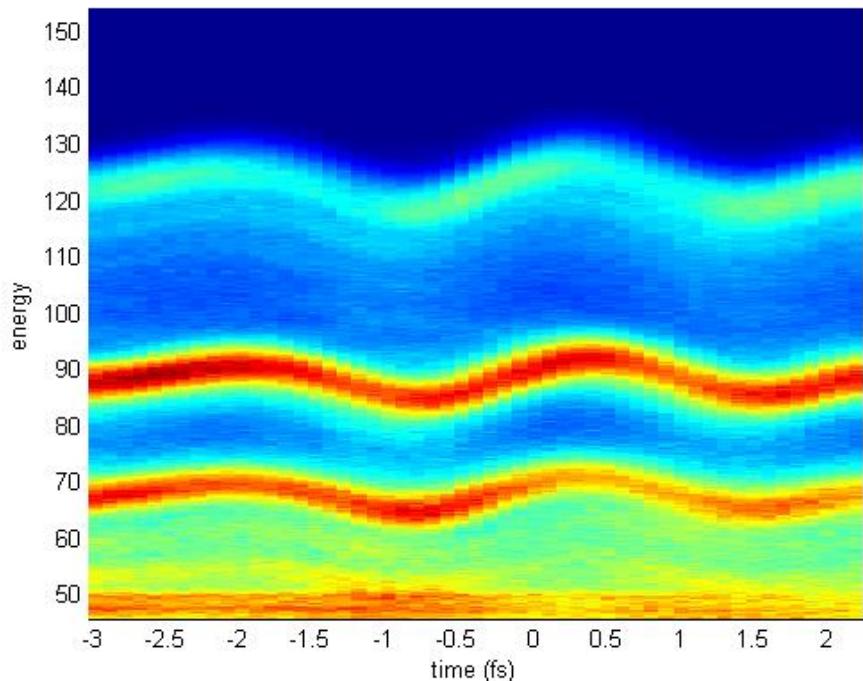
Idea:

Introduce electronic reference state localized at the surface

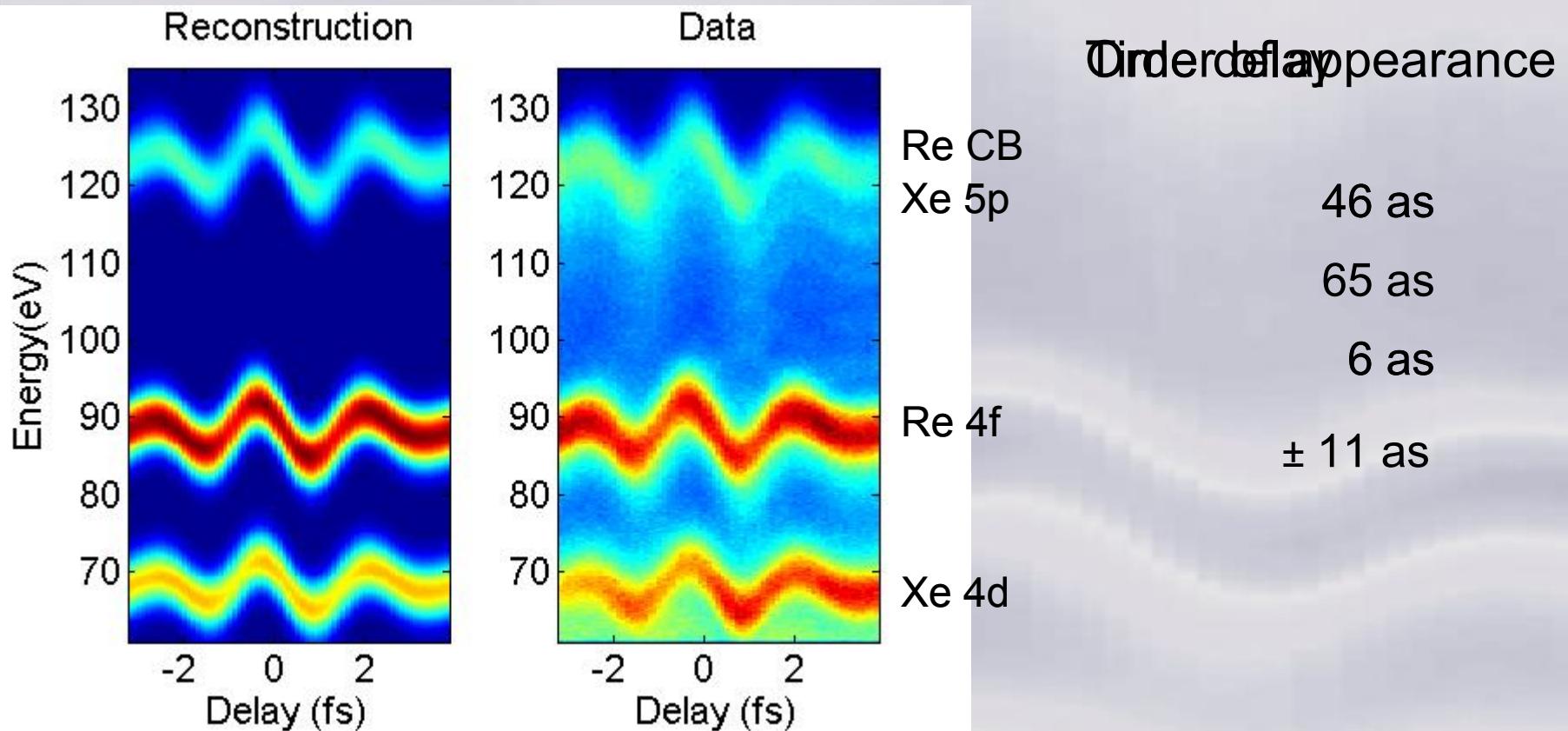
→ time zero marker



Xe monolayer on Re



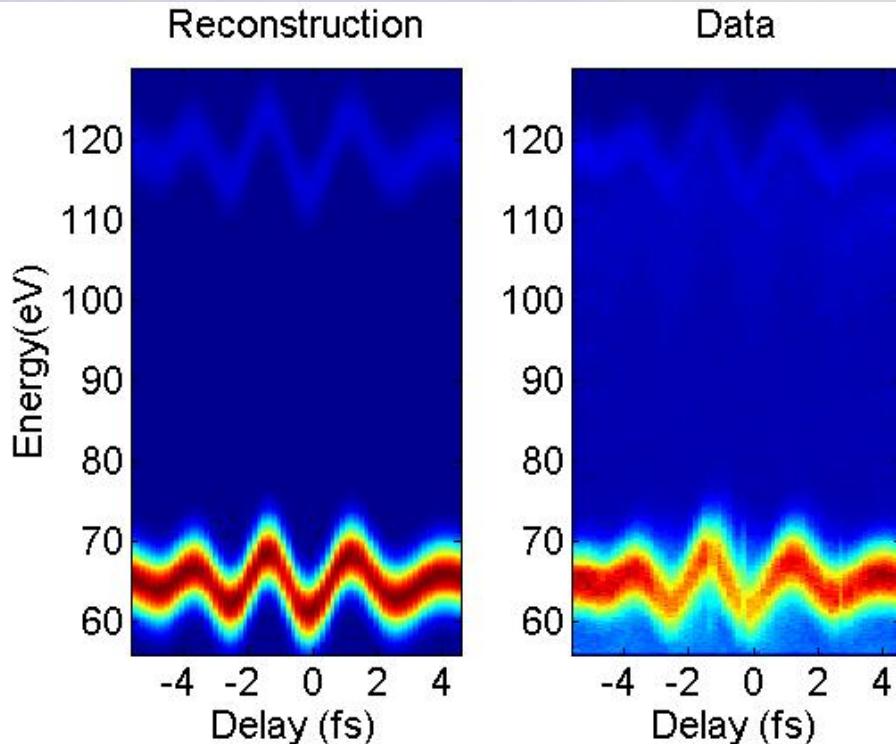
Xe monolayer on Re



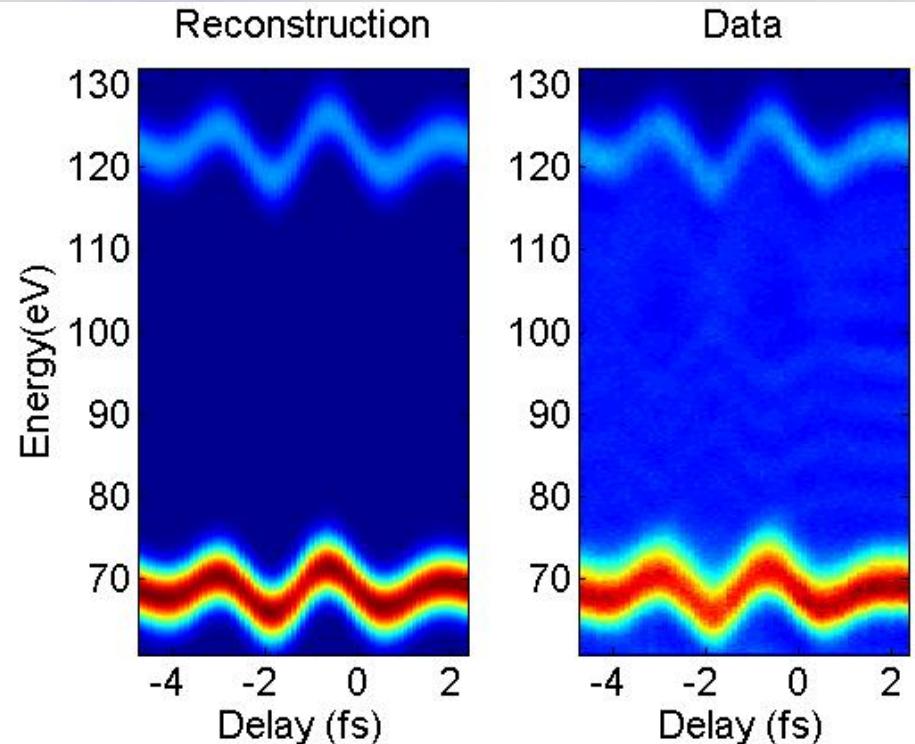
NIR intensity $\sim 5.5 \cdot 10^{11} \text{ W/cm}^2$

Comparison atomic vs. solid xenon

atomic xenon



solid xenon
(50 layers)

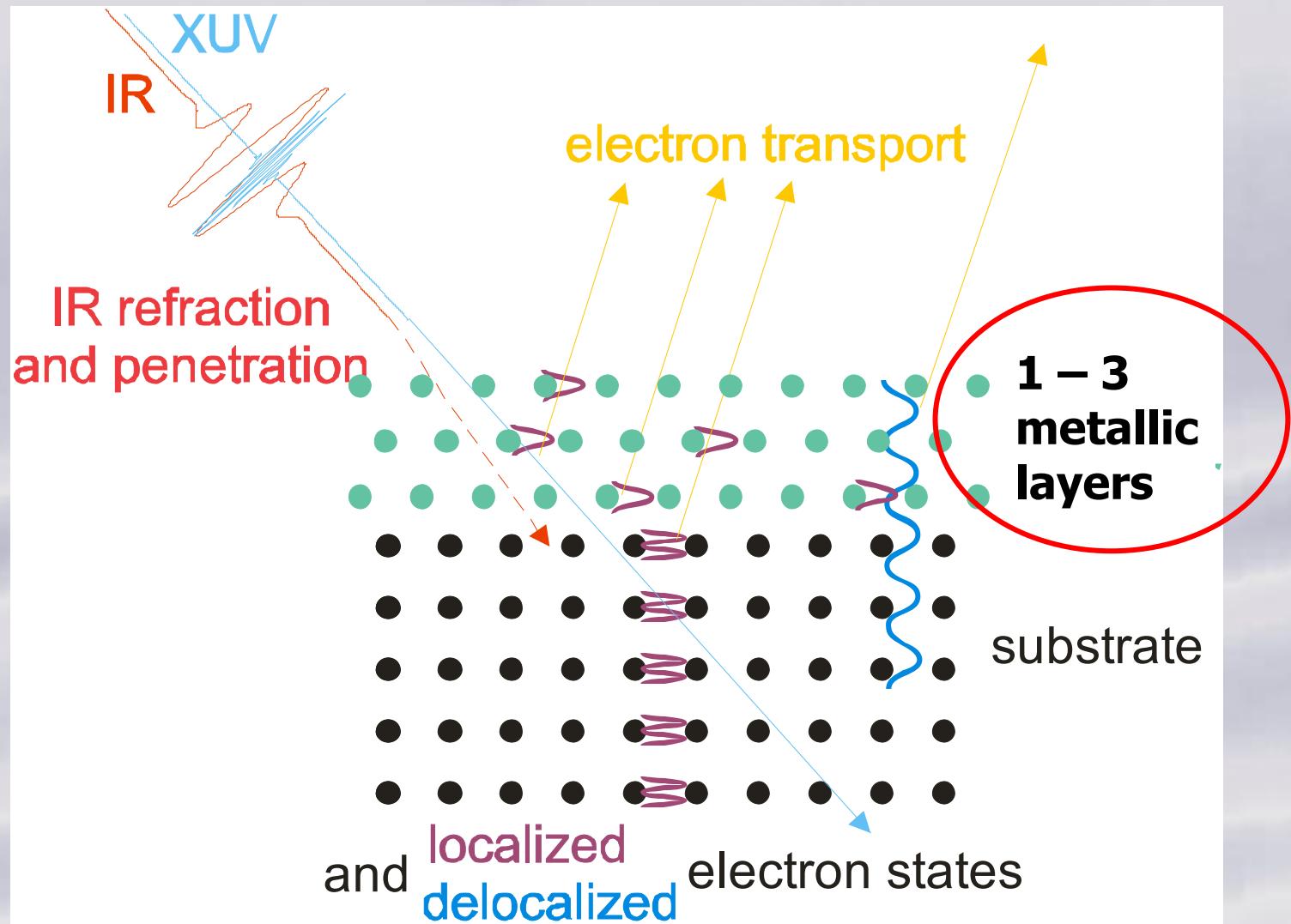


+22 ± 12 as
4d is first

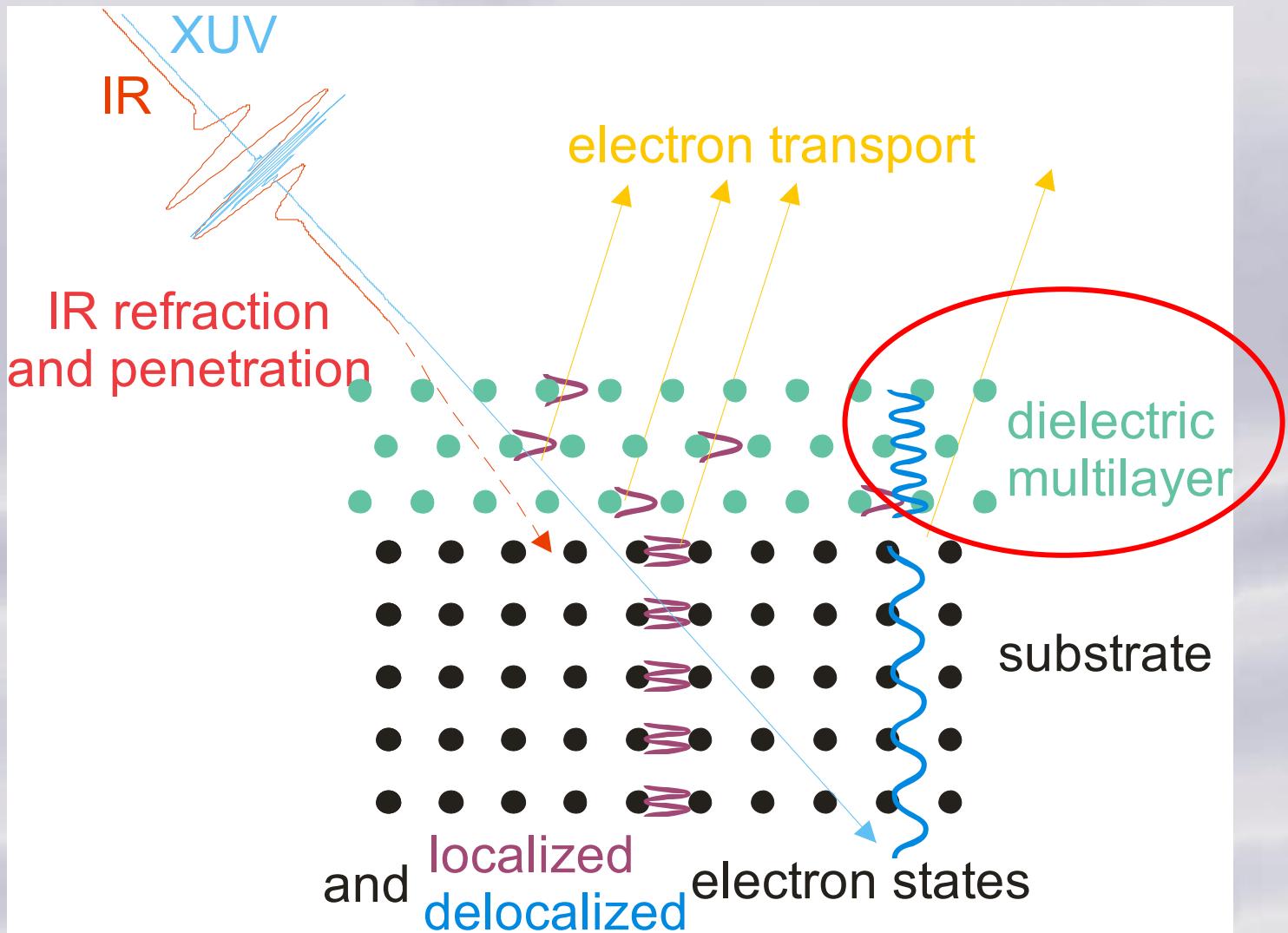
electron
propagation
effects

-20 ± 3 as
5p is first

Electron transport through defined adlayers



Electron transport through defined adlayers



Outlook

- Generation of UV pulses for excitation of molecules: UV-pump/XUV-probe
- Charge-transfer in molecules and soldis

Attosecond 2PPE

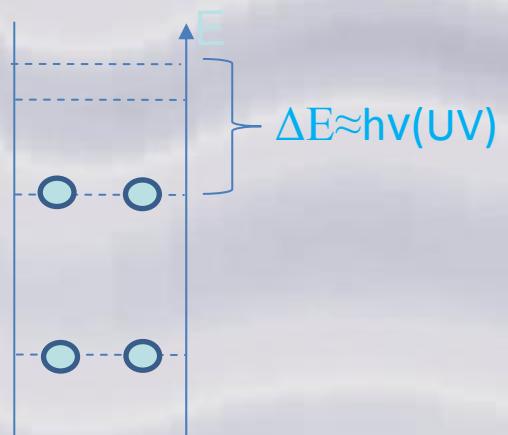
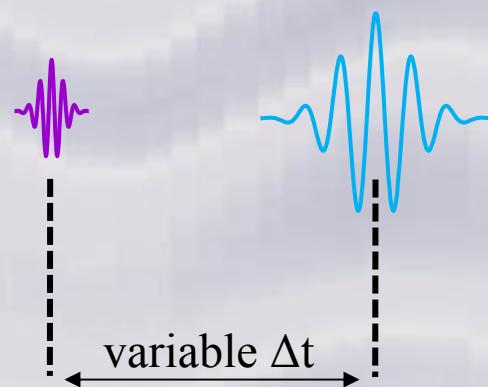
Goal: study excited state dynamics with as to few-fs time resolution

Requirements:

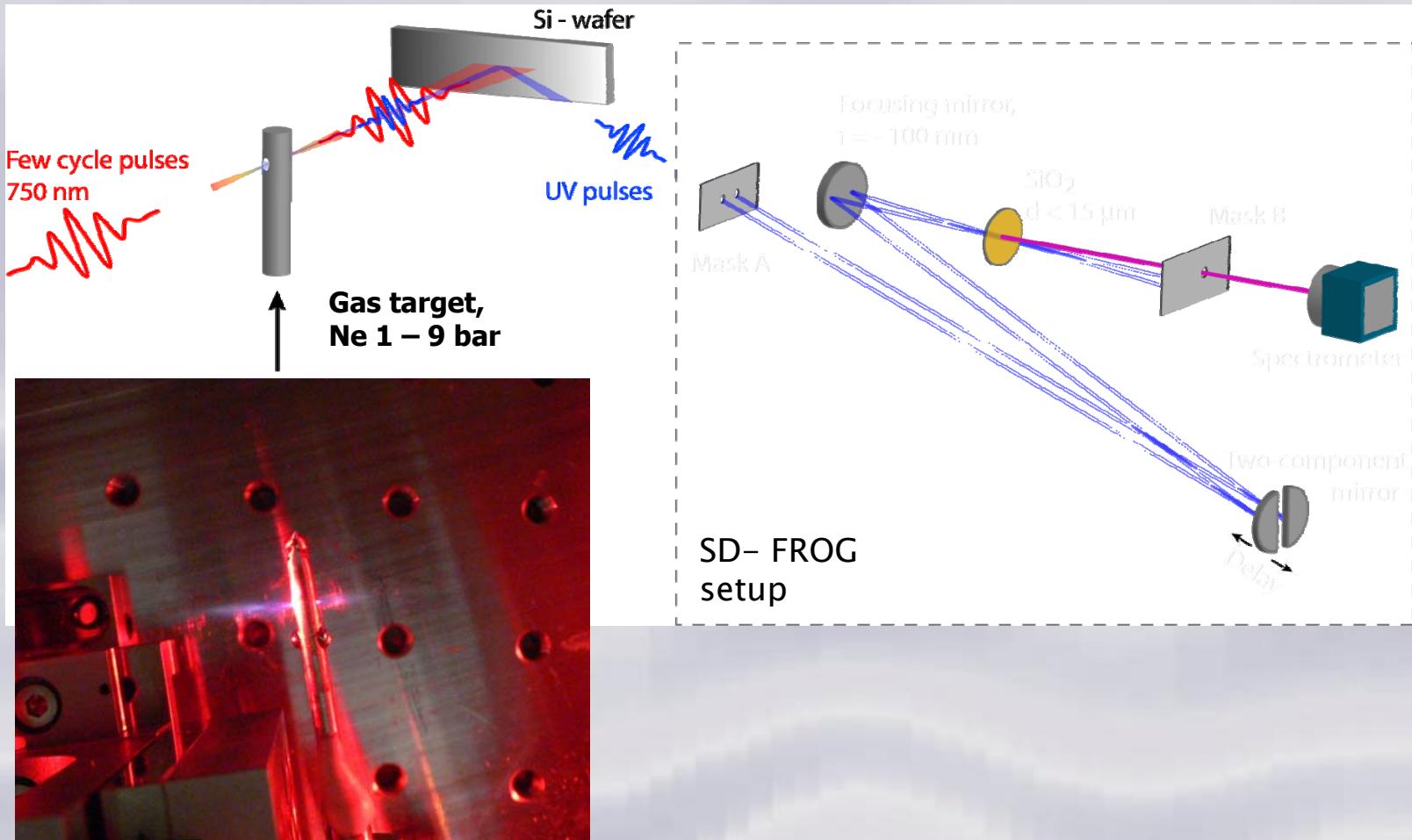
few-cycle UV pump pulses ($h\nu = 4\text{-}6 \text{ eV}$)

synchronized with

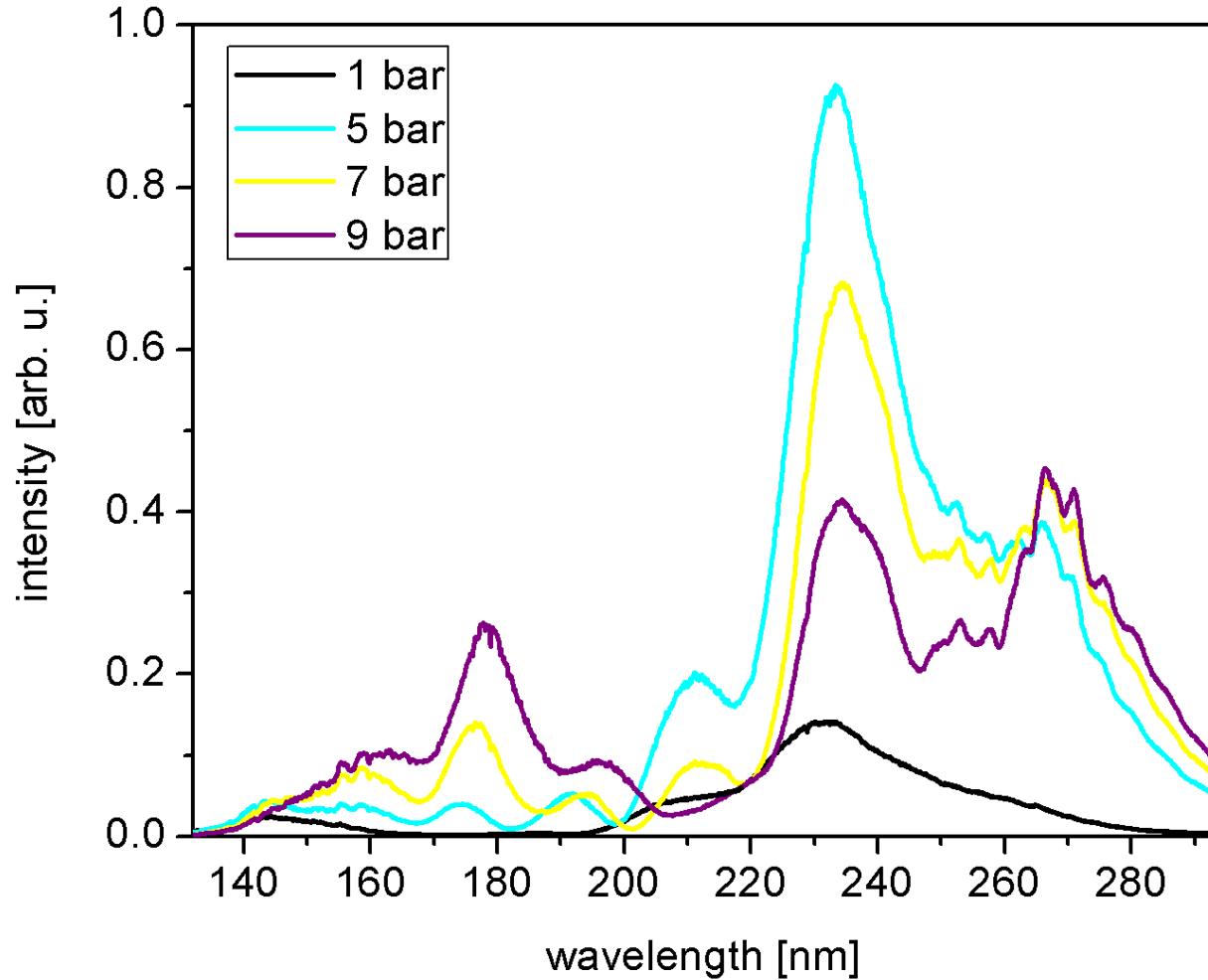
Isolated XUV probe pulse ($h\nu = 80\text{-}130 \text{ eV}$)



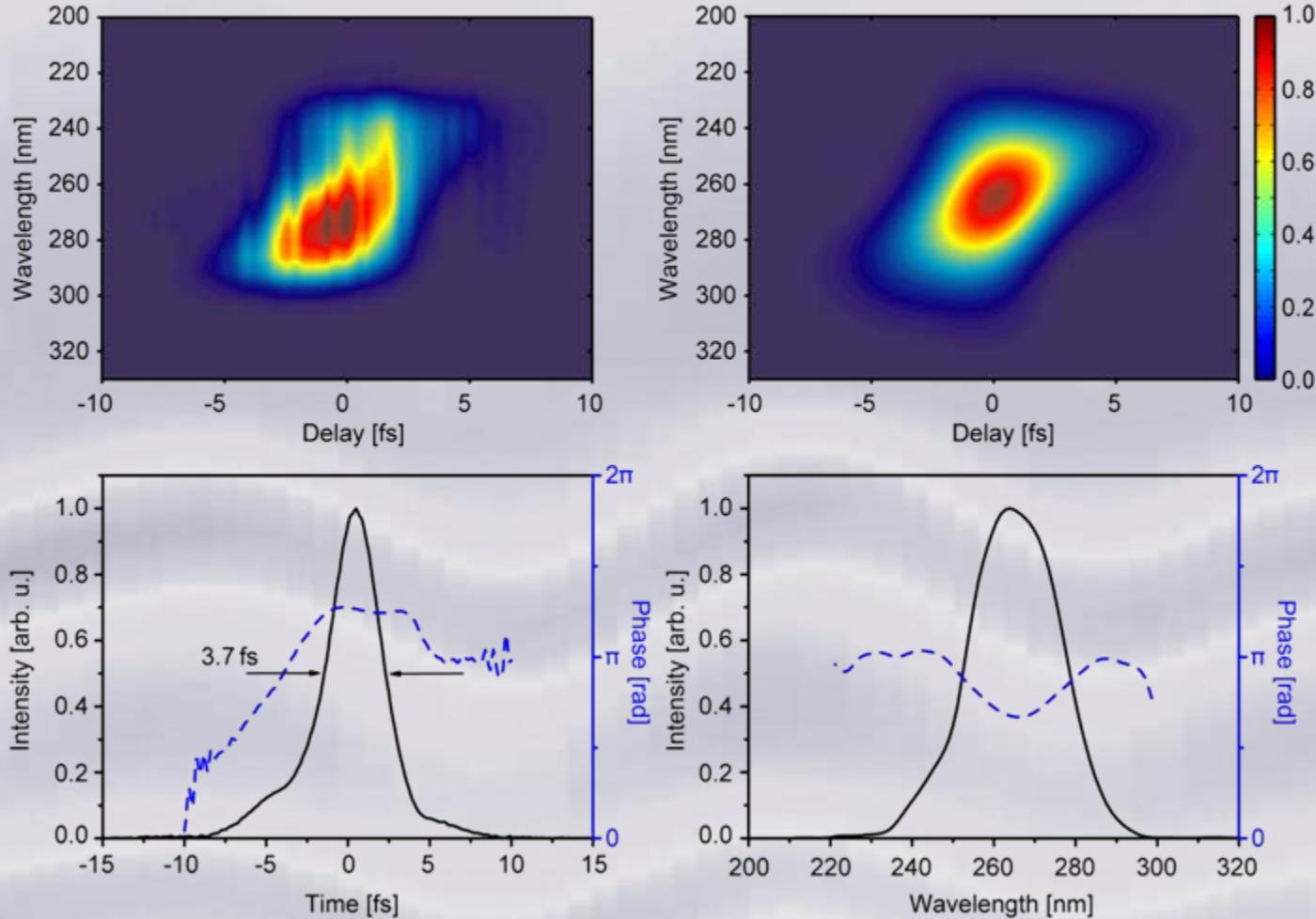
Intense pulses in the deep ultraviolet



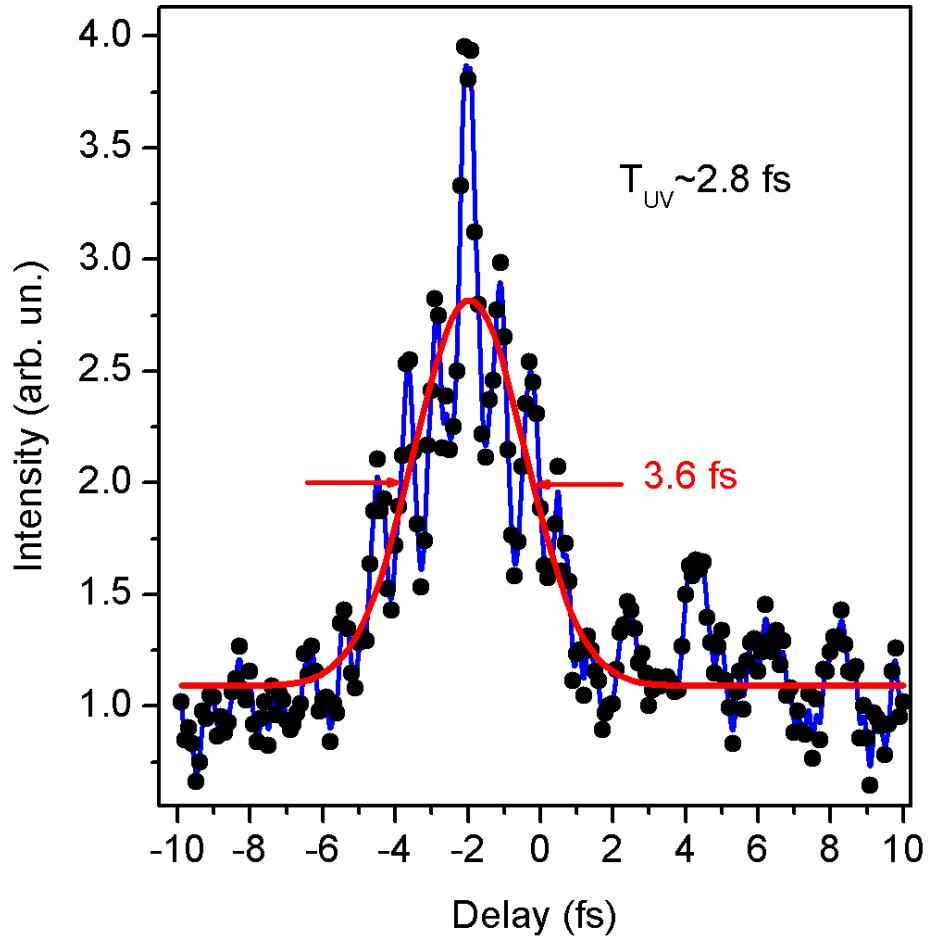
Emission of UV light at different target pressures



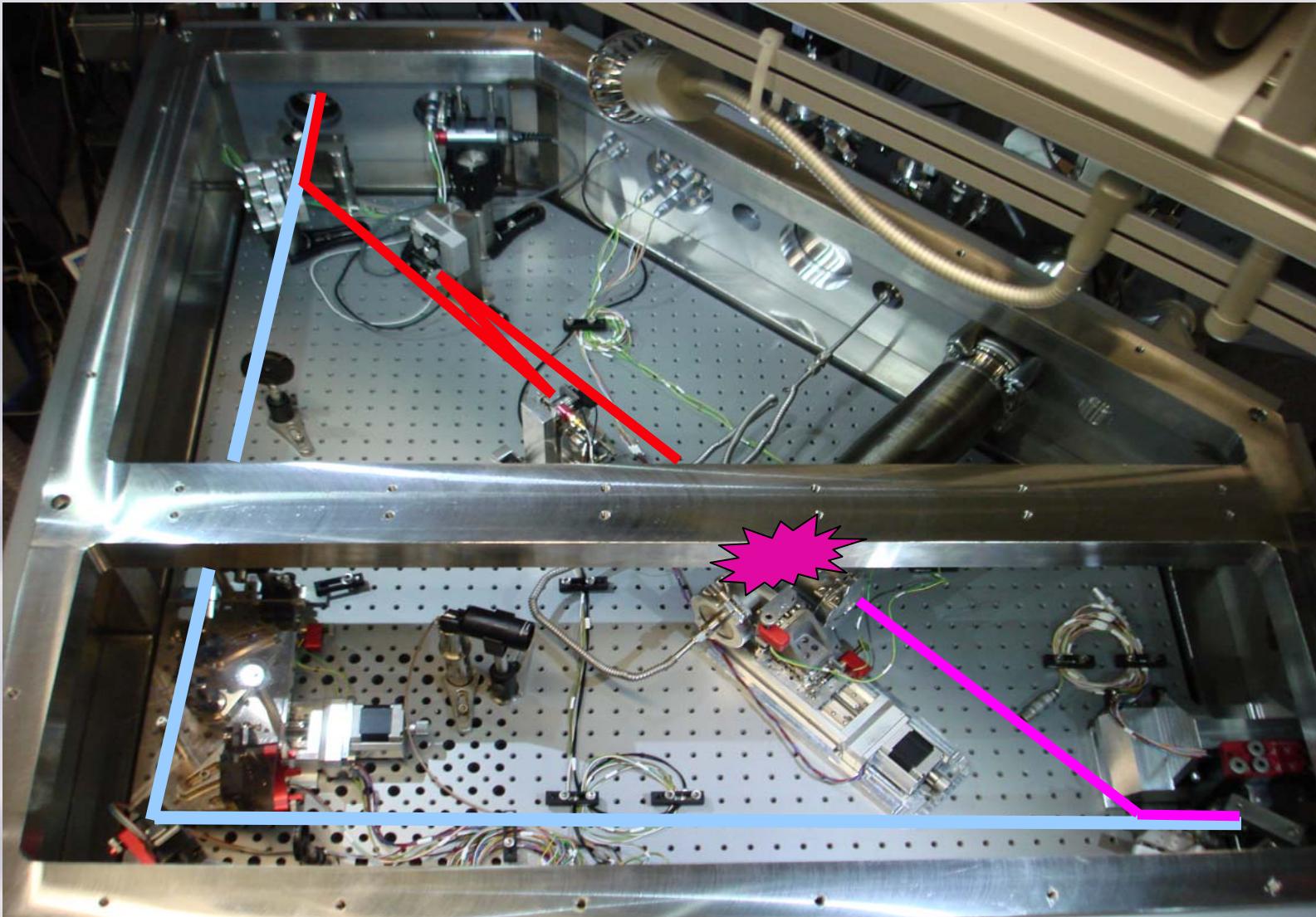
Intense ($>2 \mu\text{J}$) pulses in the deep ultraviolet



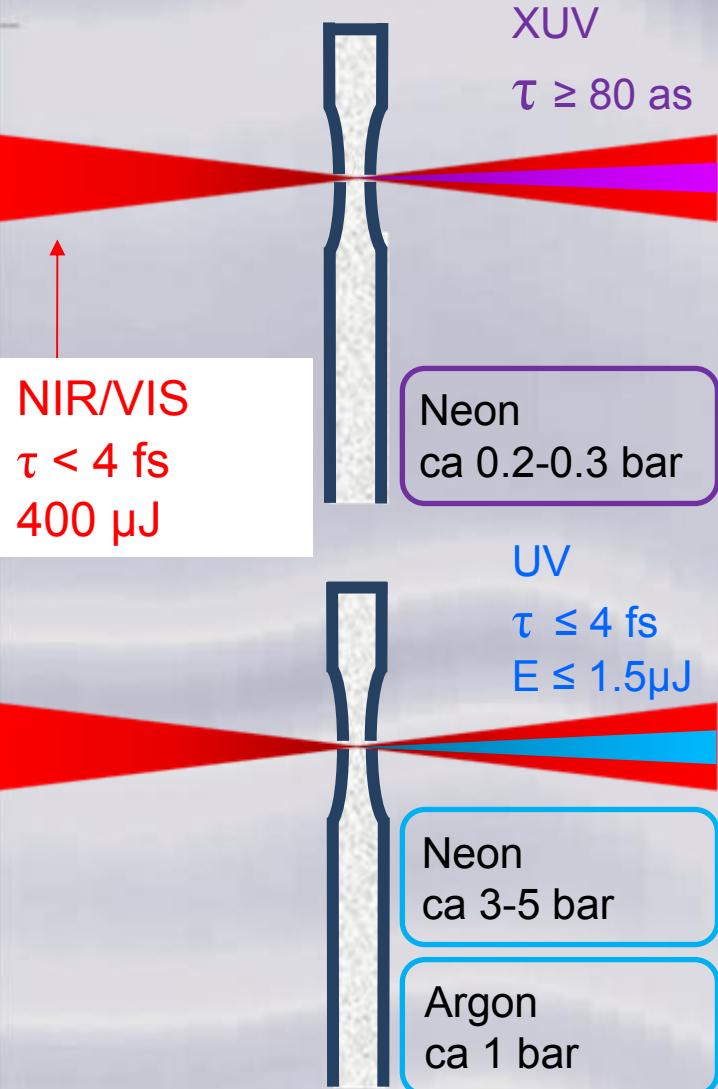
Autocorrelation trace on Kr ion yield



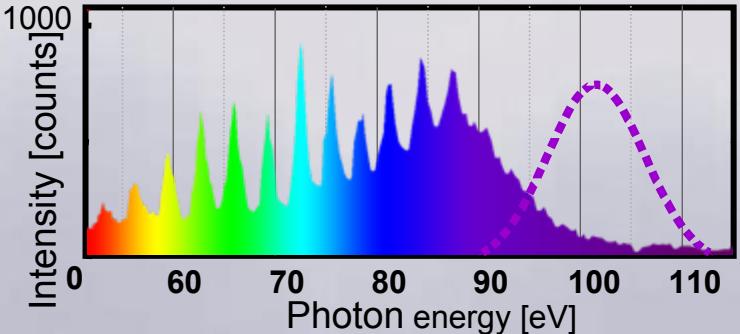
delay chamber



Collinear generation of UV and XUV



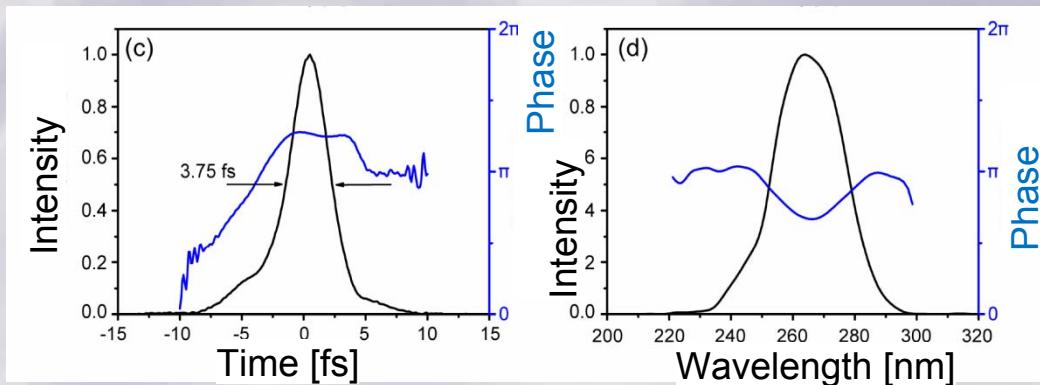
Isolated XUV attosecond pulses



A. Baltuska *et al.*, *Nature* 421, 611 (2003)

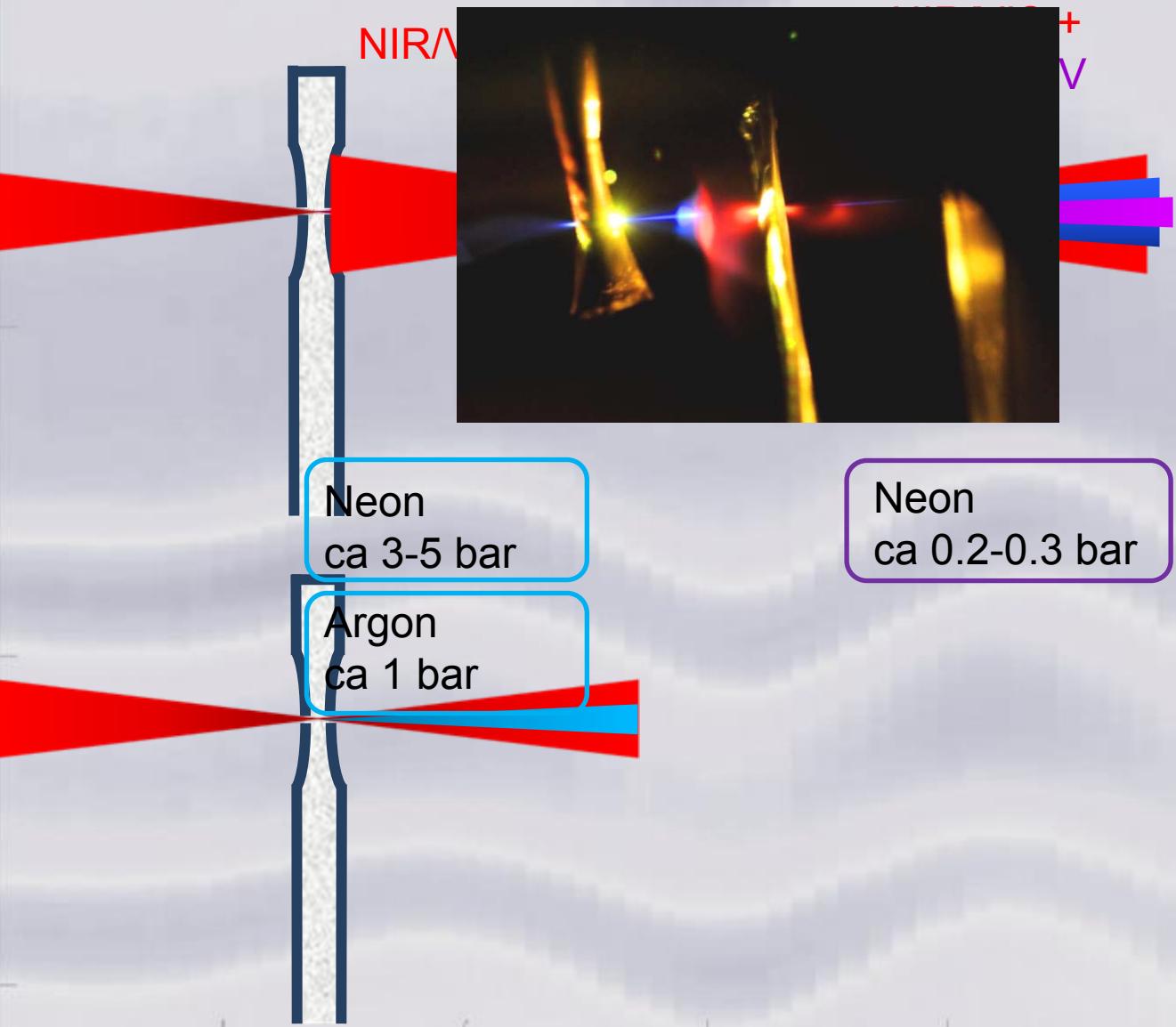
E. Goulielmakis *et al.*, *Science* Vol. 320 (2008)

Intense sub-4 fs pulses in the deep UV



U. Graf *et al.*, *Optics Express* 16, 18956 (2008)

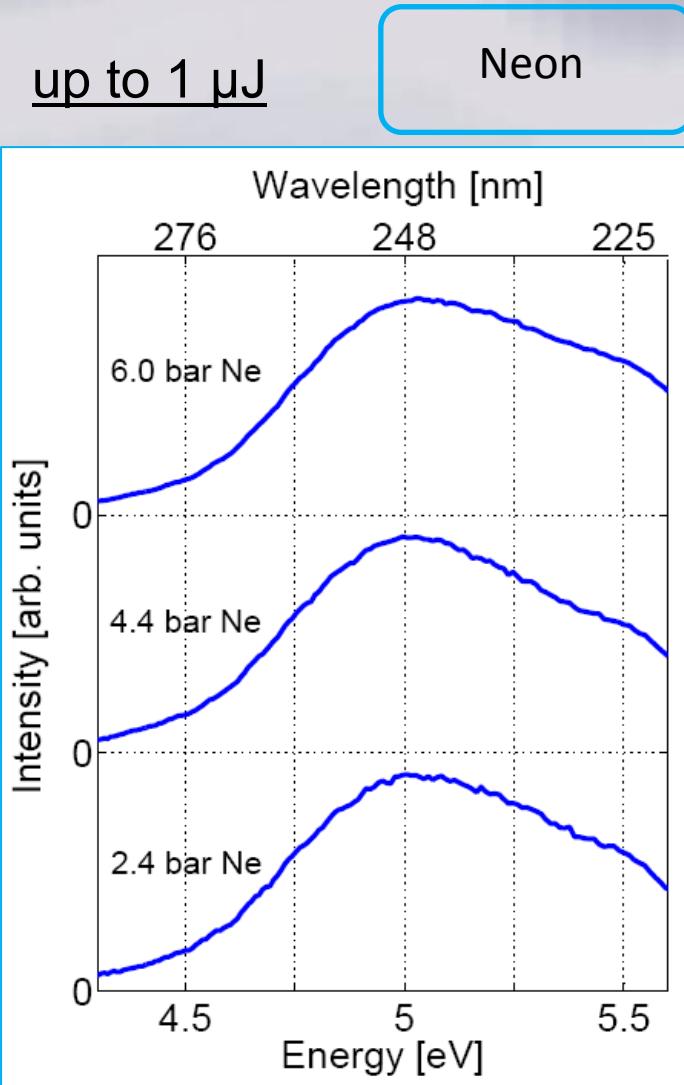
Collinear generation of UV and XUV



Collinear generation of UV and XUV

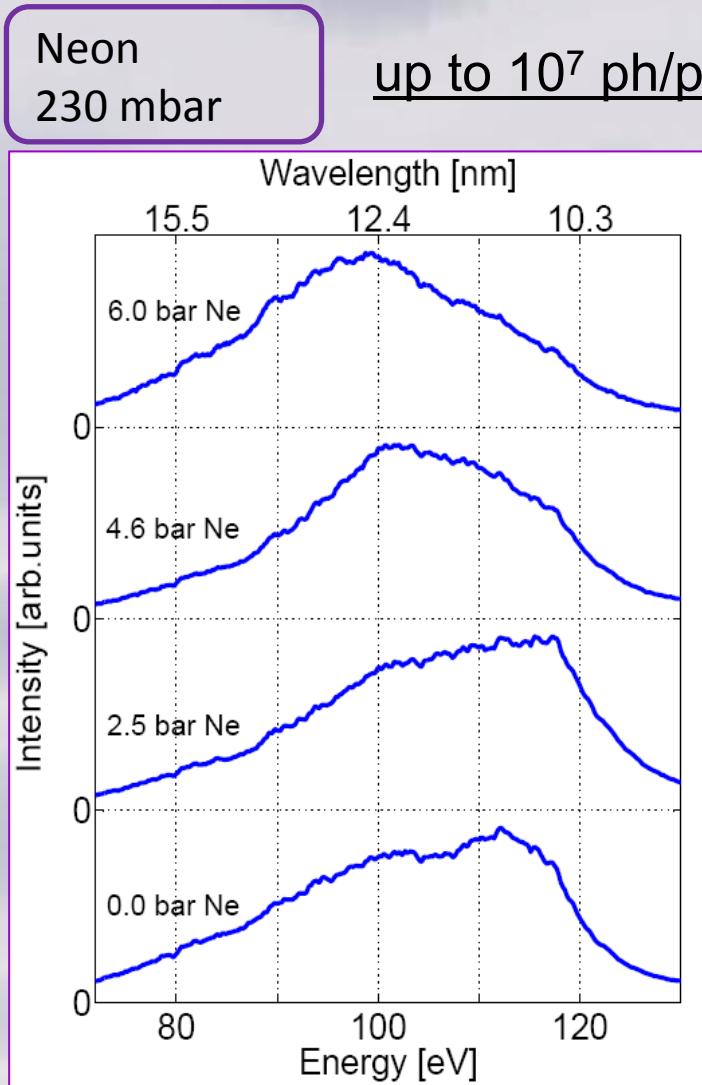
up to 1 μ J

Neon



Neon
230 mbar

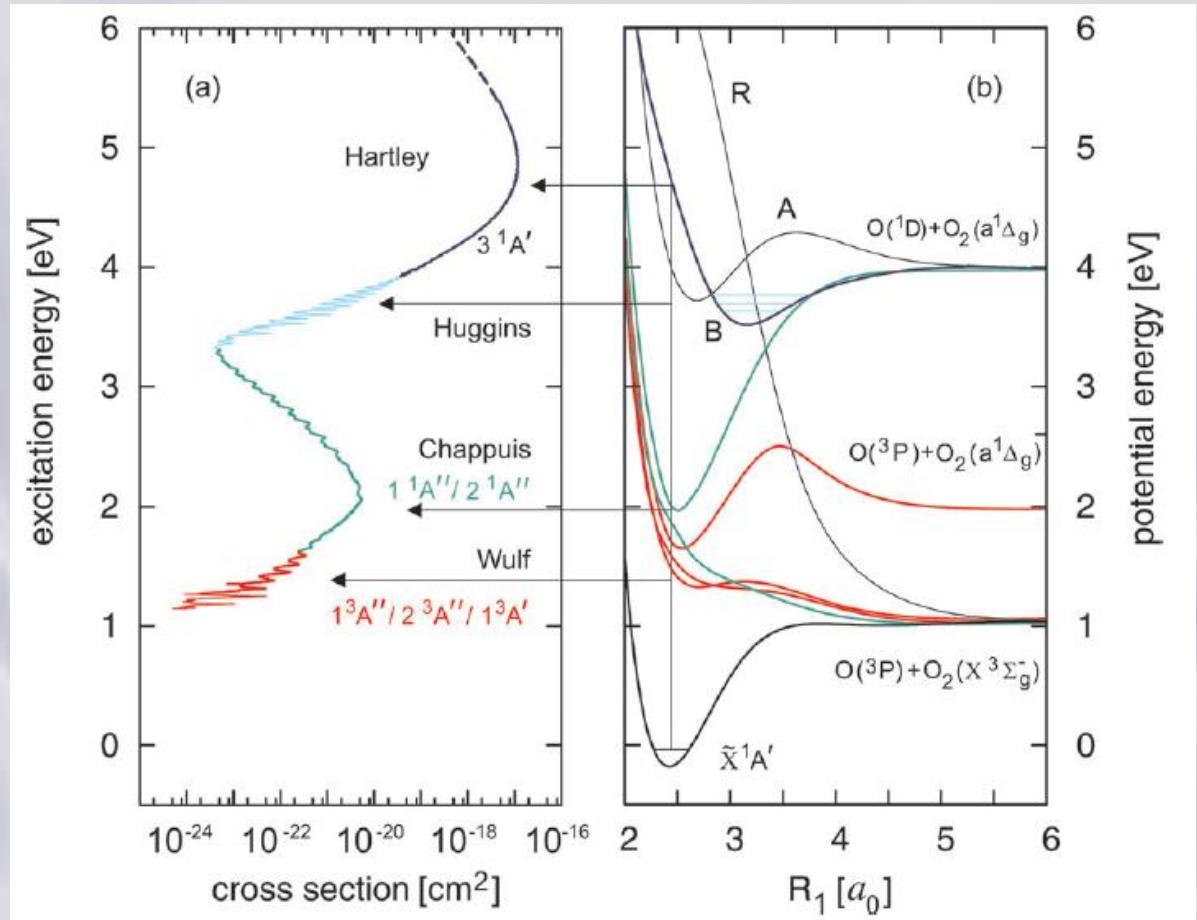
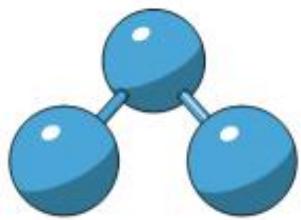
up to 10^7 ph/pulse



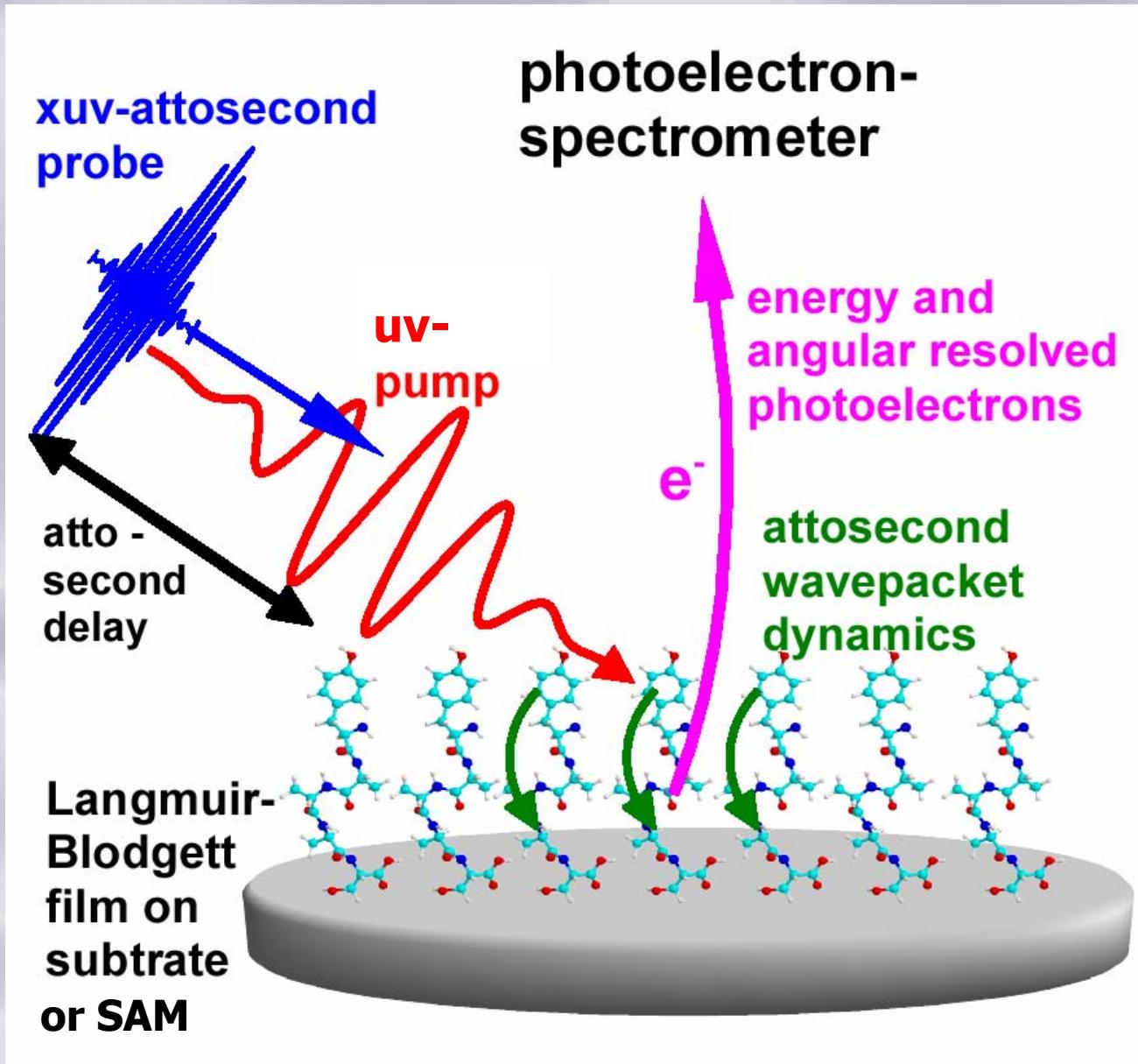
Exciting an electron wavepacket in a molecule

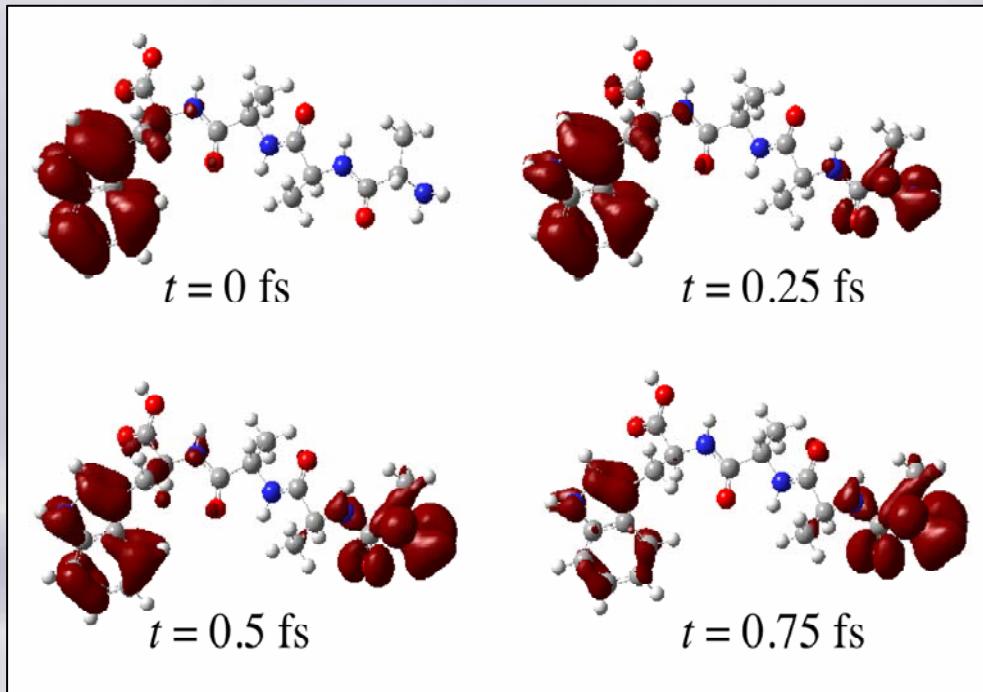
**Sampling the evolution
with XUV AS-pulses**

**First candidate:
ozone**

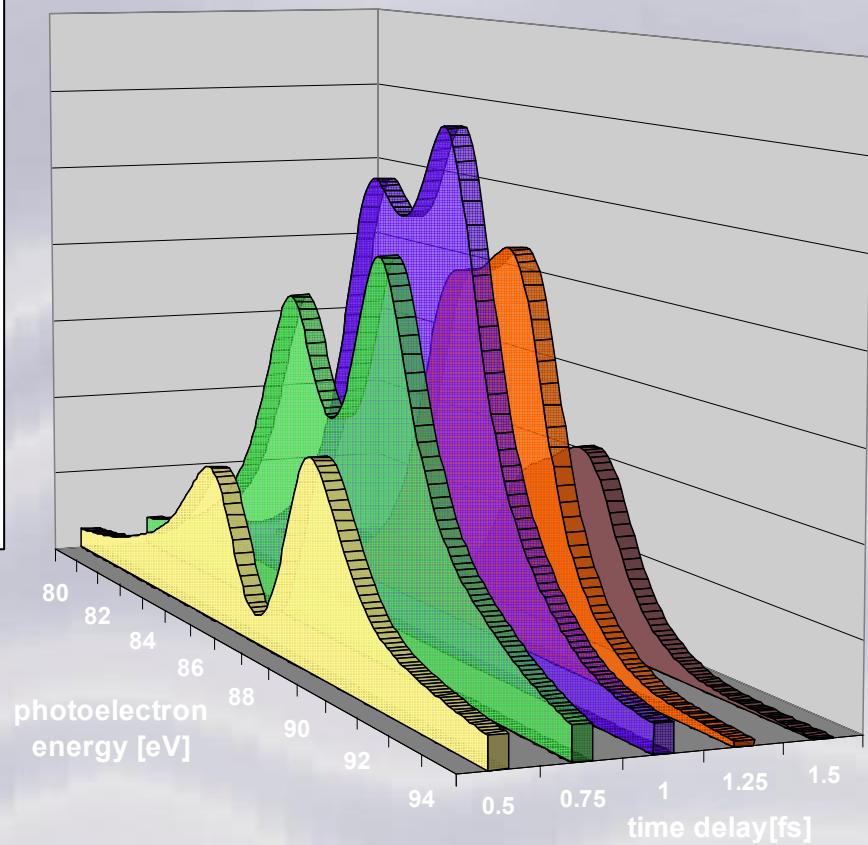


Oriented molecules on a surface





F. Remacle, R. D. Levine, *PNAS* 103, 6793 (2005)



Colleagues & Cooperations

F. Krausz

N. Karpowics

I. Grguras
Jobst

F. Reiter

M. Hassan
theory:

R. Ernststorfer

E. Goulielmakis

M. Fiess

B. Horvath

T. Wittmann

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XUV optics & spectroscopy:

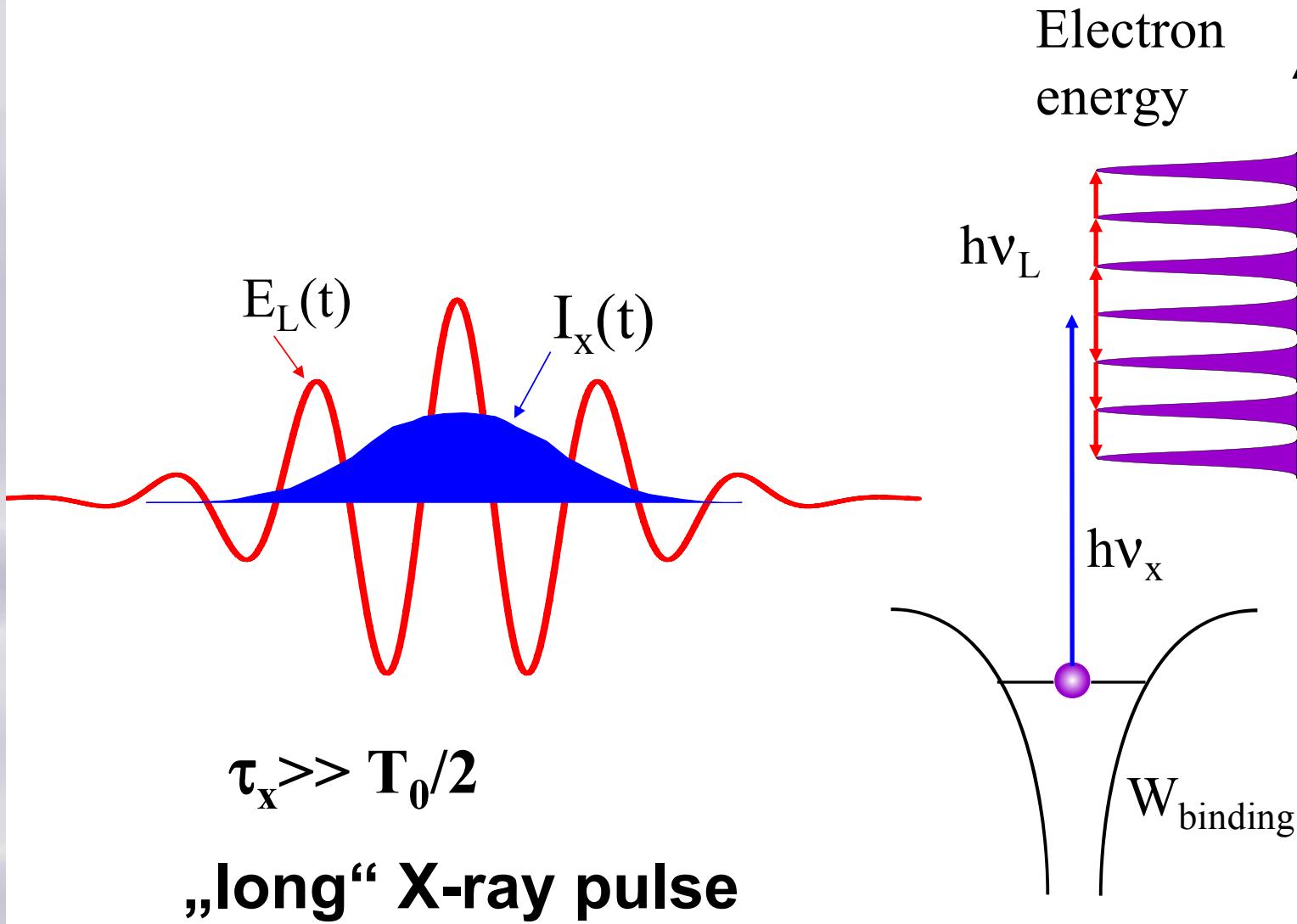
surface dynamics:

CEP measurement:

X-ray Pulse duration measurement at LCLS

In collaboration with:
DESY, CFEL, Ohio State U., Dublin U., SLAC

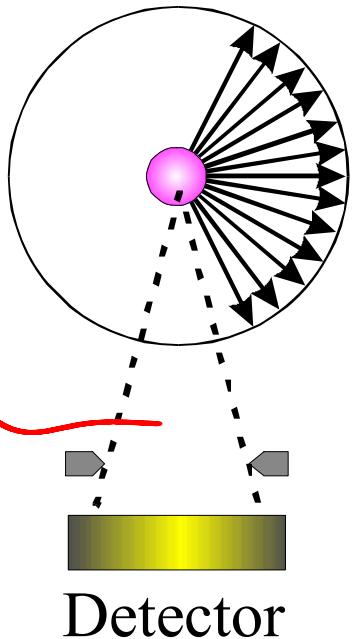
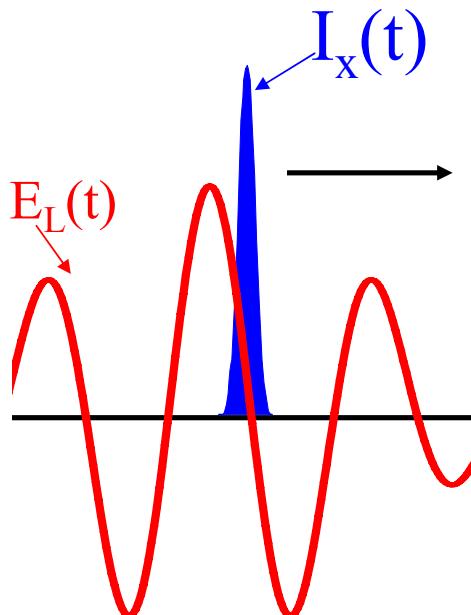
Laser-field dressed photoelectrons



Laser-field dressed photoelectrons

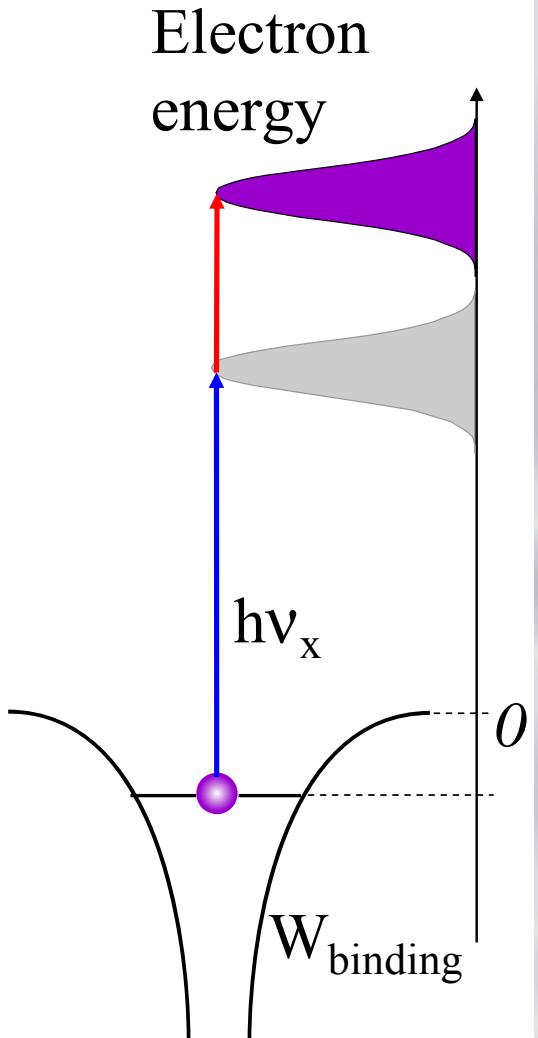
$$\lambda_{\text{IR}} = 2 \mu\text{m}$$

$$T_0/2 = 3.3 \text{ fs}$$

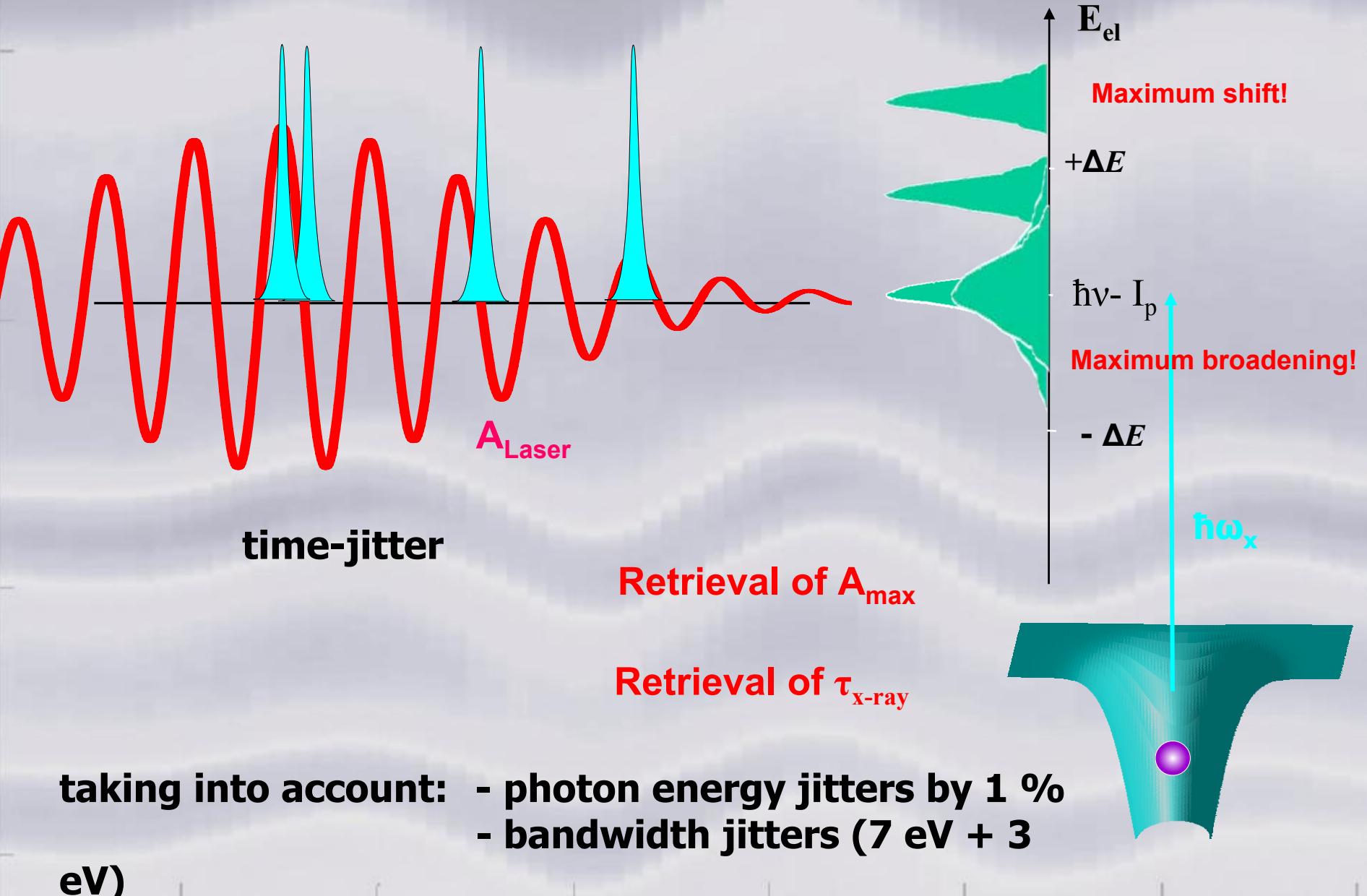


$$\tau_x < T_0/2$$

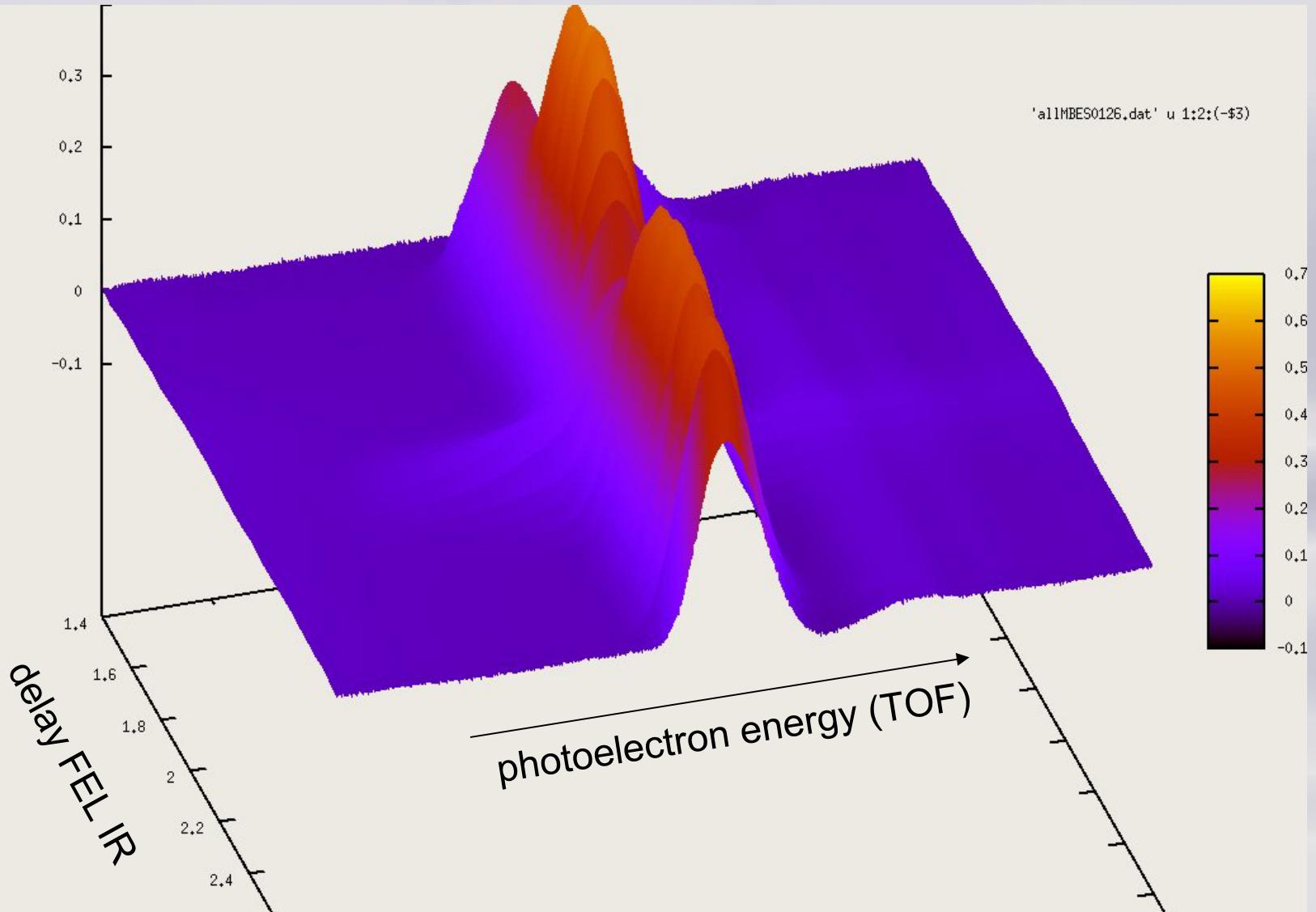
„short“ X-ray pulse



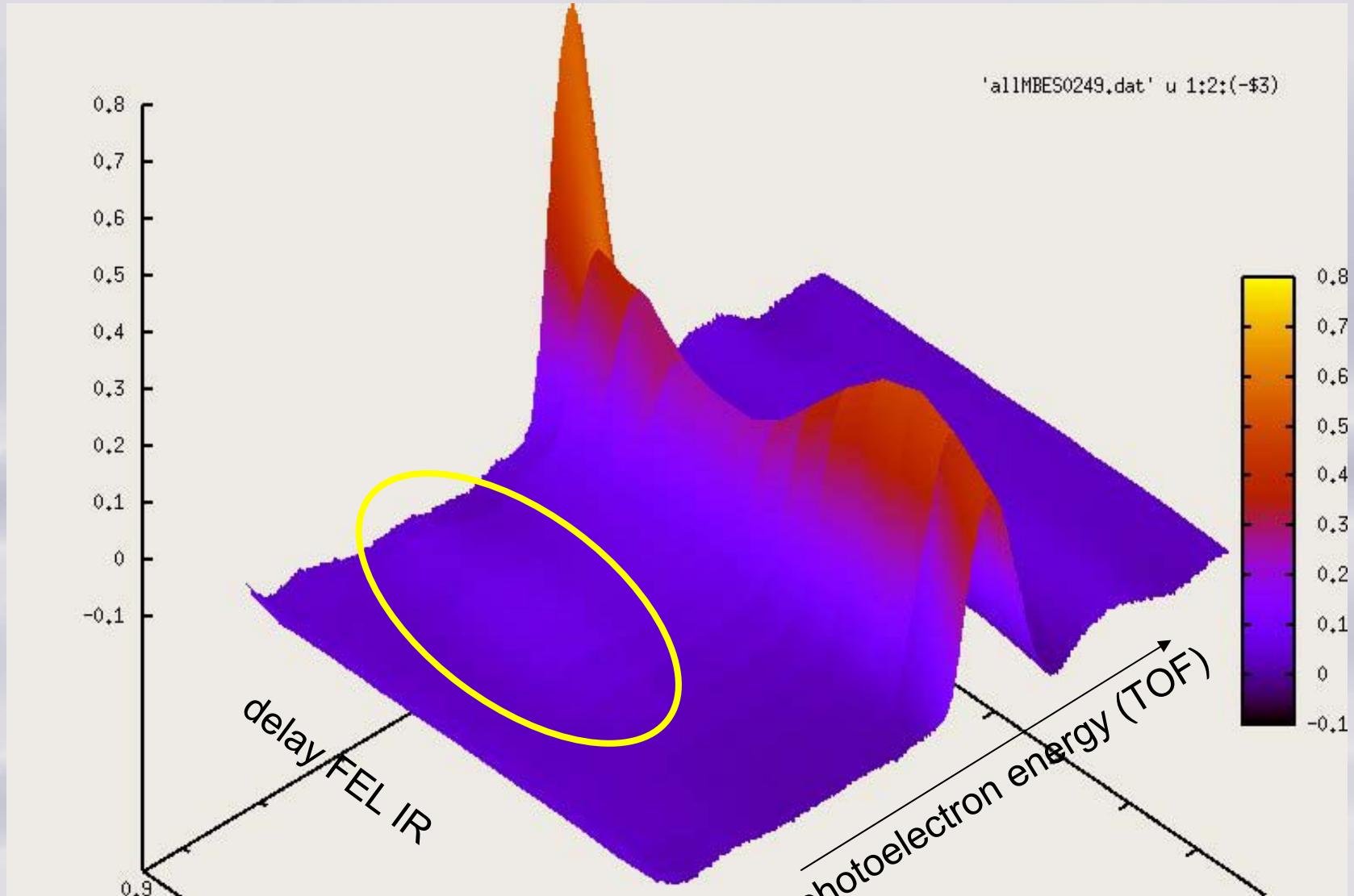
single-shot streaking measurement of $\tau_{\text{x-ray}}$



spectra sorted acc. to phase cavity timer „long“ pulse, $\lambda_{\text{Laser}} = 2 \mu\text{m}$



spectra sorted acc. to phase cavity timer „short“ pulse, $\lambda_{\text{Laser}} = 2 \mu\text{m}$



The End