Lecture Series Buenos Aires

18-3-2024 until 22-3-2024

Lecture M5 – Attosecond atomic physics - 1

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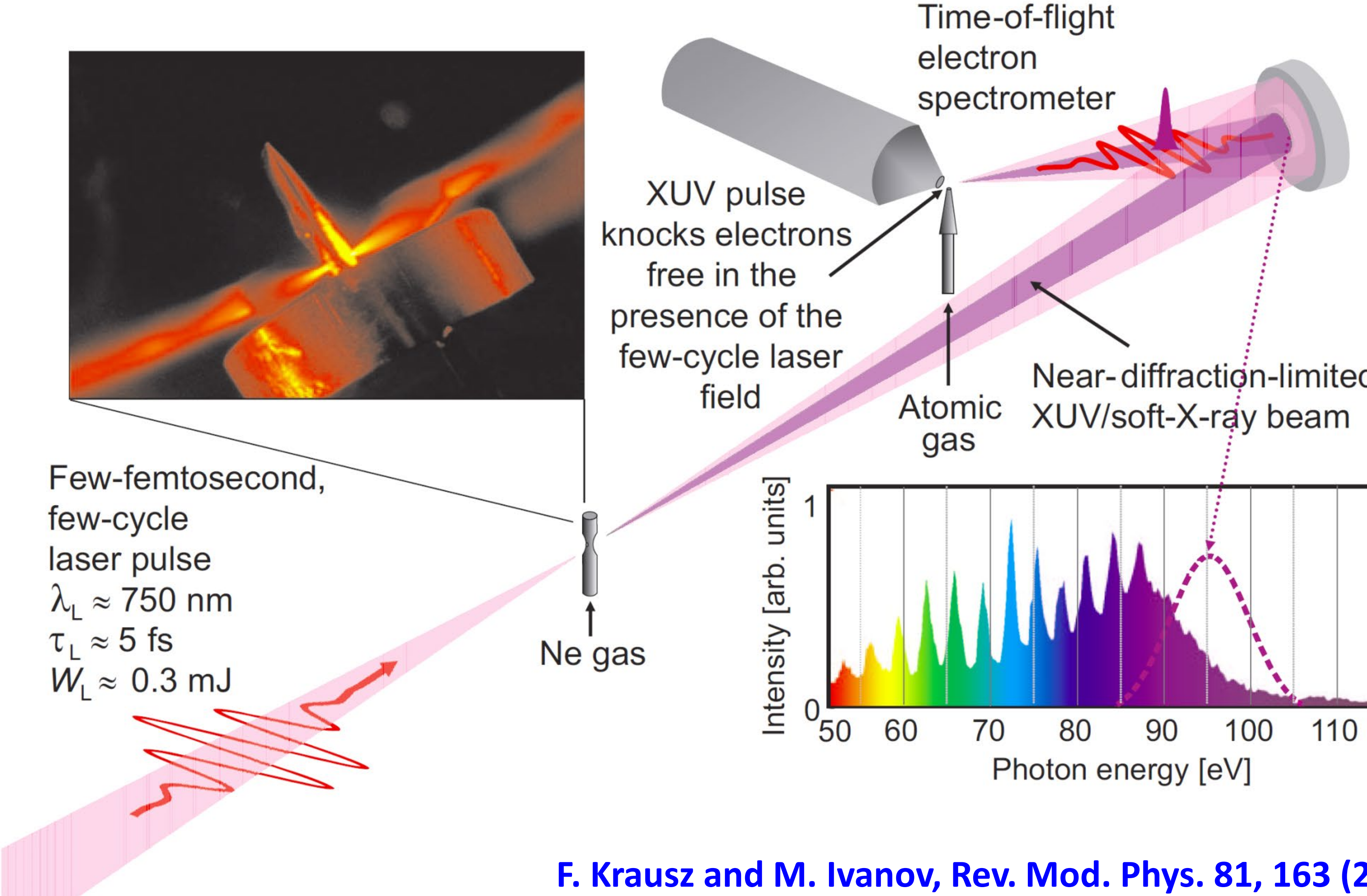
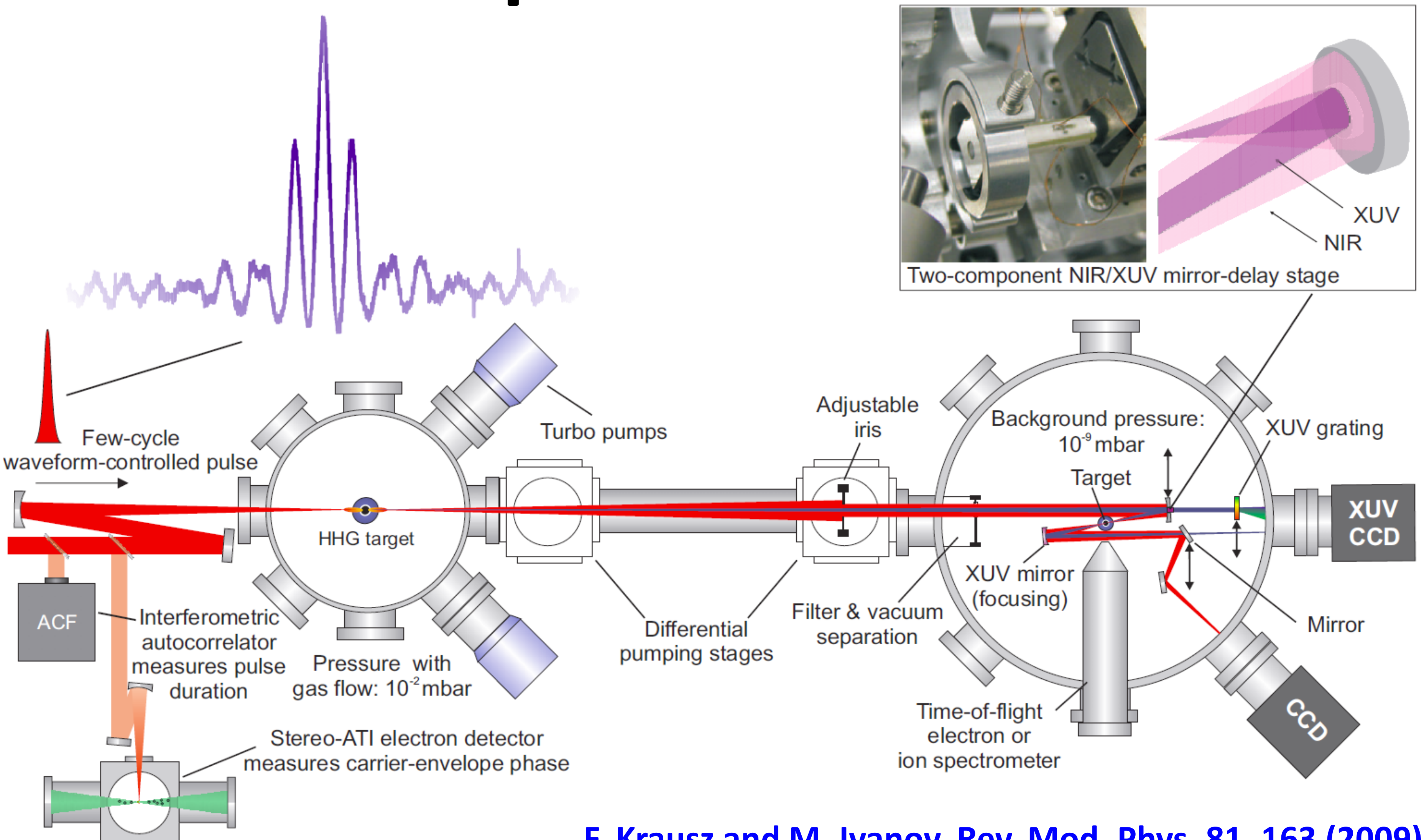
Configuring attosecond pump-probe experiments - 1



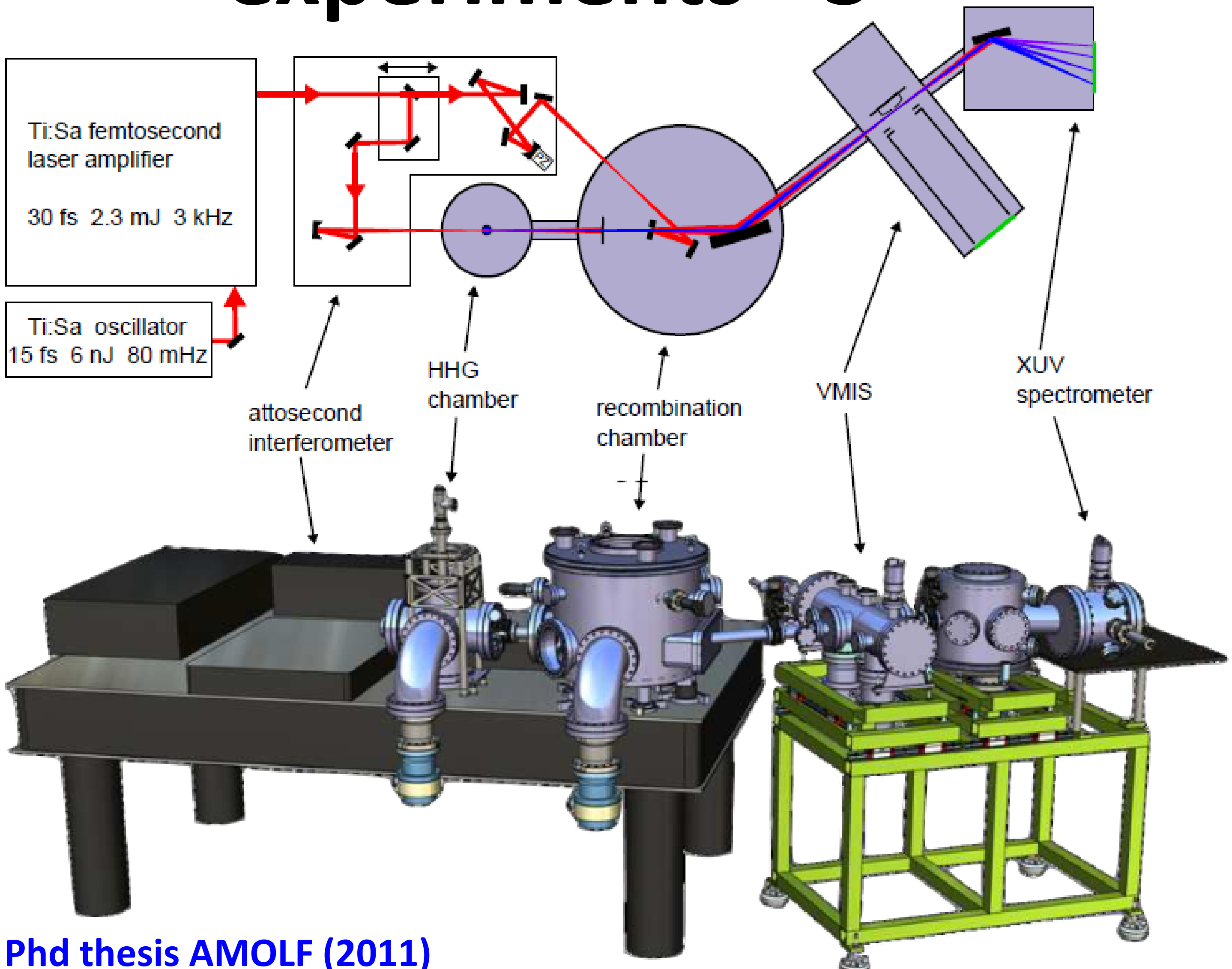
F. Krausz and M. Ivanov, Rev. Mod. Phys. 81, 163 (2009)

Configuring attosecond pump-probe experiments - 2



F. Krausz and M. Ivanov, Rev. Mod. Phys. 81, 163 (2009)

Configuring attosecond pump-probe experiments - 3

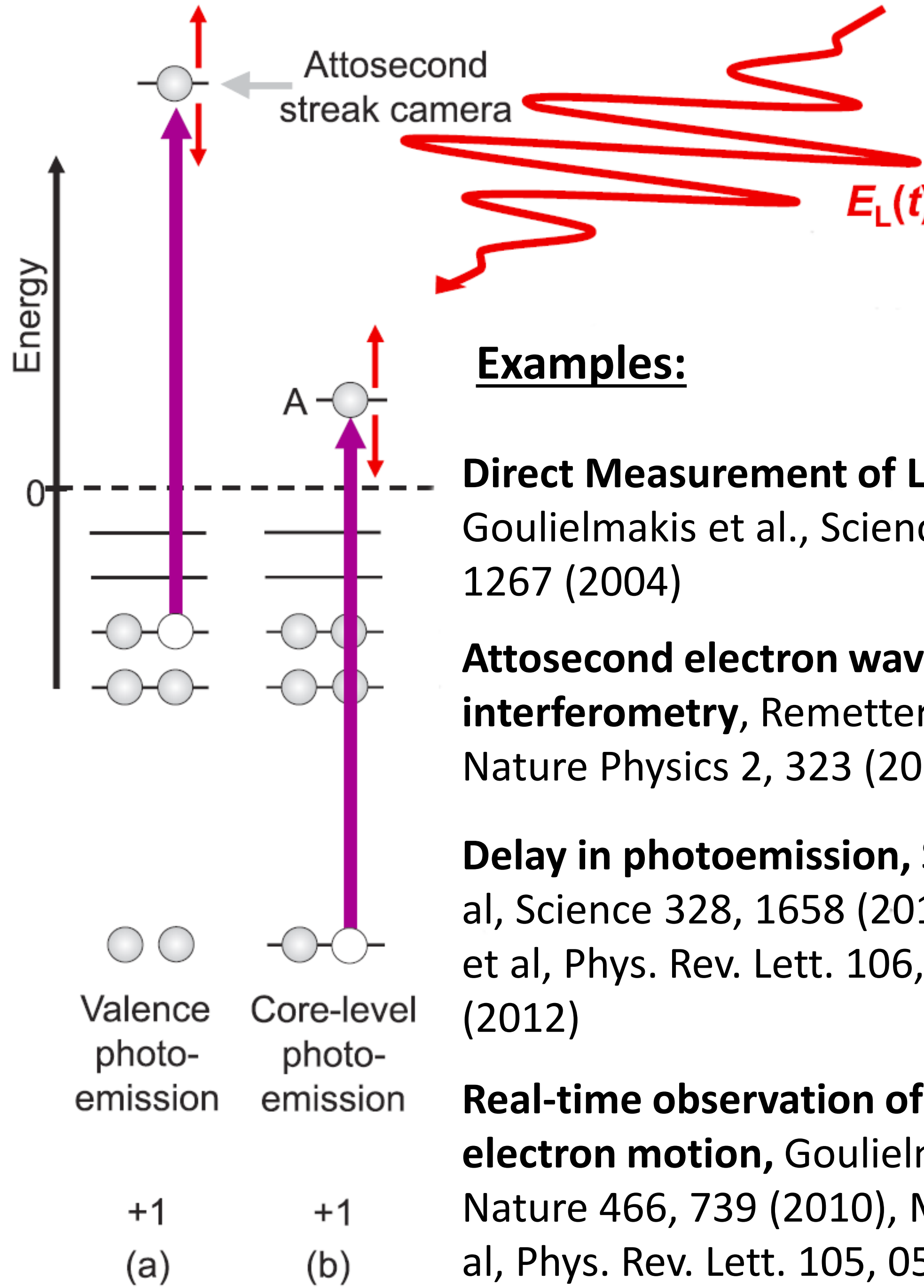


G. Gademann, Phd thesis AMOLF (2011)

Attosecond atomic physics

Single electron removal

- continuum electron dynamics following XUV photoionization (streaking)
- time delays between photoionization from different initial orbitals
- coherent electron (hole) motion following excitation of multiple orbitals or ionization from multiple orbitals



Examples:

Direct Measurement of Light Waves, Goulielmakis et al., Science 305, 1267 (2004)

Attosecond electron wave packet interferometry, Remetter et al., Nature Physics 2, 323 (2006)

Delay in photoemission, Schultze et al, Science 328, 1658 (2010), Klunder et al, Phys. Rev. Lett. 106, 143002 (2012)

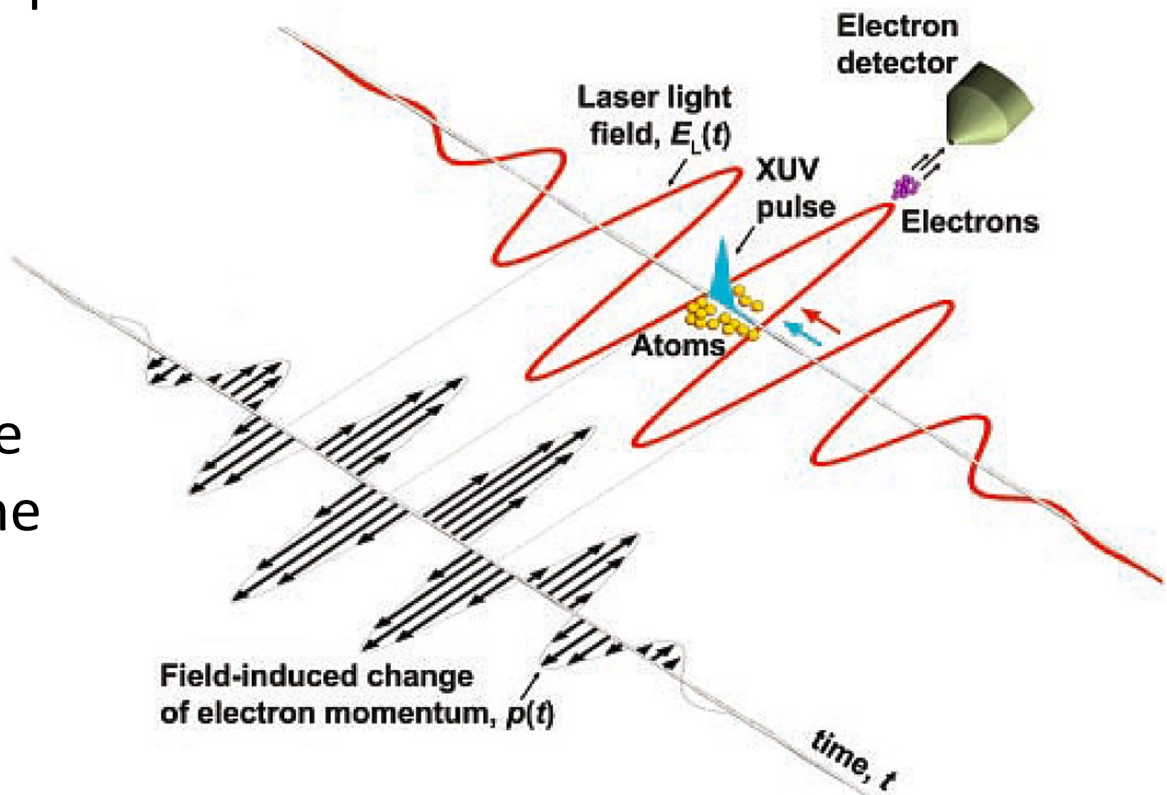
Real-time observation of valence electron motion, Goulielmakis et al., Nature 466, 739 (2010), Mauritsson et al, Phys. Rev. Lett. 105, 053001 (2010)

Direct Measurement of Light Waves

The outcome of a streaking measurement depends on

a. The properties of the XUV field
b. The properties of the NIR field
c. Atomic/molecular properties

If a. and c. are known, we can learn about b., i.e. the shape of the NIR field



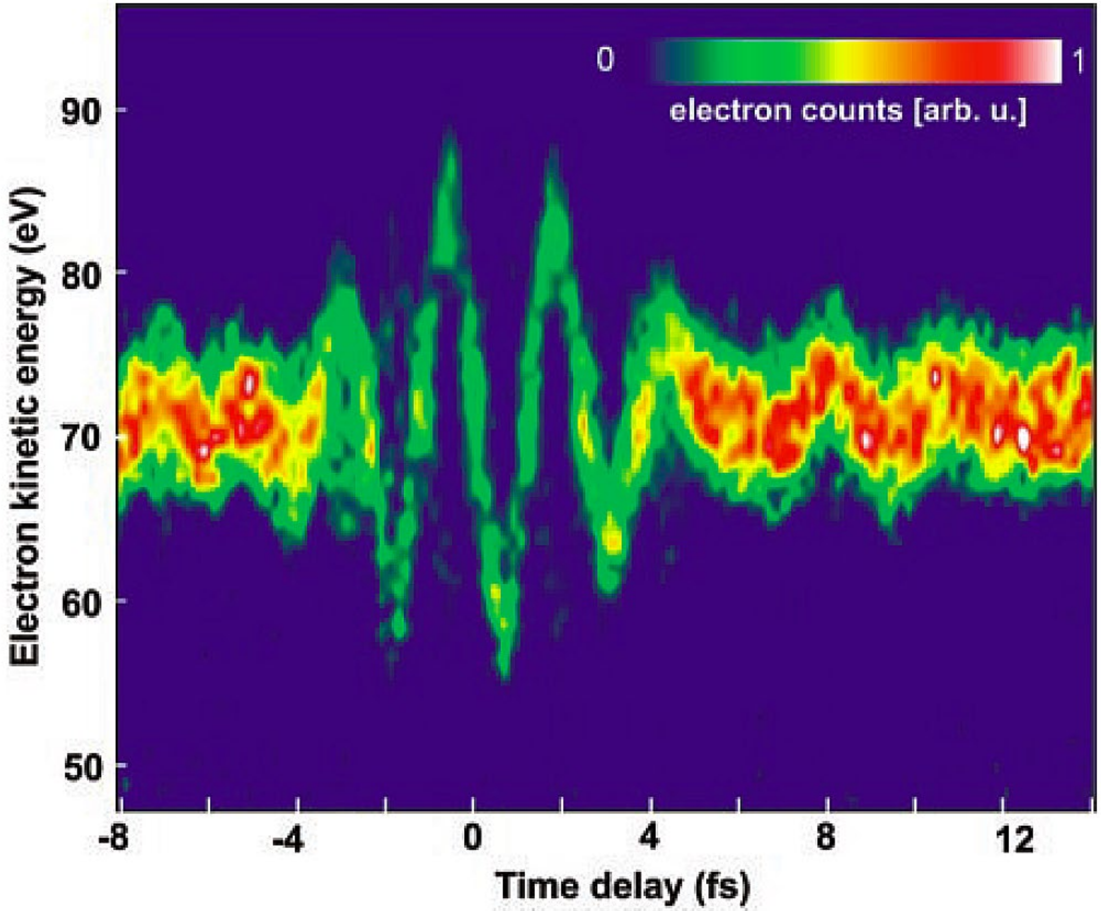
Goulielmakis et al., Science 305, 1267 (2004)

Direct Measurement of Light Waves

In an attosecond streaking measurement, the photoelectron acquires a momentum-shift that is proportional to the NIR vector potential at the time of ionization

$$\Delta p = -A(t_{ionization})$$

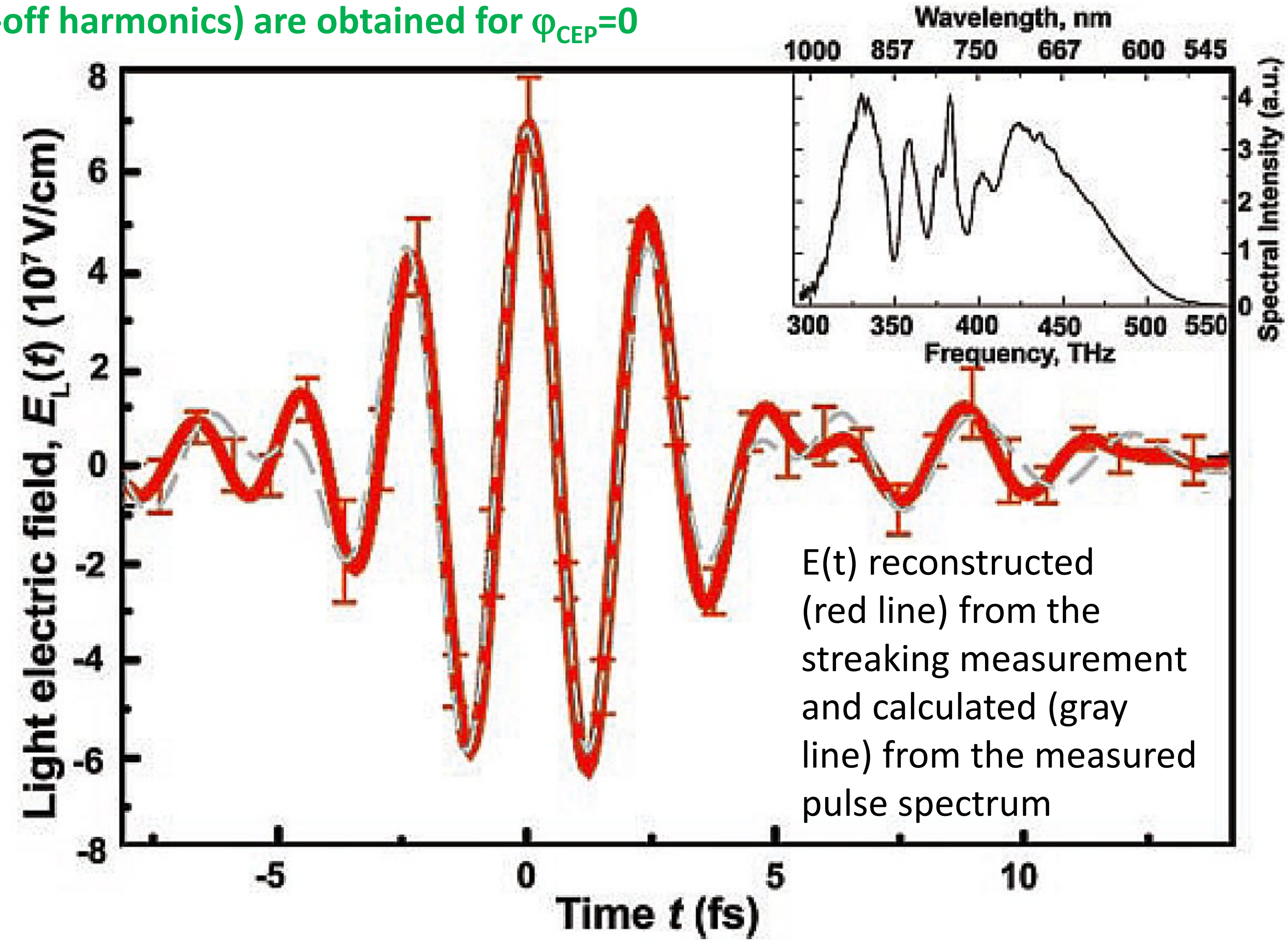
$$E(t) = -\frac{\partial A(t)}{\partial t}$$



Goulielmakis et al., Science 305, 1267 (2004)

Direct Measurement of Light Waves

Experimental proof that isolated attosecond pulses (cut-off harmonics) are obtained for $\varphi_{CEP}=0$



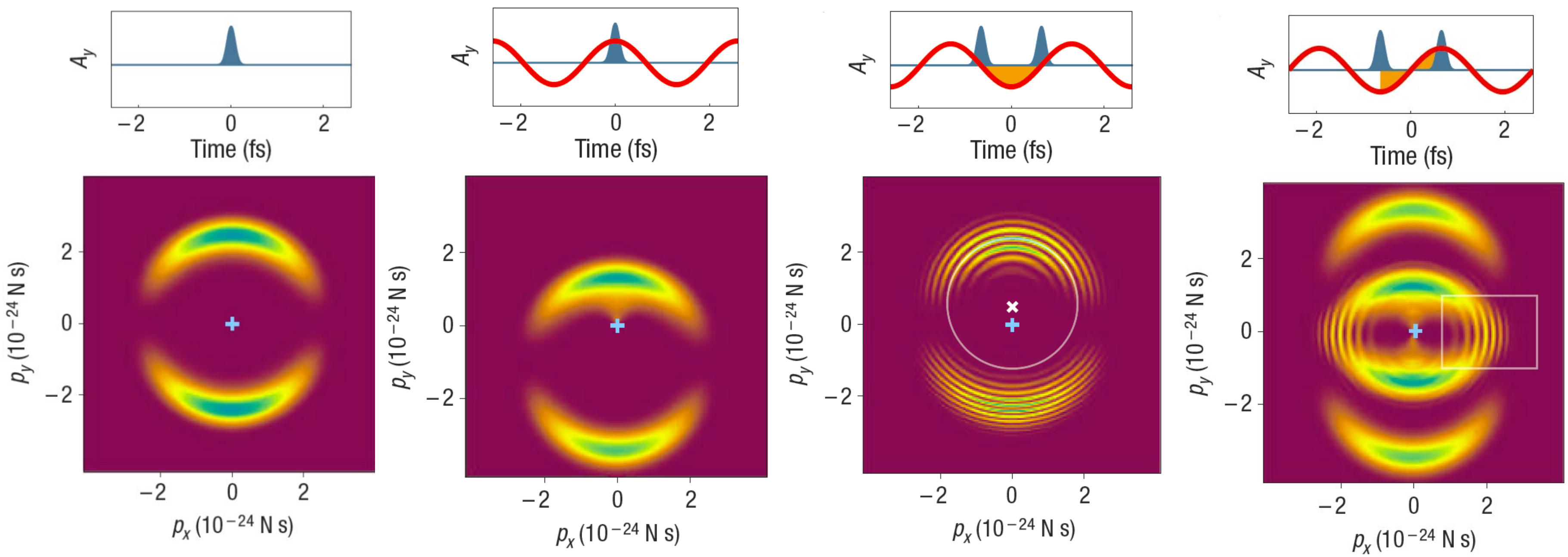
E(t) reconstructed (red line) from the streaking measurement and calculated (gray line) from the measured pulse spectrum

Goulielmakis et al., Science 305, 1267 (2004)

Attosecond electron wave packet interferometry

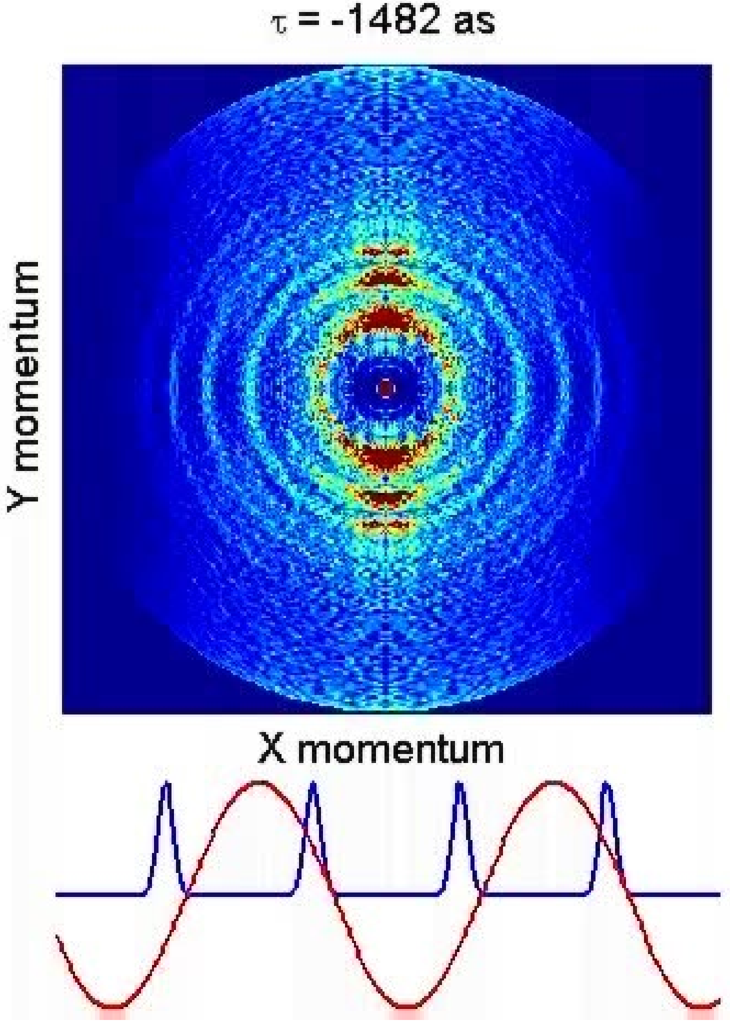
Extension of streaking spectroscopy to attosecond pulse trains
(or: RABBITT with non-perturbative fields)

Red line: A(t)



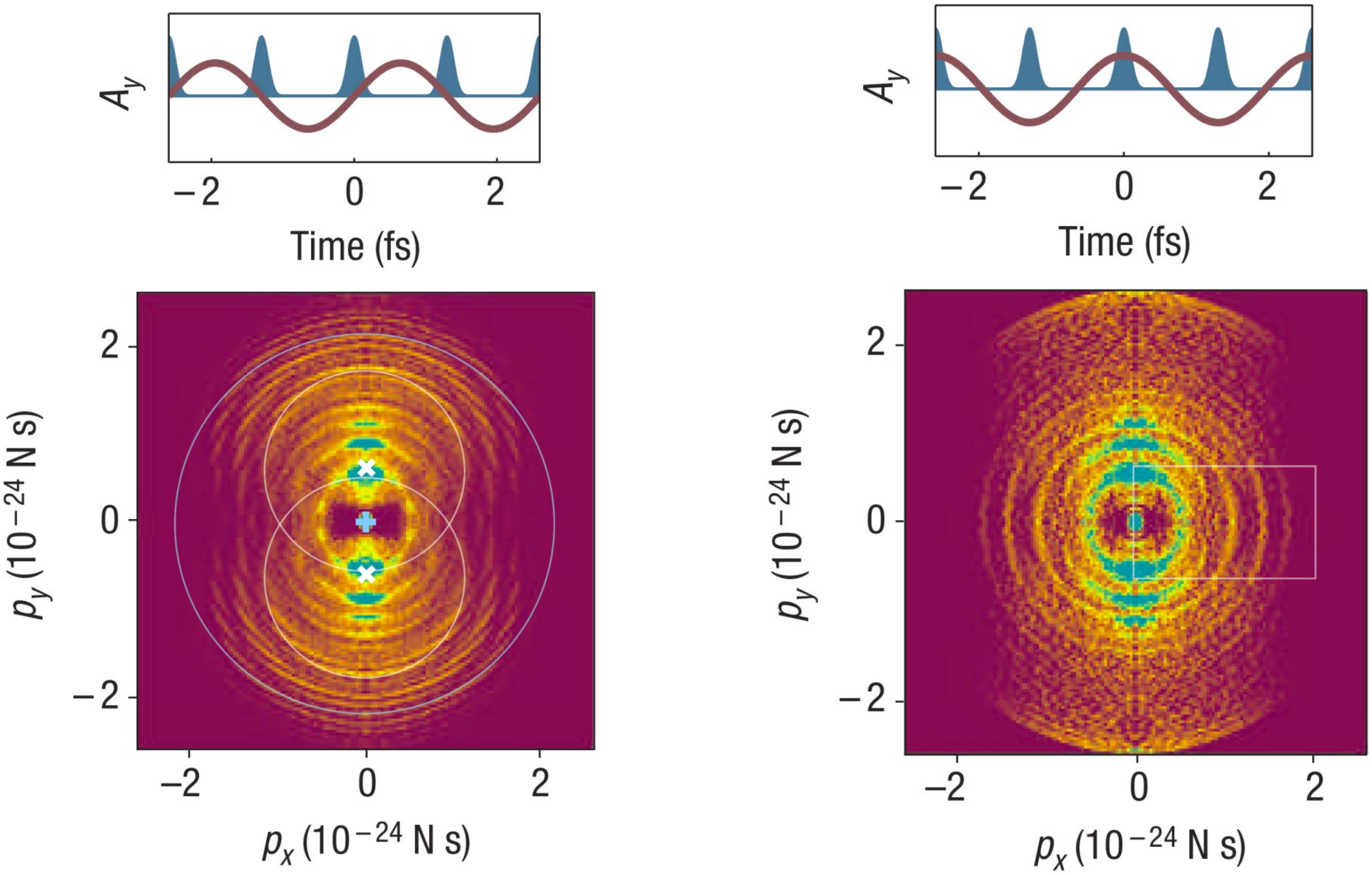
Remetter et al., Nature Physics 2, 323 (2006)

Attosecond electron wave packet interferometry



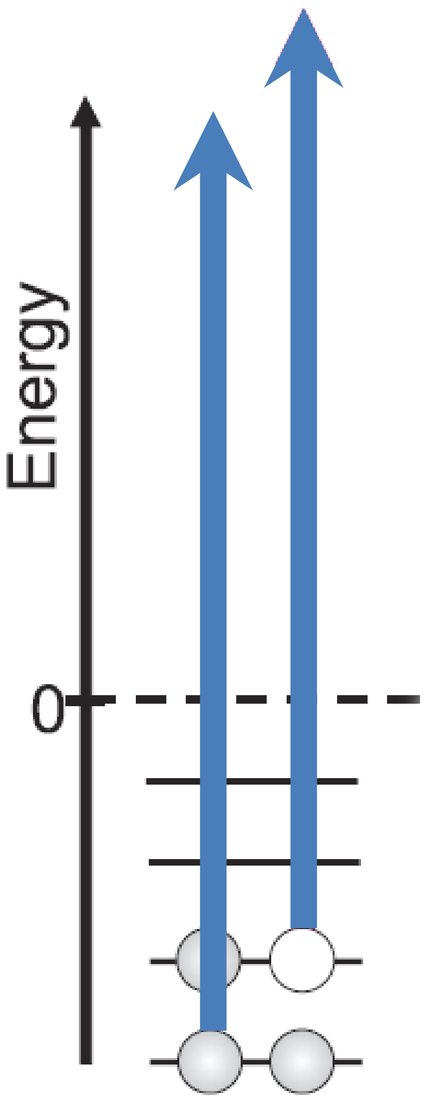
Remetter et al., Nature Physics 2, 323 (2006)

Attosecond electron wave packet interferometry



Remetter et al., Nature Physics 2, 323 (2006)

Delay in photoemission



Questions: do two electrons that originate from different orbitals ionize at the same time or is there a delay between the two?

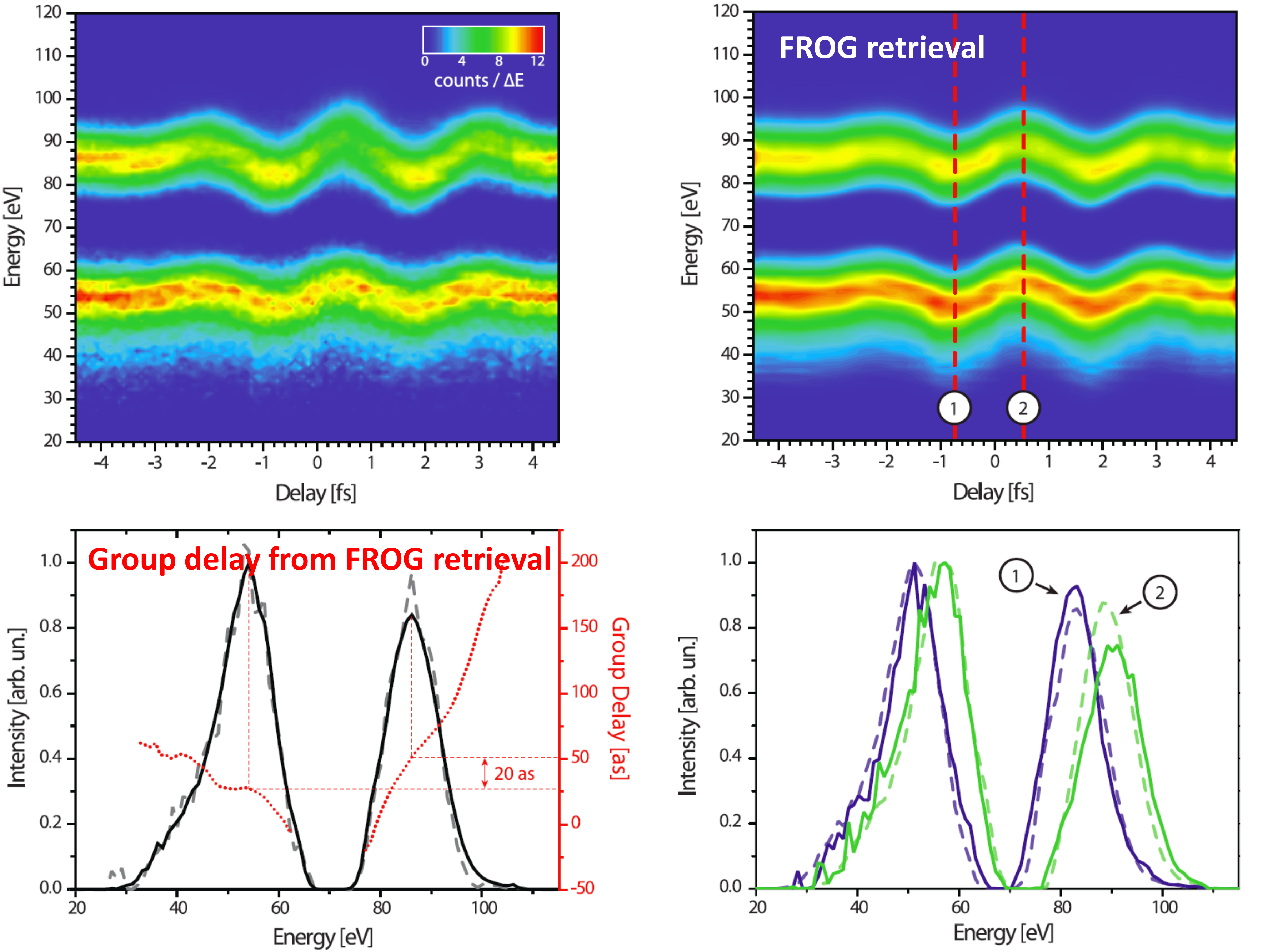
Two experimental approaches:

- Ionization by an isolated attosecond pulse (IAP) in combination with a streaking measurement

Schultze et al, Science 328, 1658 (2010)

- Ionization by a train of attosecond pulse (APT) in combination with a RABBITT measurement

Kluender et al, Phys. Rev. Lett. 106, 143002 (2012)

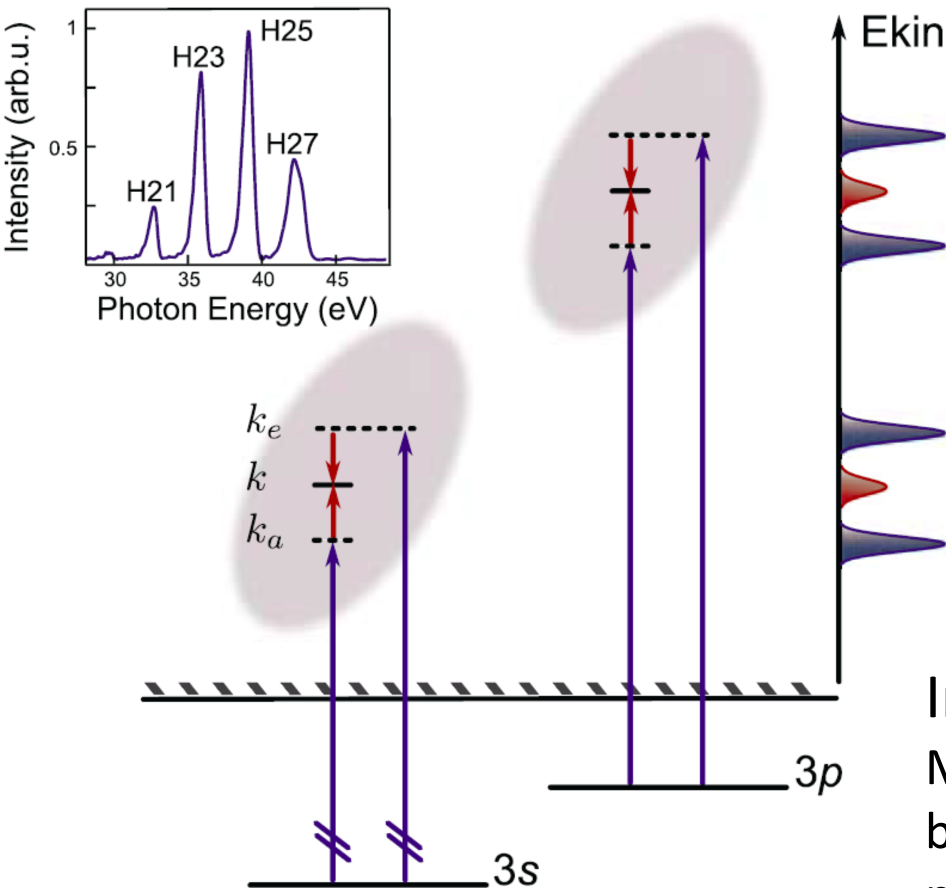


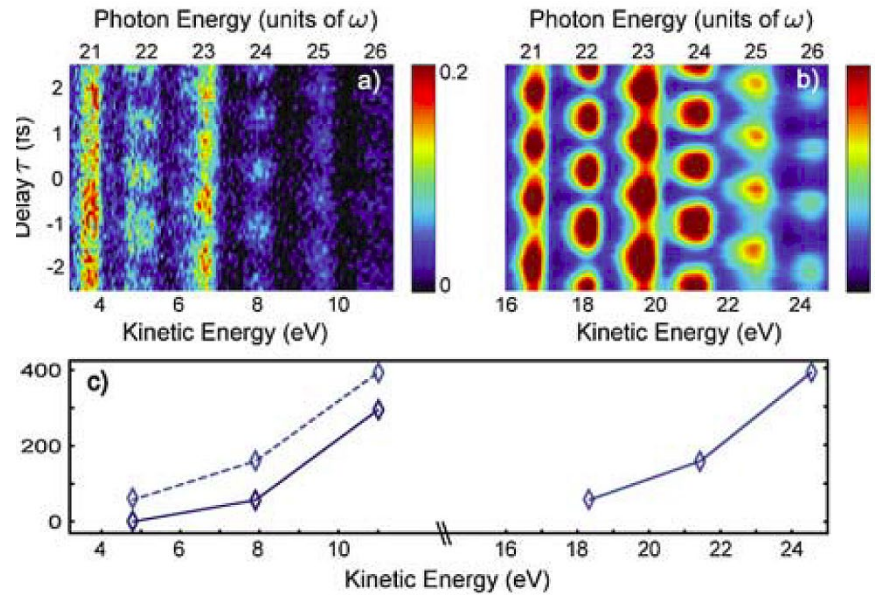
Schultze et al, Science 328, 1658 (2010)

Delay in photoemission

$$S(\tau) = \alpha + \beta \cos[2\omega(\tau - \tau_A - \tau_I)],$$

τ_A = group delay of the attosecond pulses
τ_I = atomic delay two-color ionization

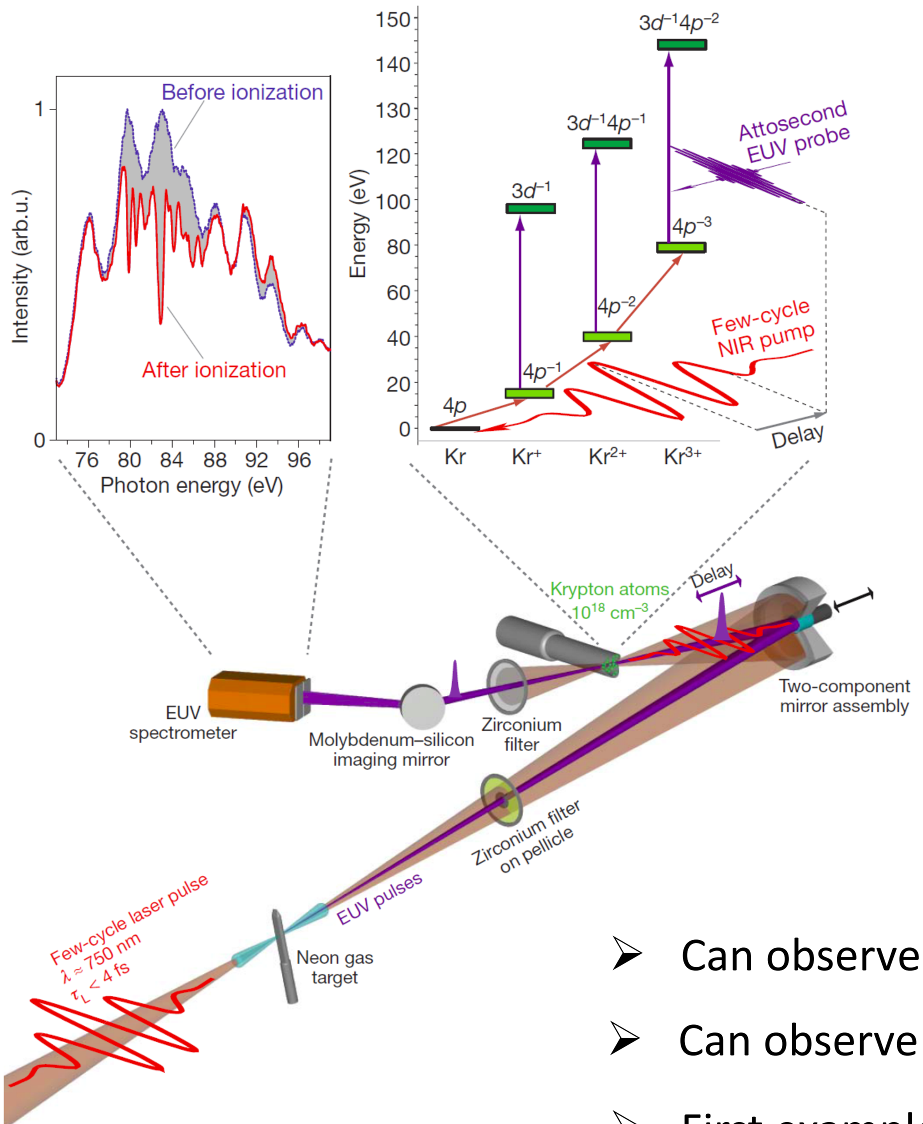




Important:
Measured time delays /phases accumulate both during ionization process and during propagation in the Coulomb+laser fields

Kluender et al, Phys. Rev. Lett. 106, 143002 (2012)

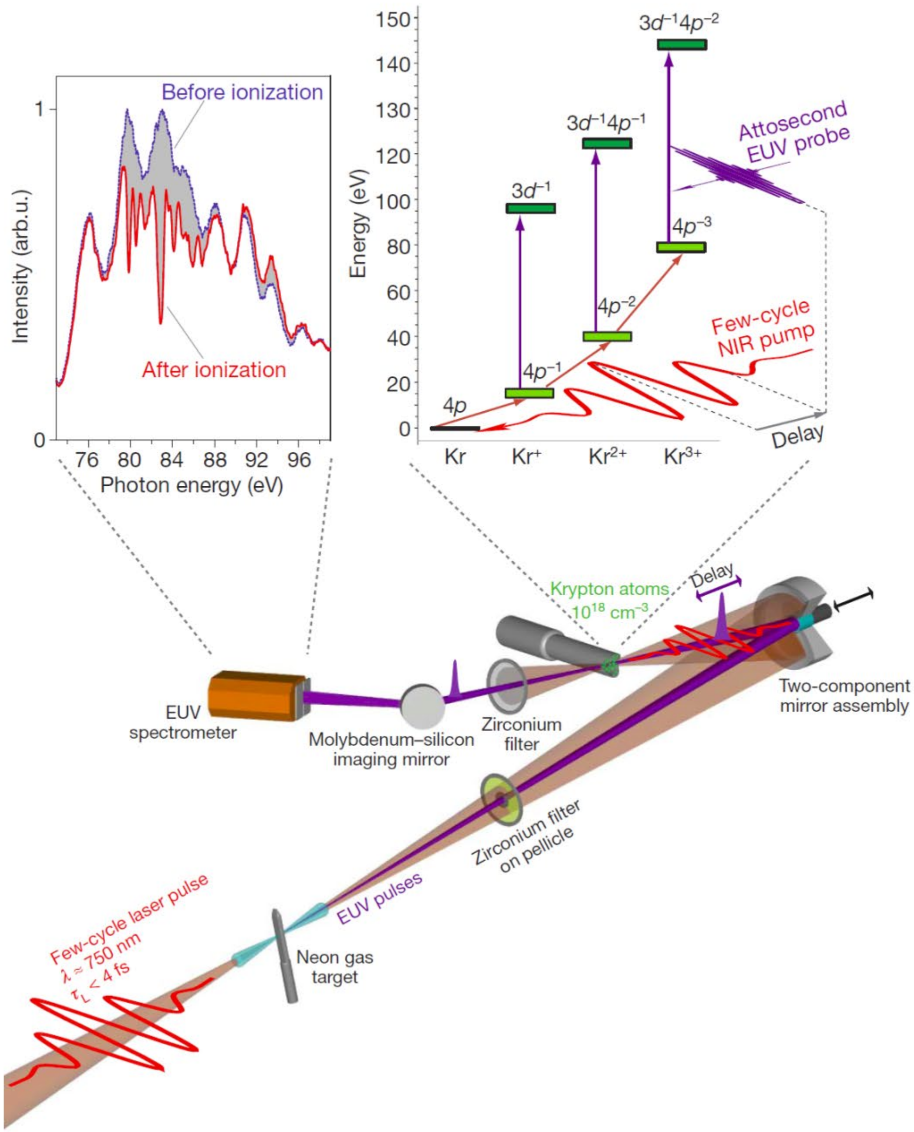
Real-time observation of valence electron motion

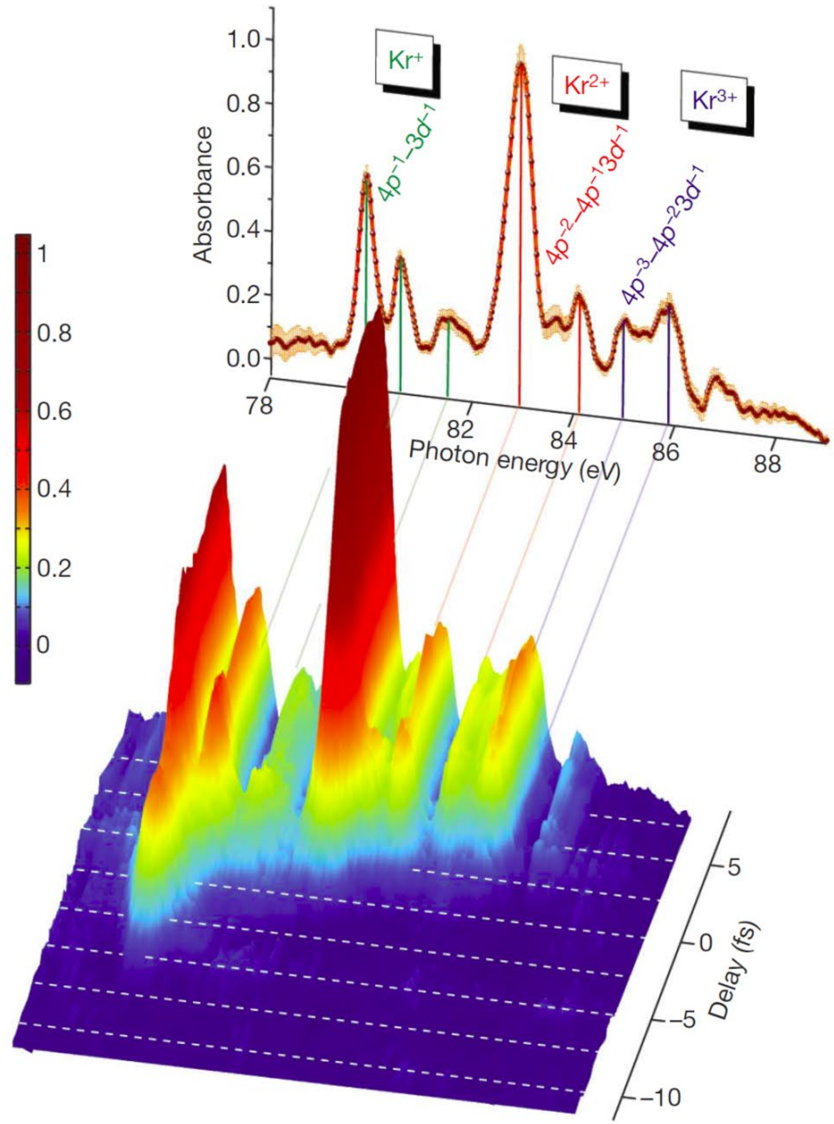


Ionization produces the ion in a superposition of two states that are probed by the XUV

- Can observe stepsize formation of different ionic states
- Can observe coherence between different ionic states
- First example of Attosecond Transient Absorption (ATAS)

Goulielmakis et al., Nature 466, 739 (2010)





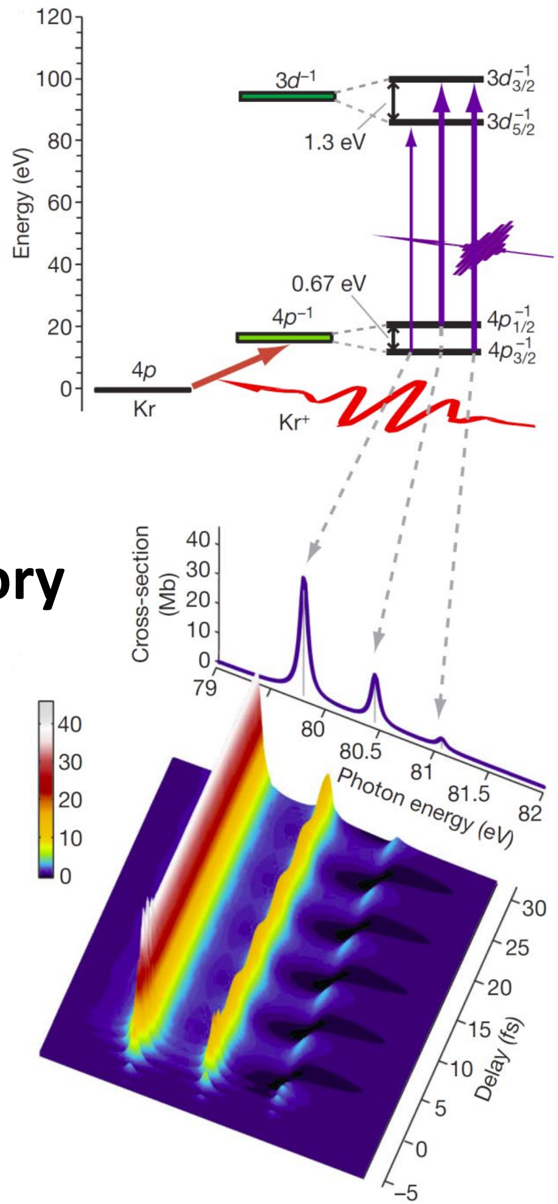
Goulielmakis et al., Nature 466, 739 (2010)

- ➢ stepsize formation of different ionic states

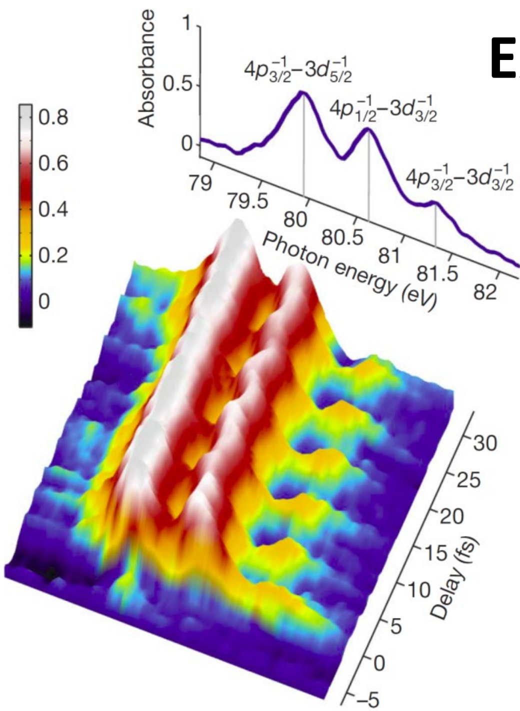
Observation of electronic coherence

After ionization the Kr^{+} ion is in a $4p_{1/2}$ or $4p_{3/2}$ state
Both configuration can be excited to a $3d_{3/2}$ state of the ion → interference

Theory

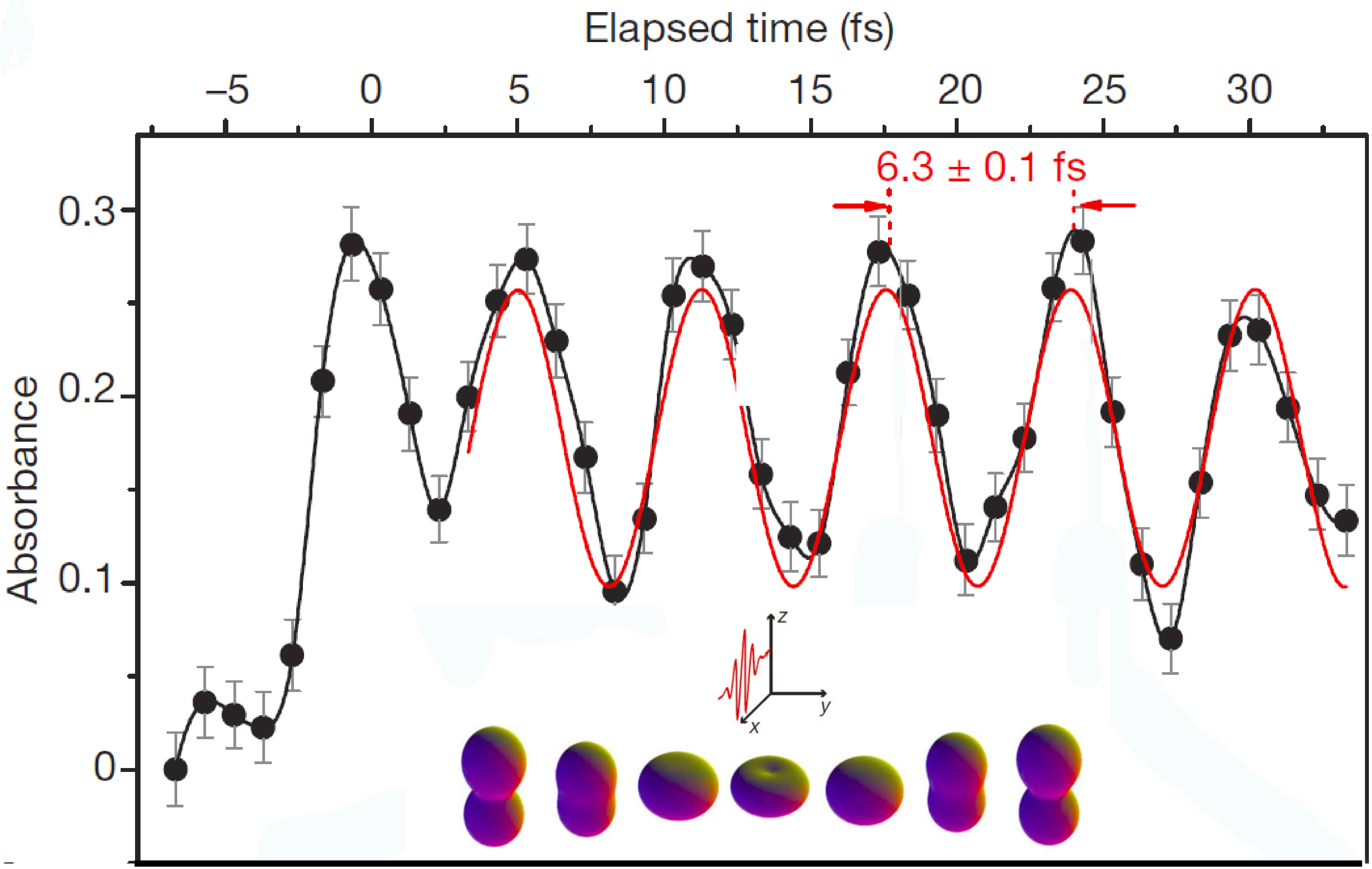


Experiment



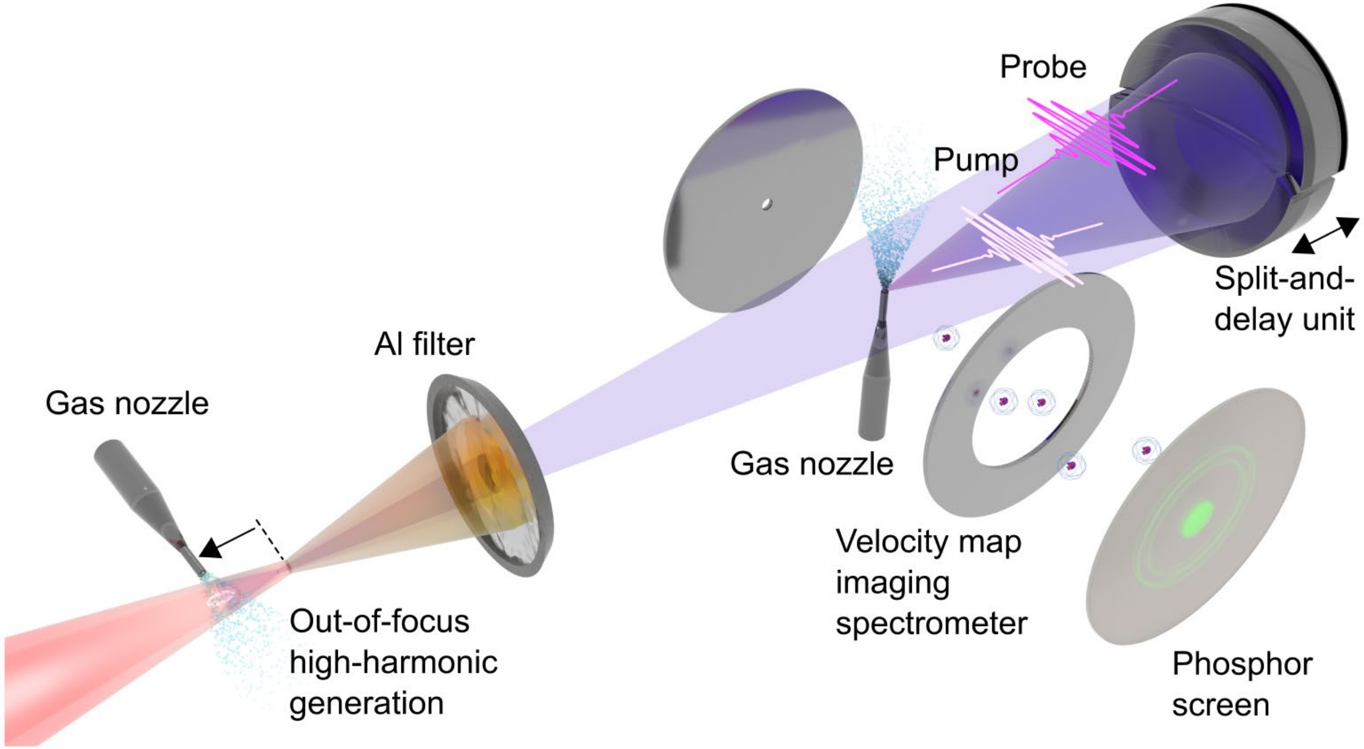
Goulielmakis et al., Nature 466, 739 (2010)

Observation of electronic coherence



Goulielmakis et al., Nature 466, 739 (2010)

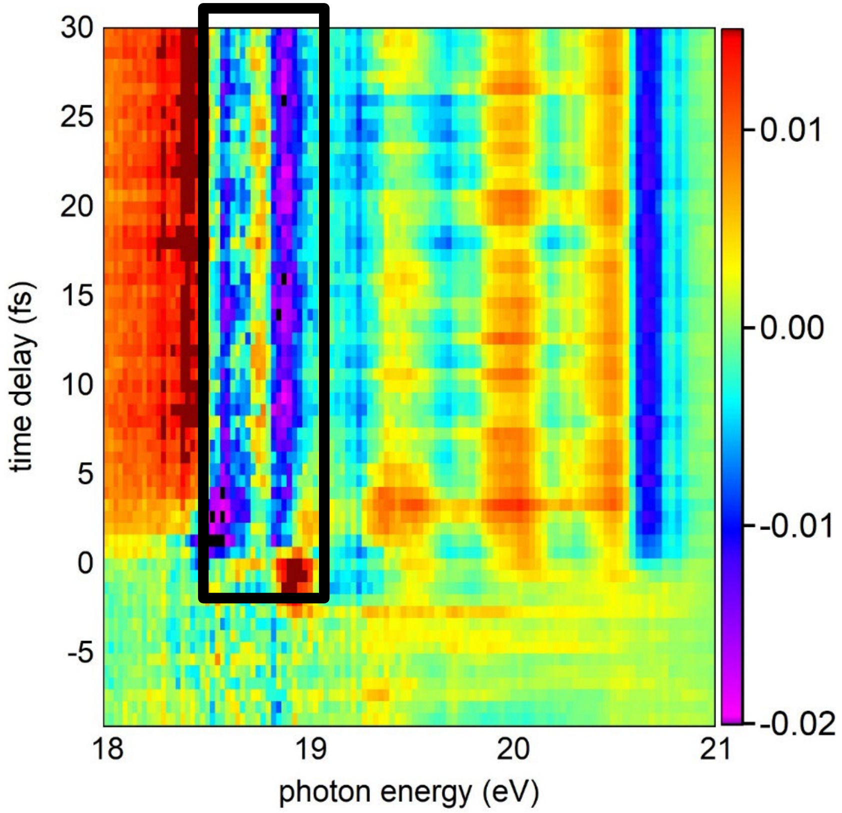
All-attosecond pump-probe spectroscopy

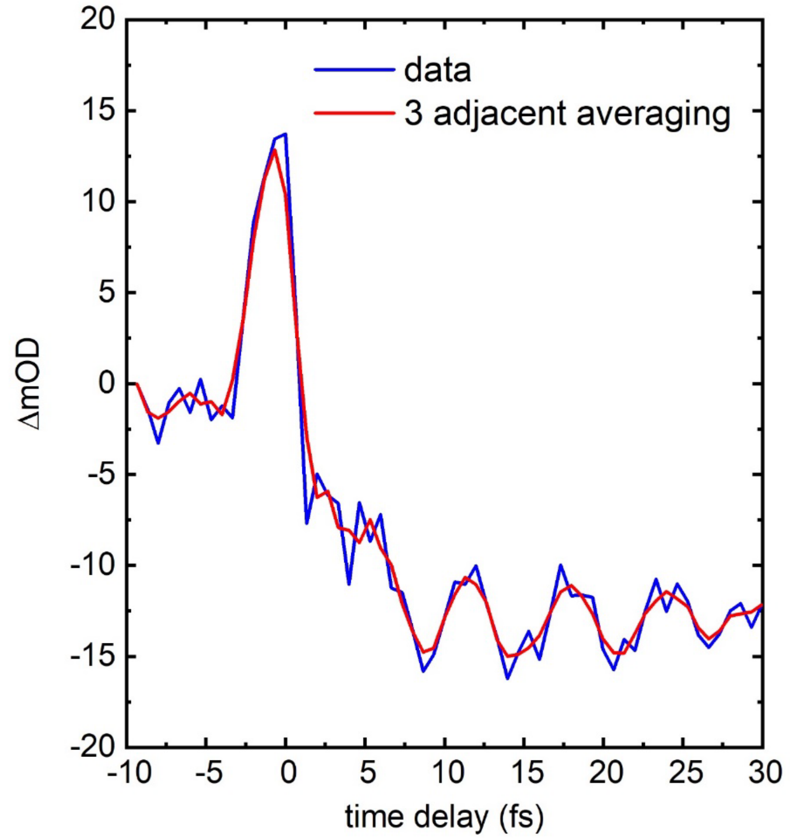


- Post-compressed kHz Ti:Sa laser (3.8 fs, 1 mJ)
- Tight focusing to ~$6\text{x}10^{15}$ W/cm^2
- *Out of focus* HHG in a high density gas jet producing divergent XUV with a few-μm virtual source size
- 5-10 fold demagnification to waist radius ~1μm

Optica 8, 960 (2021)
Science Advances 10, eadk9605 (2024)

First-ever all-attosecond transient absorption (MBI, 2/2024)

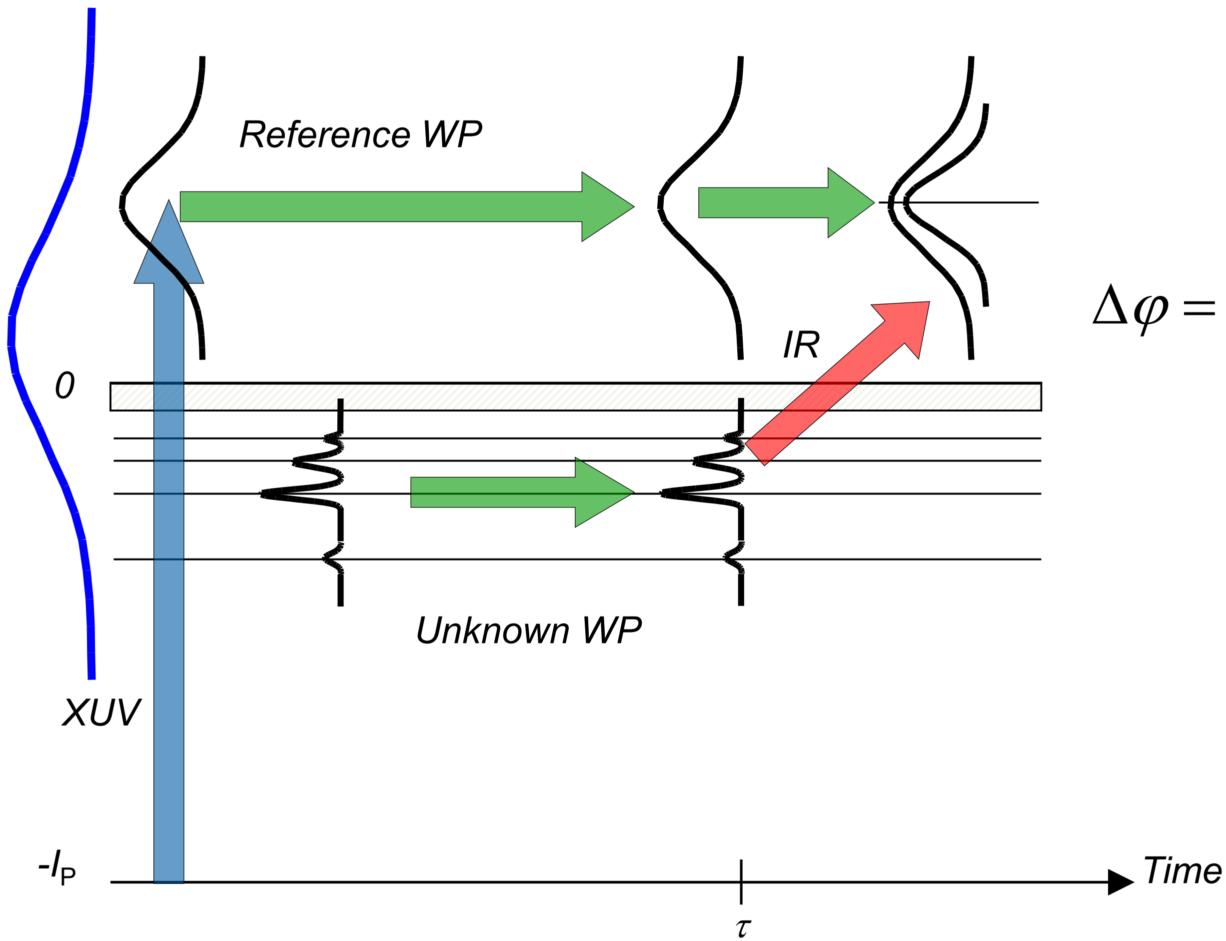




- Ionization of Kr with a (near-)isolated *pump* attosecond pulse
- Probing of electronic coherence in Kr^+ by measuring the absorption of a (near)-isolated *probe* attosecond pulse

- Observation of spin-orbit wavepacket in Kr^+ (coherent superposition of $^2P_{1/2}$ and $^2P_{1/2}$ spin-orbit states

Holographic observation of electronic coherence

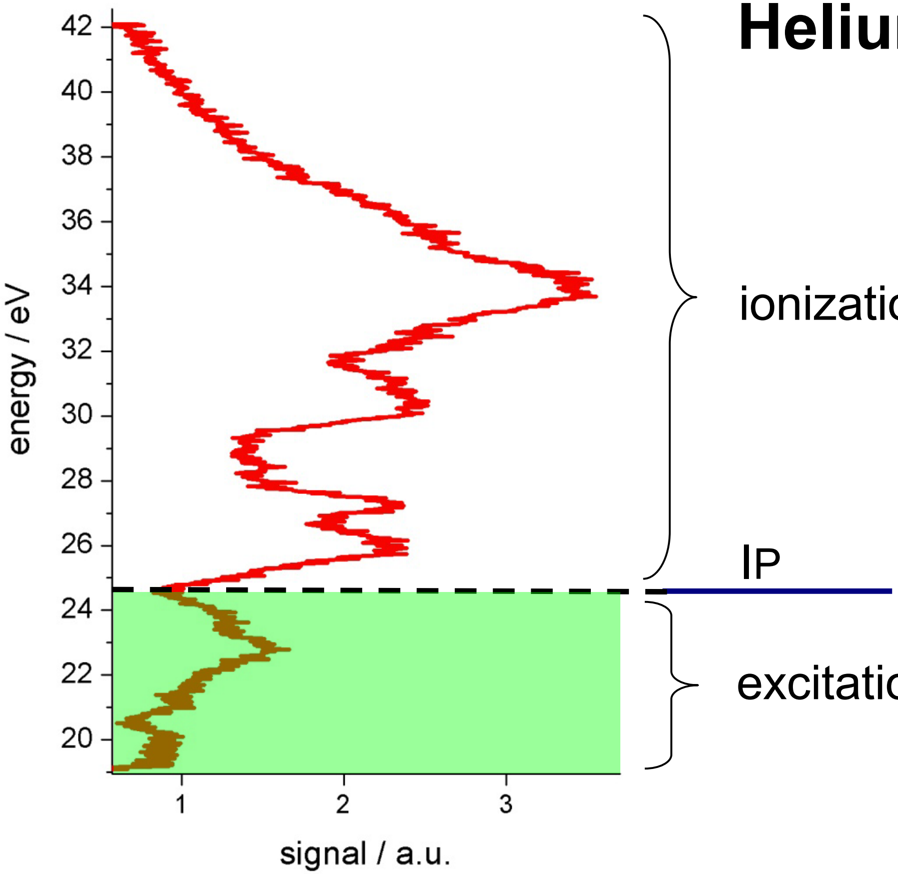


$$\Delta\varphi = (E_{ref} - E_A)\tau \;/\hbar$$

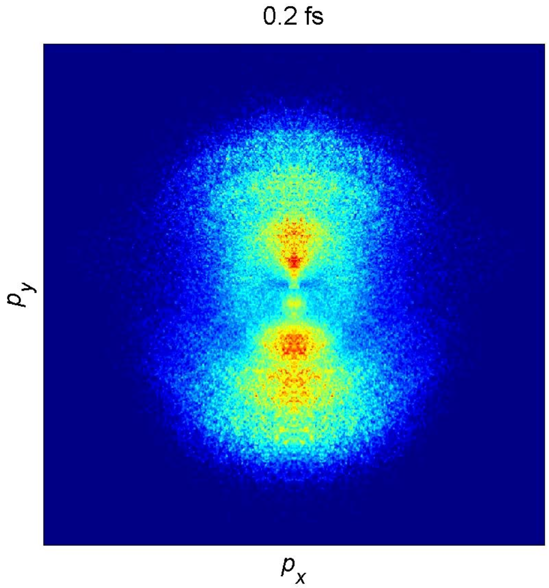
Mauritsson et al, Phys. Rev. Lett. 105, 053001 (2010)

Near-threshold Electron Wavepackets

HHG in Xenon, polarization gated
100 nm Al filter



VMIS image



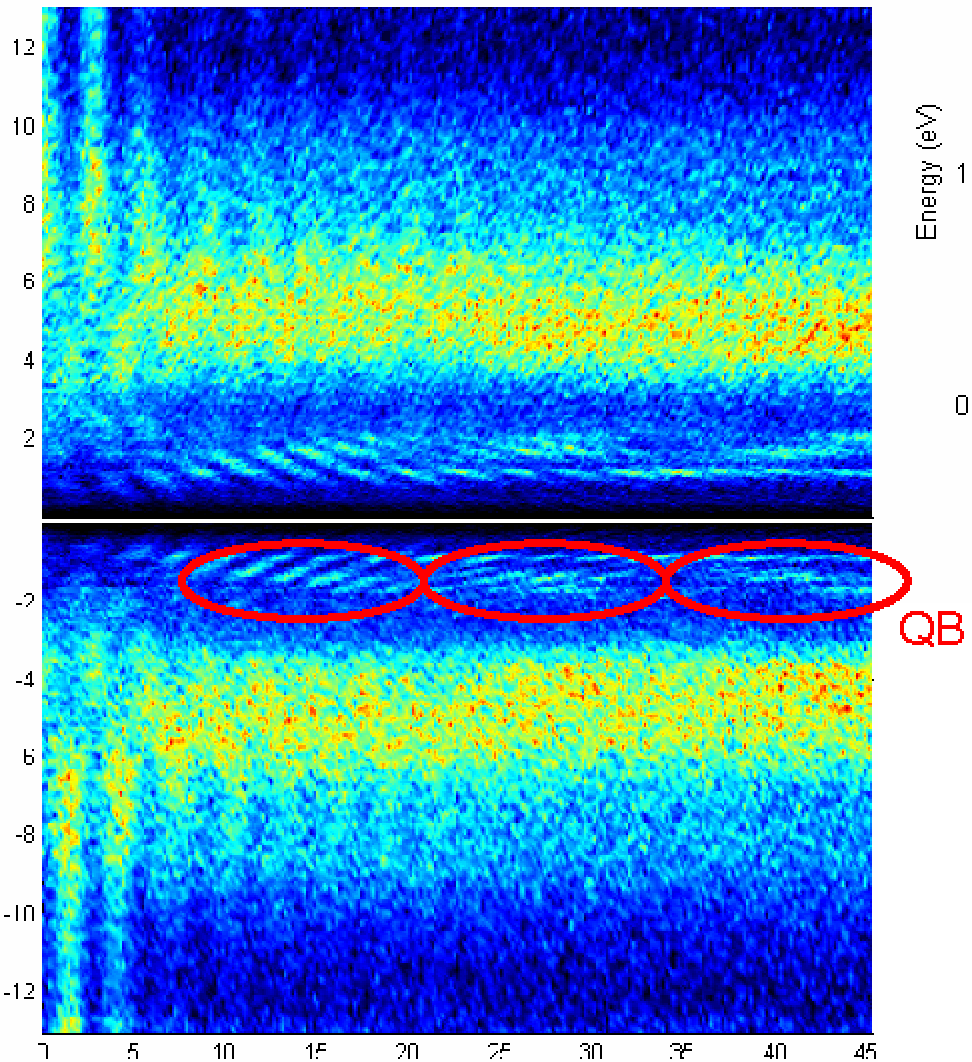
Helium ionization (raw image)

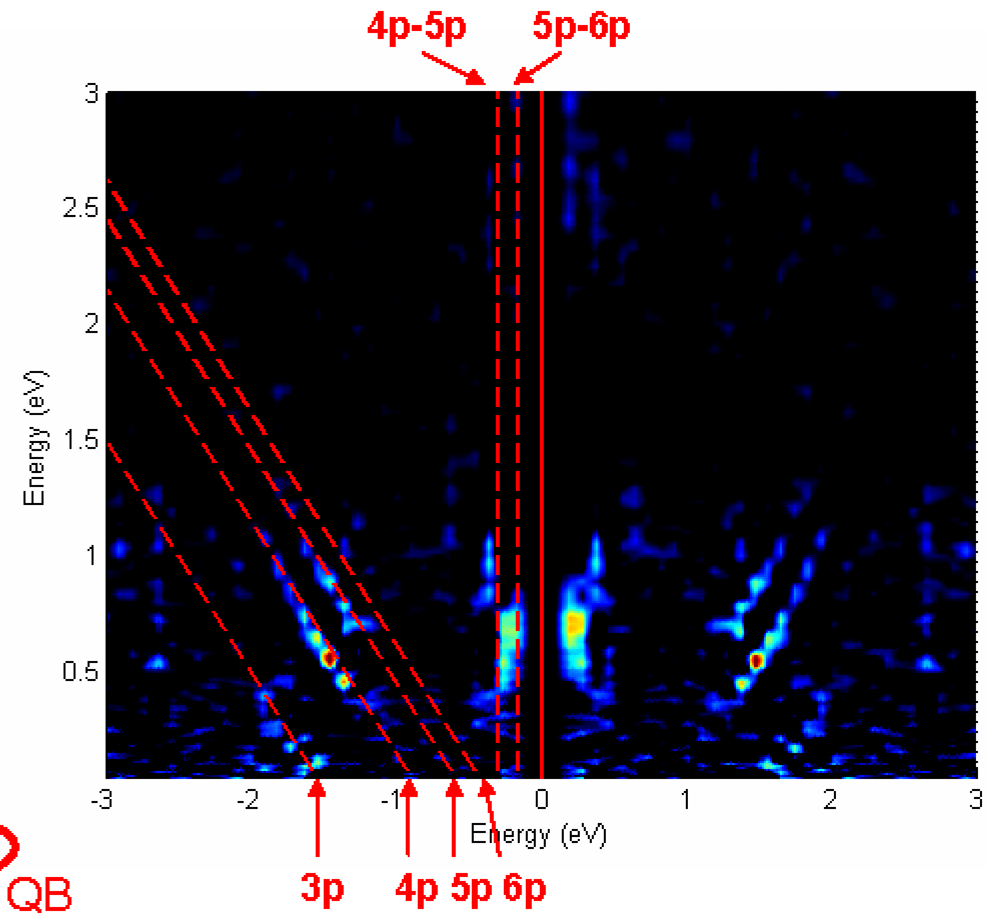
Mauritsson et al, Phys. Rev. Lett. 105, 053001 (2010)

Near-threshold Electron Wavepackets

Mauritsson et al, Phys. Rev. Lett. 105, 053001 (2010)

First-ever observation of bound electron dynamics using attosecond lasers!!!





In (E,E) plot the beats of individual states against the continuum and beats among the states can be observed → access to energy, amplitude and phase!!!

Mauritsson et al, Phys. Rev. Lett. 105, 053001 (2010)

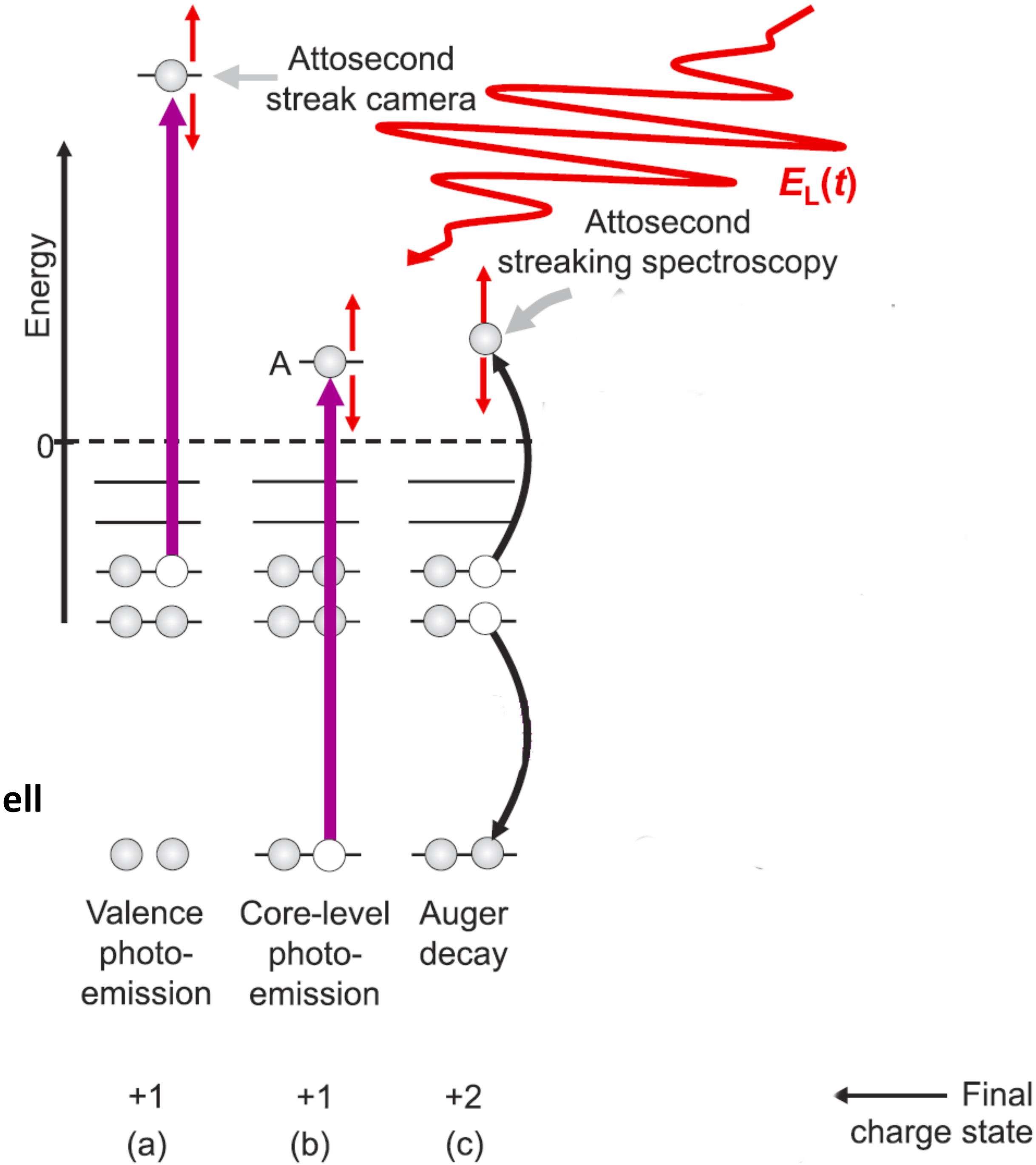
Attosecond atomic physics

Multi-electron dynamics

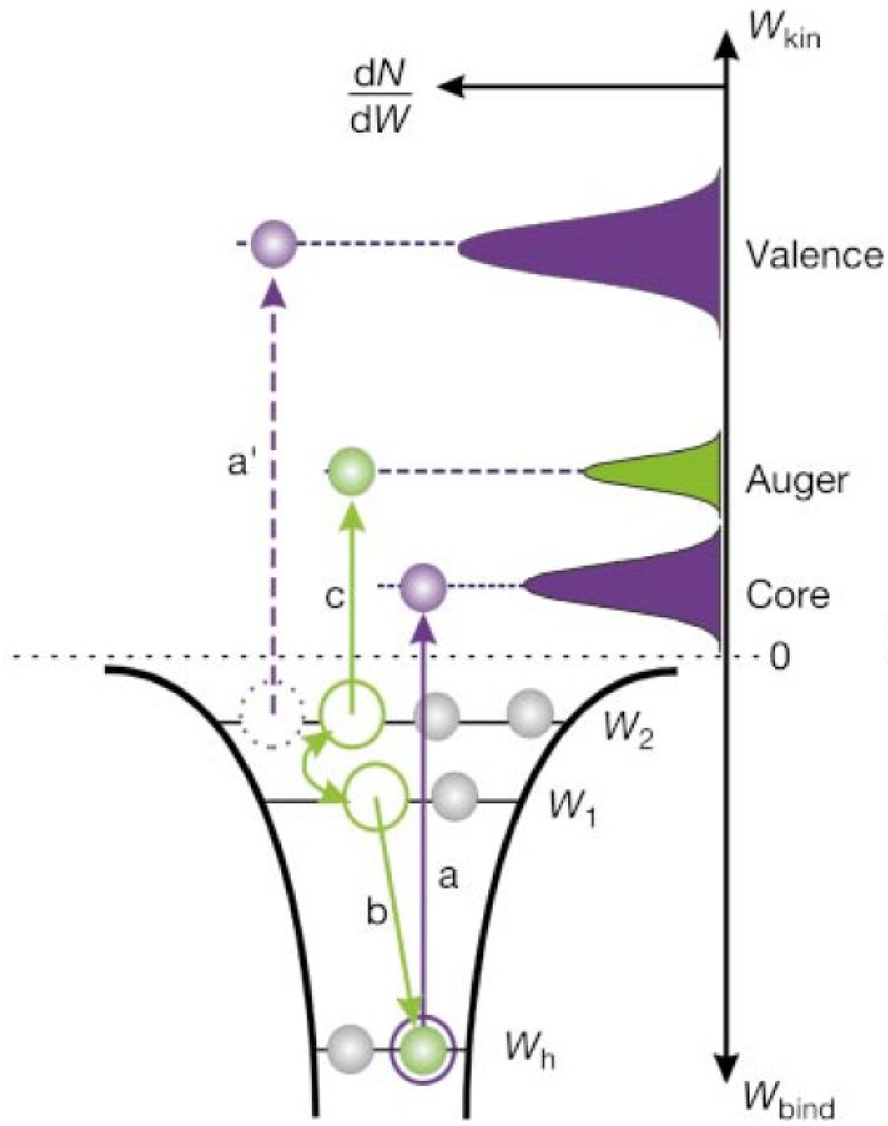
- Auger decay

Example:

Time-resolved atomic inner shell spectroscopy, Drescher et al., Nature 419, 803 (2002)



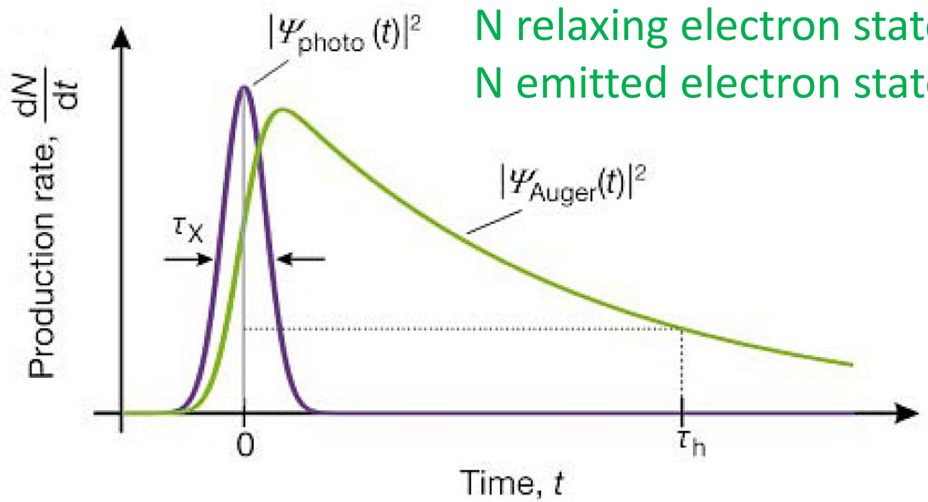
Attosecond measurement of Auger decay



Photoionization of Kr at 95 eV leads to both the removal of valence electrons and that of 3d core M-shell electrons (purple)

The removal of a core electron may be followed by an MNN Auger decay (green), allowing a measurement of the liftime of the core hole

M core level hole,
N relaxing electron state
N emitted electron state



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

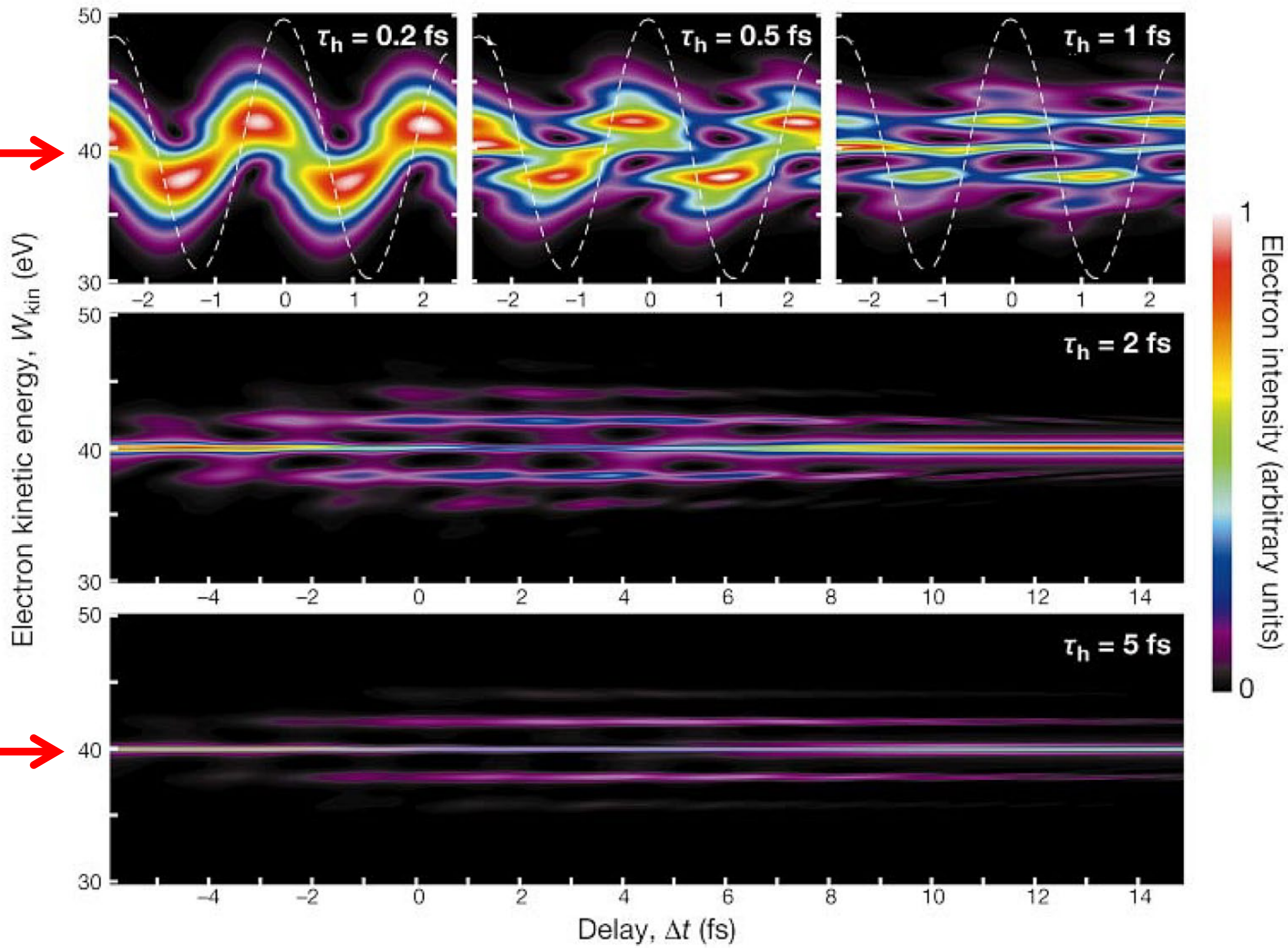
Attosecond measurement of Auger decay

streaking regime

$\tau_{Auger} << \tau_{laser}$

sideband regime

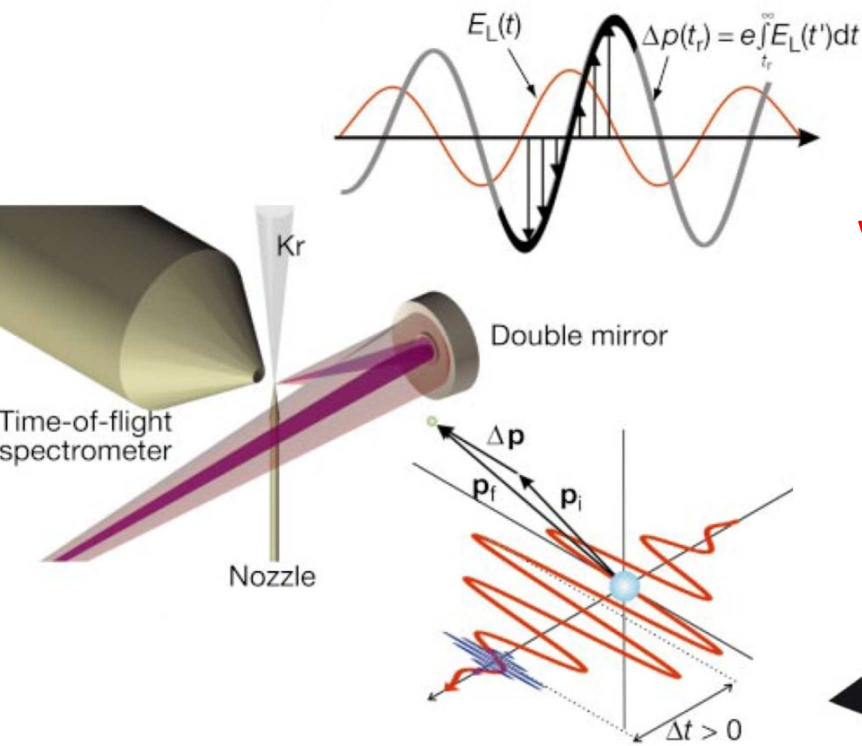
$\tau_{Auger} >> \tau_{laser}$

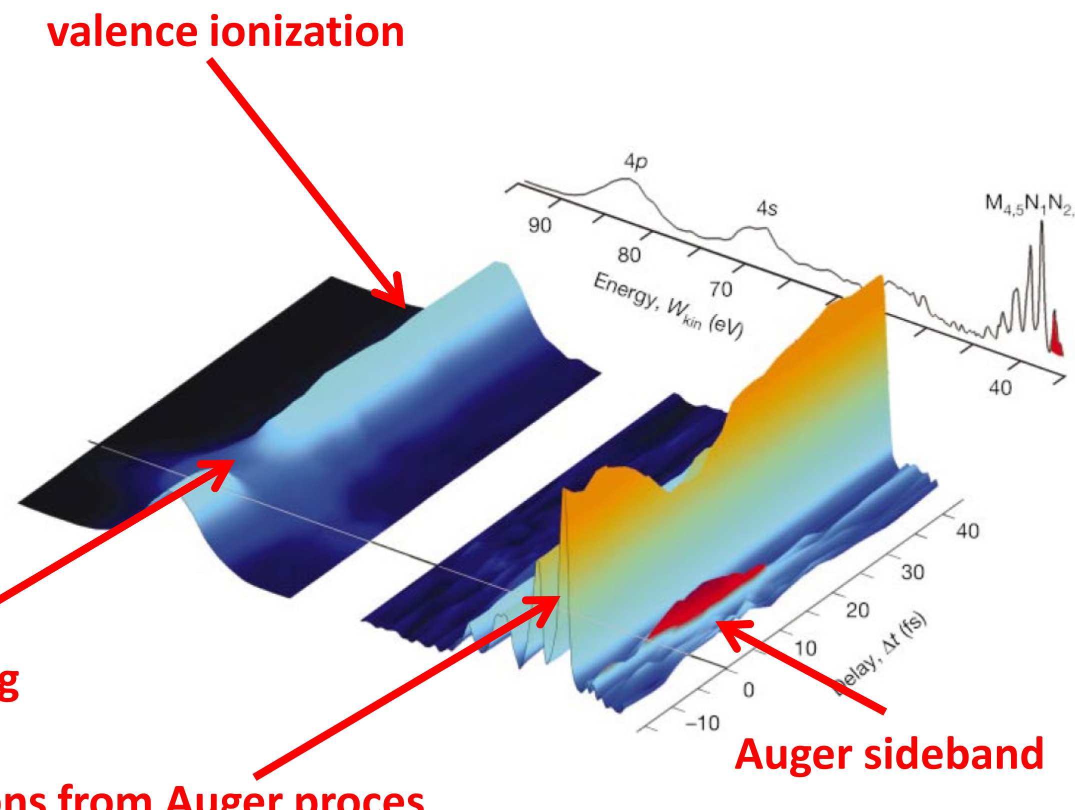


M. Drescher et al., Nature 419, 803 (2002)

Attosecond measurement of Auger decay

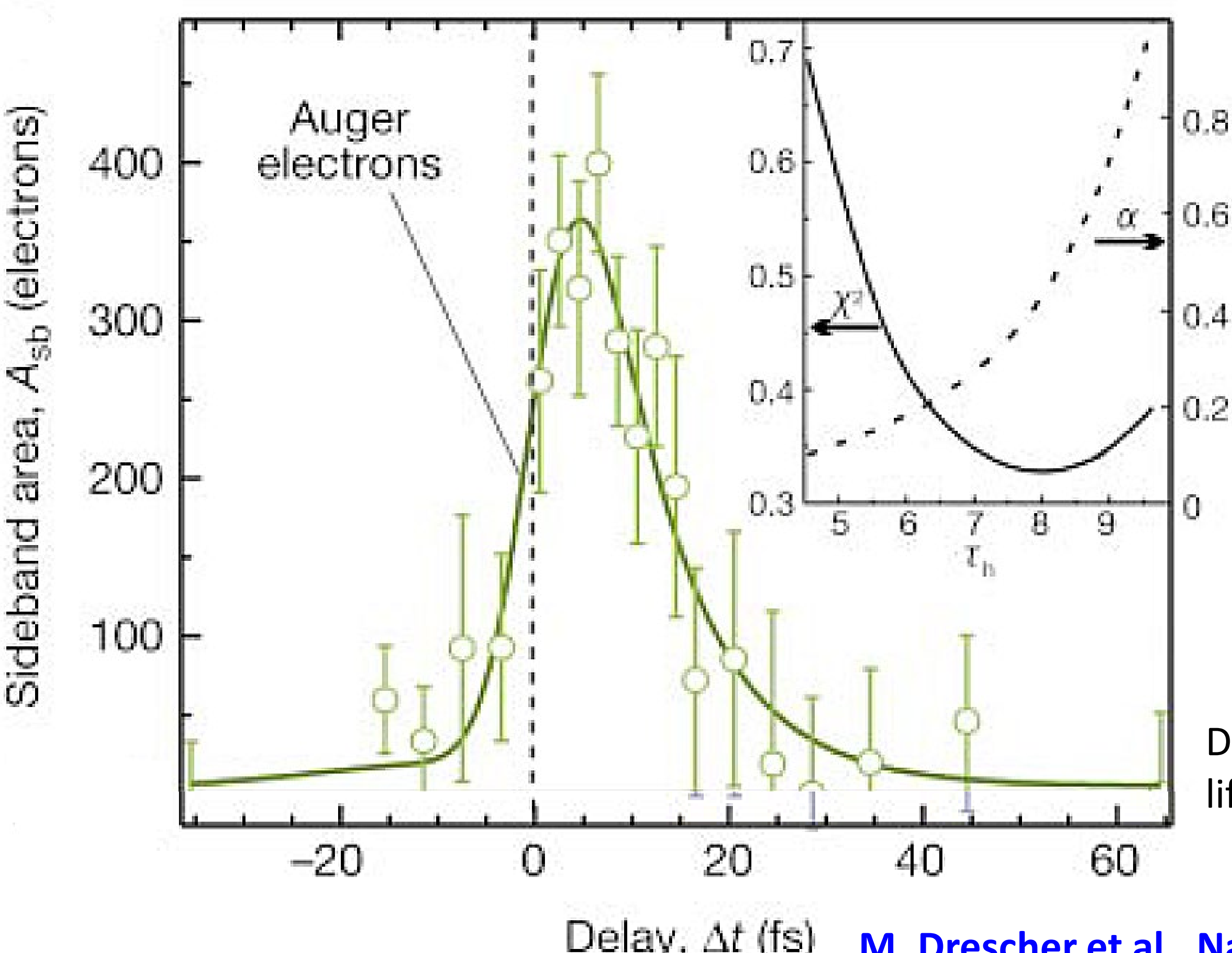
The Auger lifetime can be revealed by a streaking /sideband measurement





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

Attosecond measurement of Auger decay



Determination of Auger lifetime of 7.9 ± 1 fs

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