Optics - Extreme Nonlinear Optics, Attosecond Science and High-Field Physics

Attosecond Pump-Probe Spectroscopy - part 1

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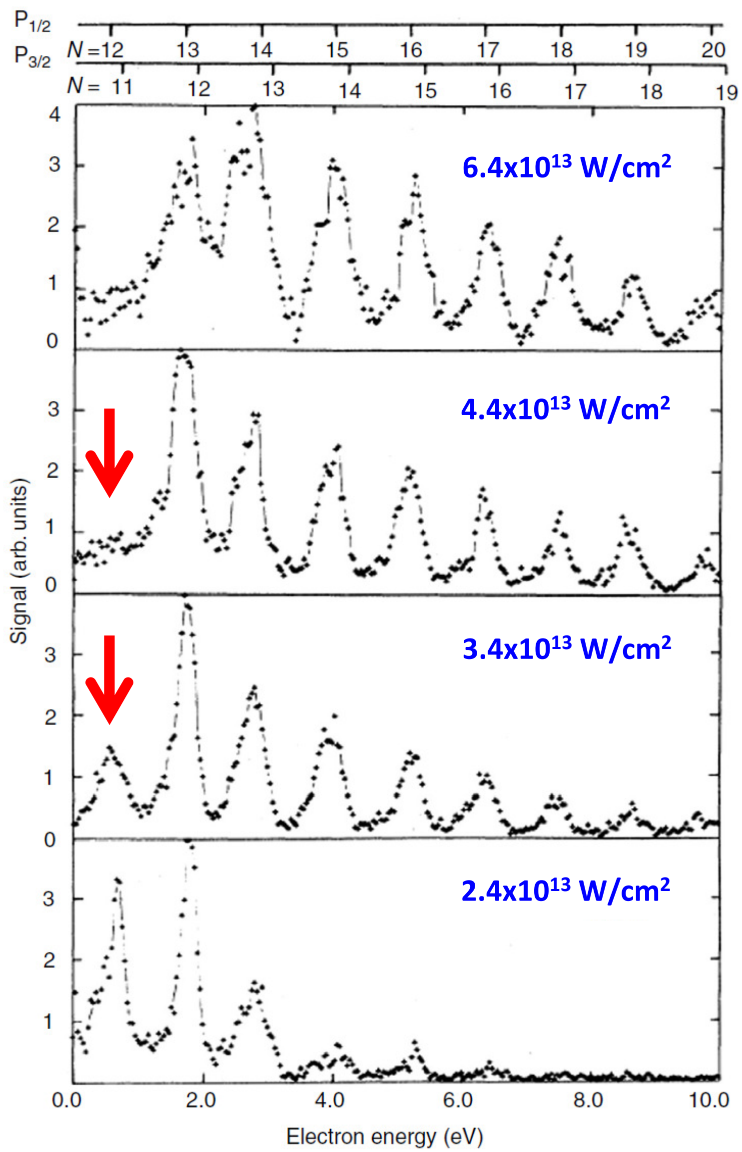
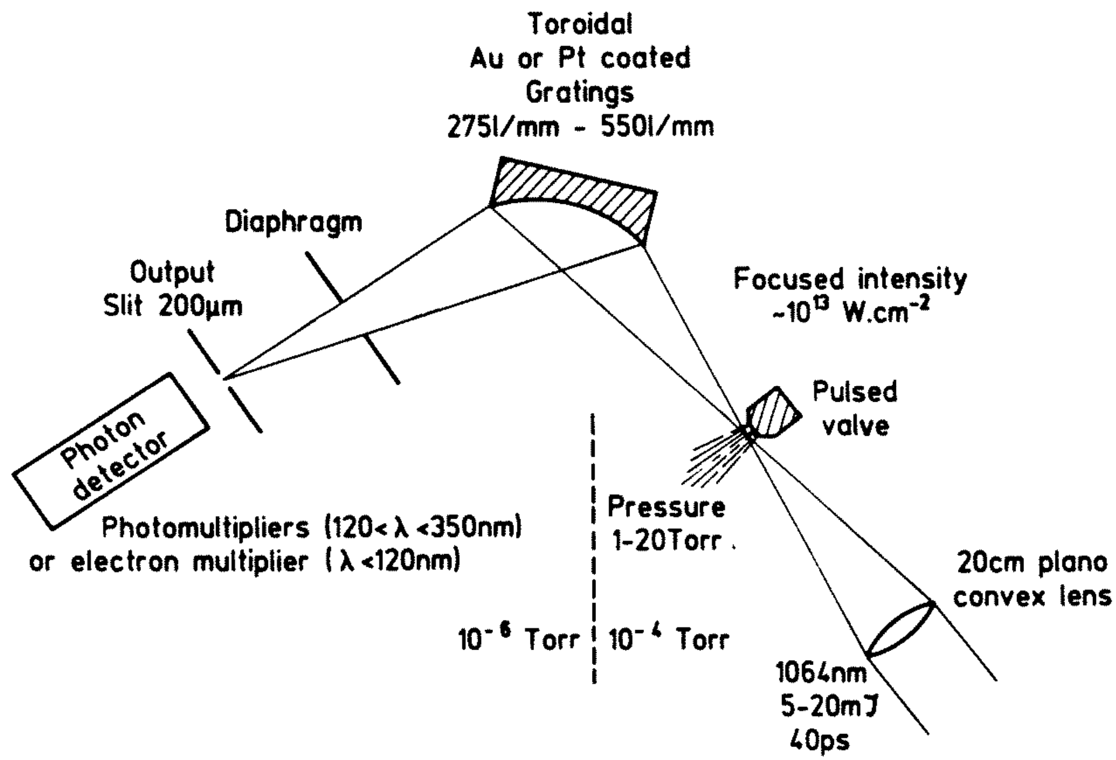
First, a short summary of a few things that you hopefully heard about last week....

Mid-1980´s: studies of ATI



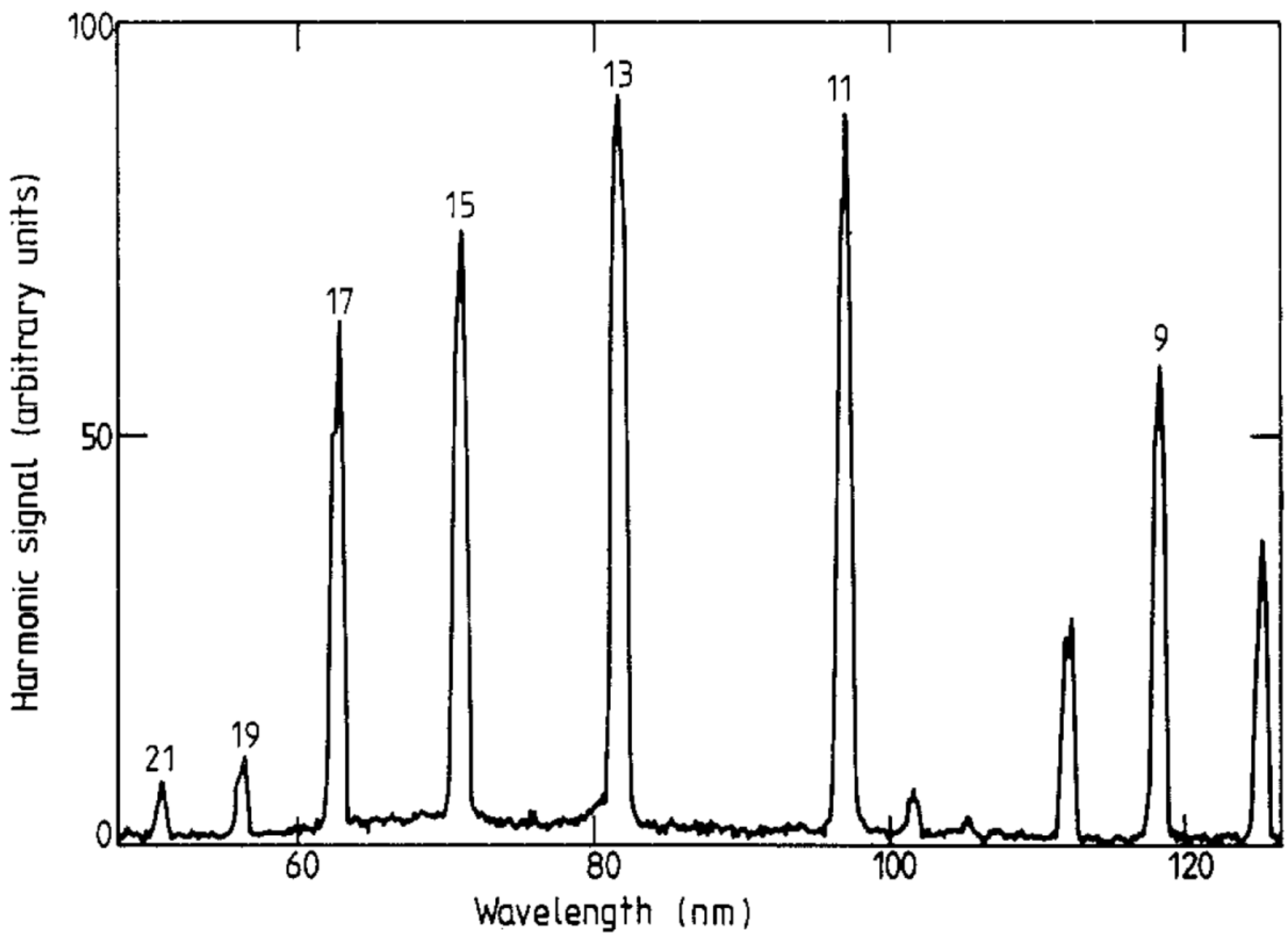
P. Kruit et al., Phys. Rev. A 28, 248 (1983)



Experimental setup at Saclay to measure the light emitted during above-threshold ionization experiments

A. L´Huillier et al., in ‘Atoms in Intense Laser Fields’, edited by Gavrila and Muller, (Academic Press, 1992)

Discovery of High-Harmonic Generation (HHG)

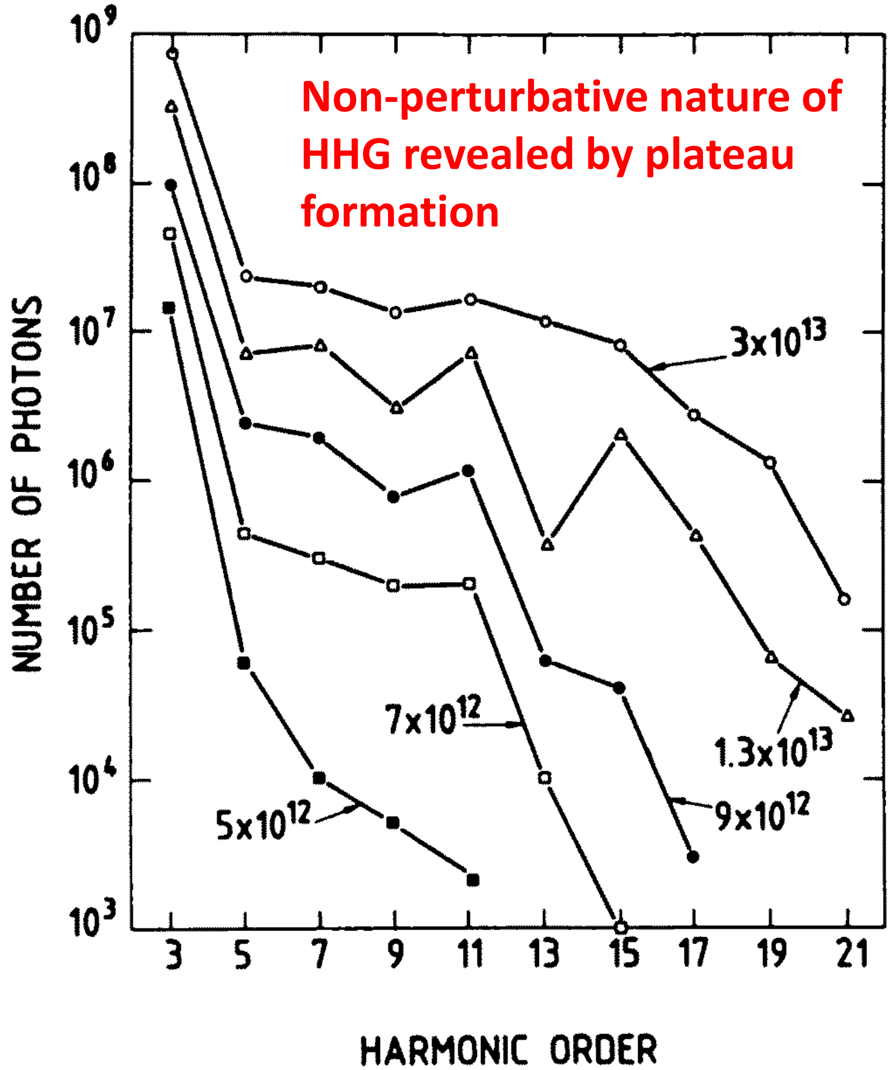


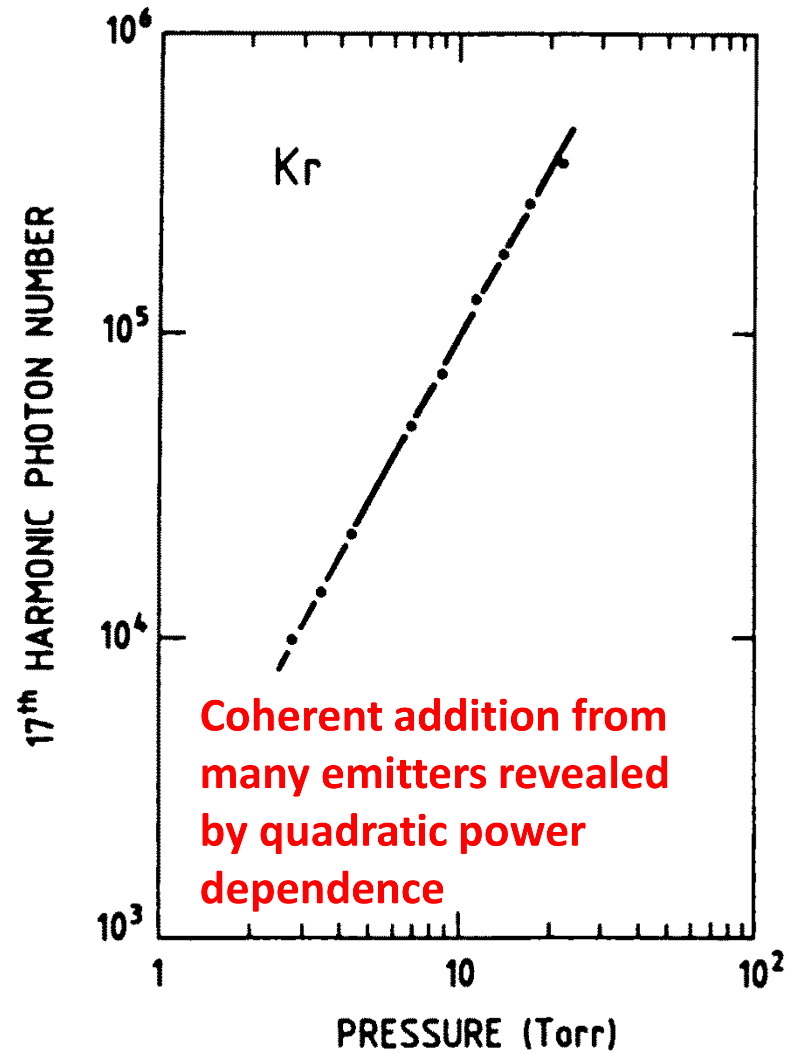
M. Ferray et al., J. Phys. B 21 L31 (1988)

High-harmonic generation in Xe using a 30 ps, 1064 nm laser focused to ca. 10^{13} W/cm^2

+ similar observations around the time in the Rhodes-group using 248 nm driver lasers

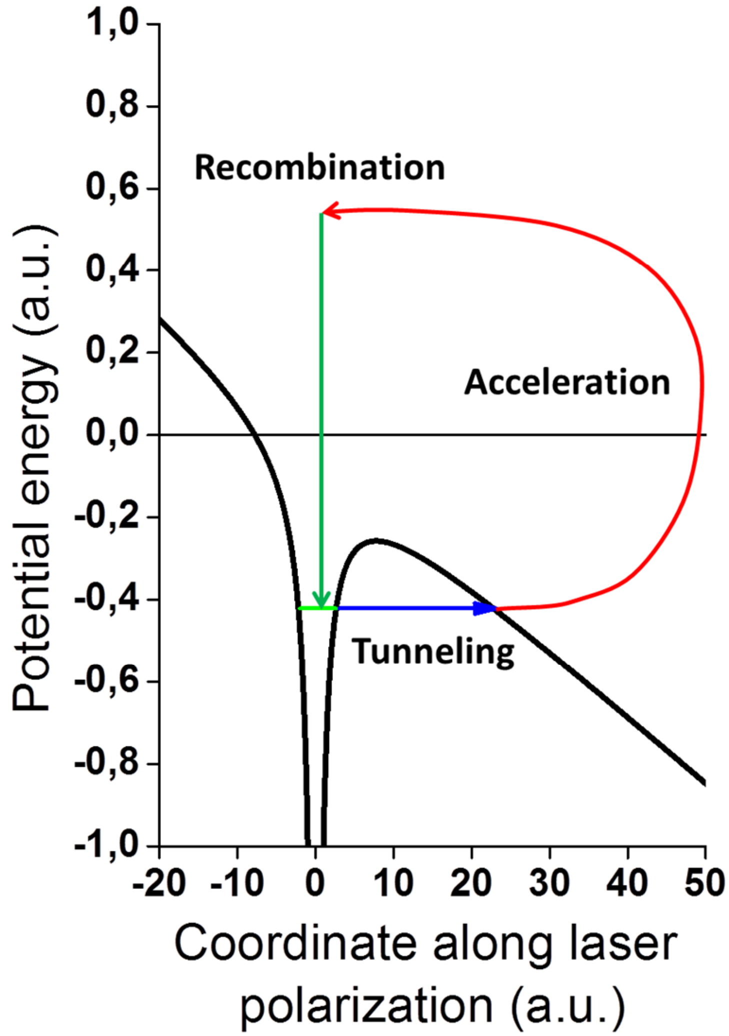
Intensity and pressure dependence

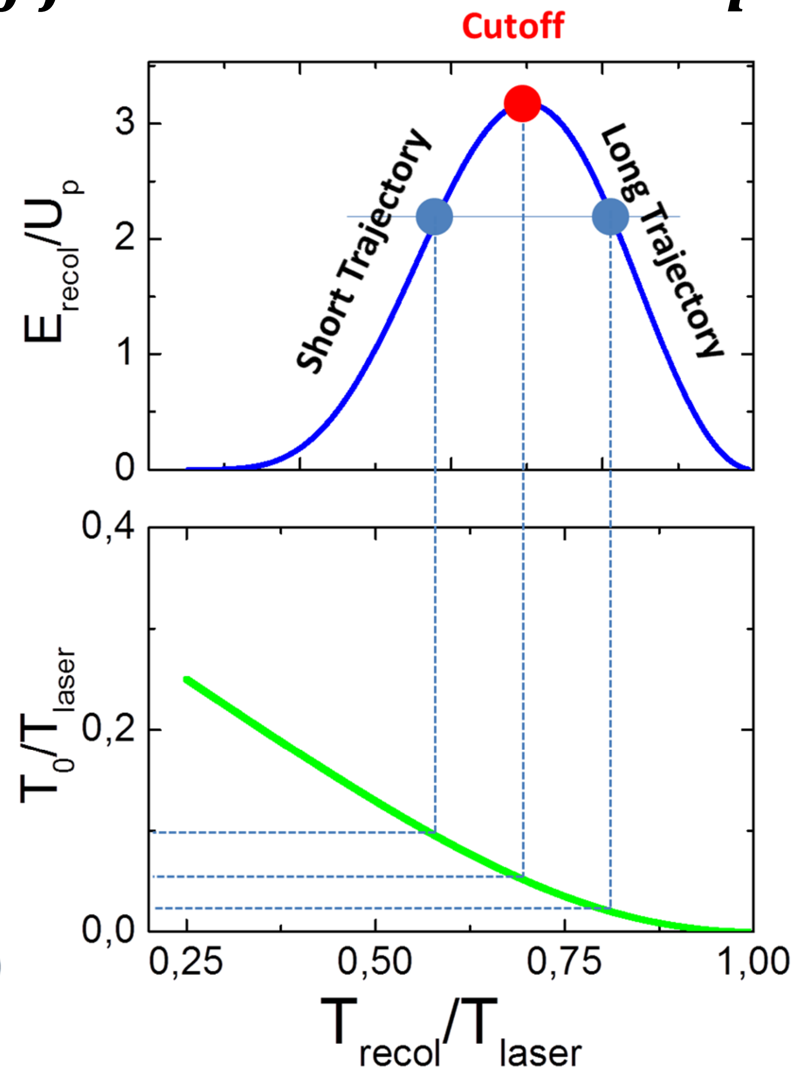




A. L´Huillier et al., in 'Atoms in Intense Laser Fields ', edited by Gavrila and Muller, (Academic Press, 1992)

Cut-off law: $E_{cutoff} = IP + 3.17\ U_p$





K. Schafer et al., Phys. Rev. Lett. 70, 1599 (1993)
P. Corkum. Phys. Rev. Lett. 71, 1994 (1993)

The main triumph of the three-step model was that it explained the cut-off law

Attosecond pulses by HHG

Physics Letters A 168 (1992) 447–450
North-Holland

PHYSICS LETTERS A

Proposal for attosecond light pulse generation using laser induced multiple-harmonic conversion processes in rare gases

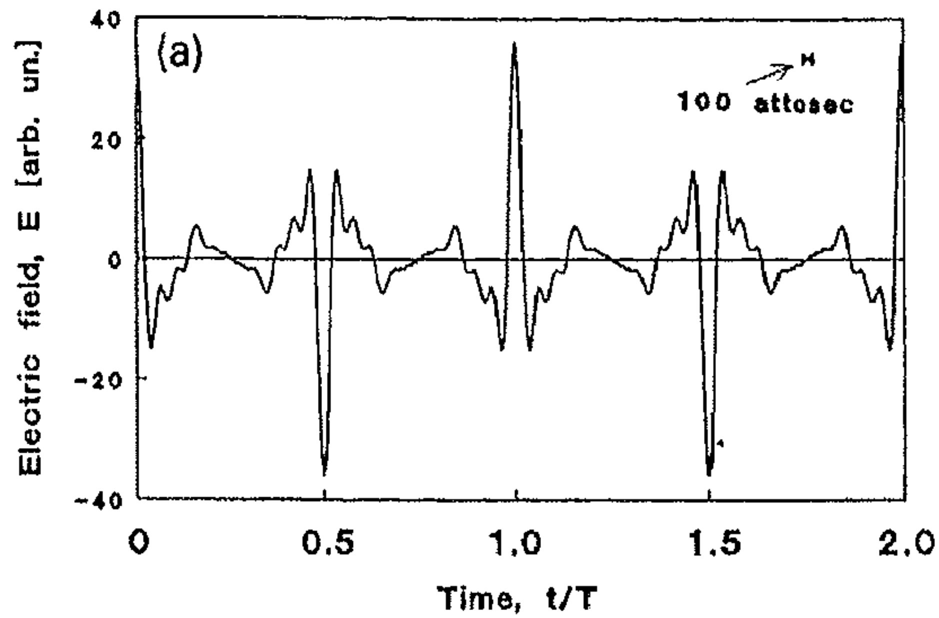
Gy. Farkas and Cs. Tóth

Research Institute for Solid State Physics, Central Research Institute for Physics, P.O. Box 49, H-1525 Budapest, Hungary

Received 11 June 1992; accepted for publication 13 July 1992
Communicated by V.M. Agranovich

Table 1
Resulting pulse durations and pulse height increase ratios after the Fourier synthesis of high harmonic components of the plateau region for various noble gases. The first (n_p) and the last (n_c) component of the plateau region are given

Gas	n_p	n_c	τ from $E(t)$ (as)	τ from $E^2(t)$ (as)	Increase ratio $(\sum E_n^2)/E_n^2$
xenon	5	21	97	72	81
krypton	5	25	76	56	169
argon	7	29	74	52	180
neon	13	53	38	28	441



Attosecond pulses by HHG

Science **292**, 1689 (2001);
DOI: 10.1126/science.1059413

P. M. Paul,[1] E. S. Toma,[2] P. Breger,[1] G. Mullot,[3] F. Augé,[3] Ph. Balcou,[3] H. G. Muller,[2*] P. Agostini[1]

In principle, the temporal beating of superposed high harmonics obtained by focusing a femtosecond laser pulse in a gas jet can produce a train of very short intensity spikes, depending on the relative phases of the harmonics. We present a method to measure such phases through two-photon, two-color photoionization. We found that the harmonics are locked in phase and form a train of 250-attosecond pulses in the time domain. Harmonic generation may be a promising source for attosecond time-resolved measurements.

Attosecond metrology

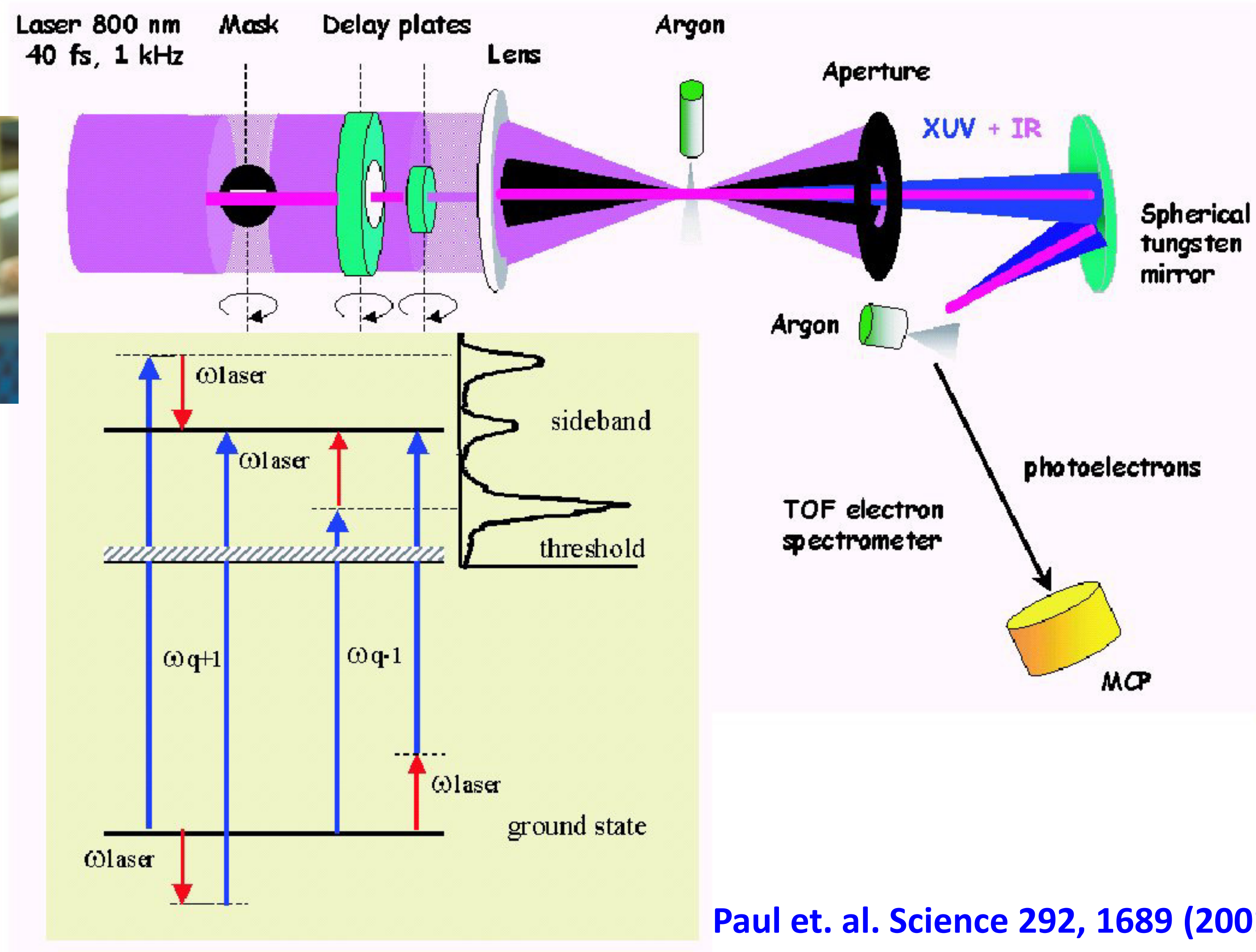
M. Hentschel*†, R. Kienberger*†, Ch. Spielmann*, G. A. Reider*, N. Milosevic*, T. Brabec*, P. Corkum‡, U. Heinzmann§, M. Drescher§ & F. Krausz*

The generation of ultrashort pulses is a key to exploring the dynamic behaviour of matter on ever-shorter timescales. Recent developments have pushed the duration of laser pulses close to its natural limit—the wave cycle, which lasts somewhat longer than one femtosecond (1 fs = 10^{-15} s) in the visible spectral range. Time-resolved measurements with these pulses are able to trace dynamics of molecular structure, but fail to capture electronic processes occurring on an attosecond (1 as = 10^{-18} s) timescale. Here we trace electronic dynamics with a time resolution of $\leq$ 150 as by using a subfemtosecond soft-X-ray pulse and a few-cycle visible light pulse. Our measurement indicates an attosecond response of the atomic system, a soft-X-ray pulse duration of 650 $\pm$ 150 as and an attosecond synchronism of the soft-X-ray pulse with the light field. The demonstrated experimental tools and techniques open the door to attosecond spectroscopy of bound electrons.

RABBITT

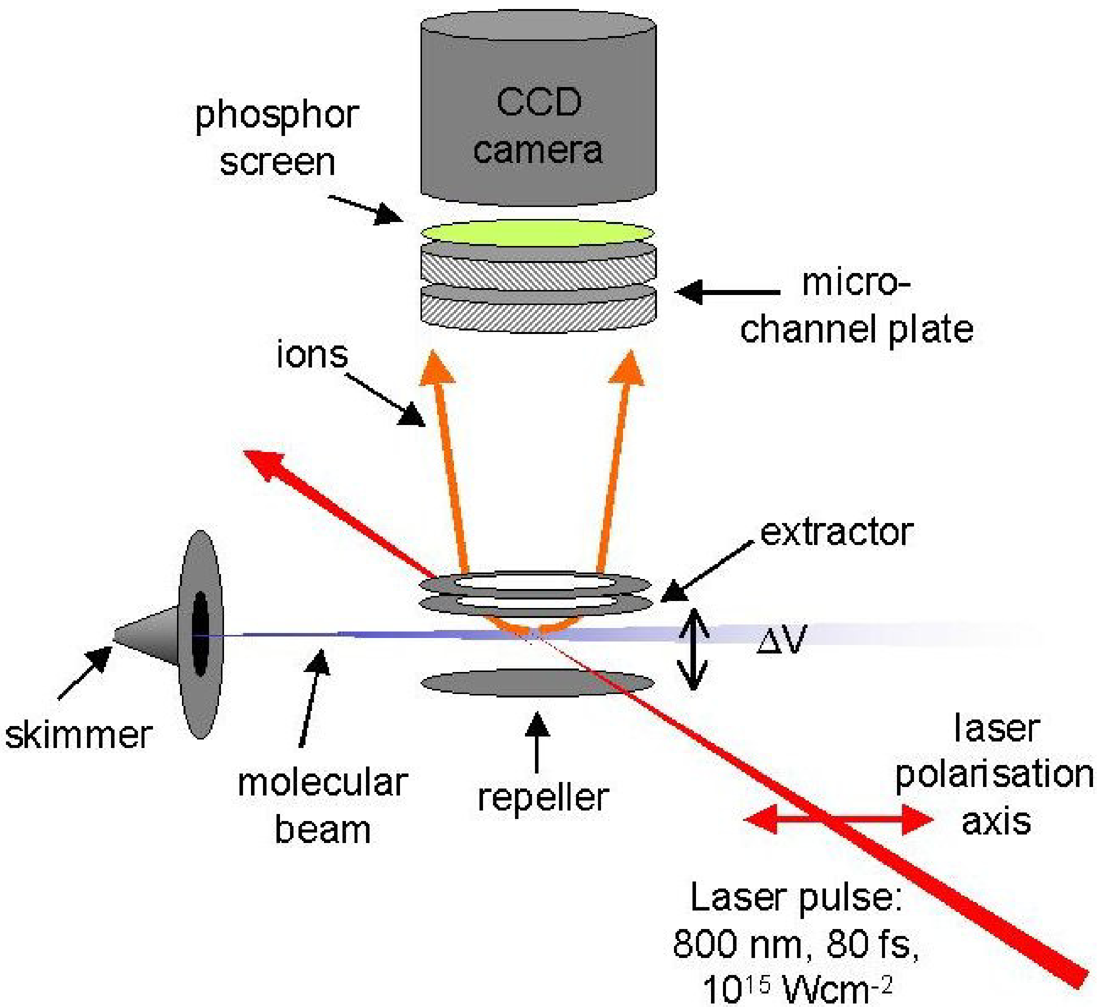
Reconstruction of Attosecond harmonic Beating By Interference of Two-photon Transitions

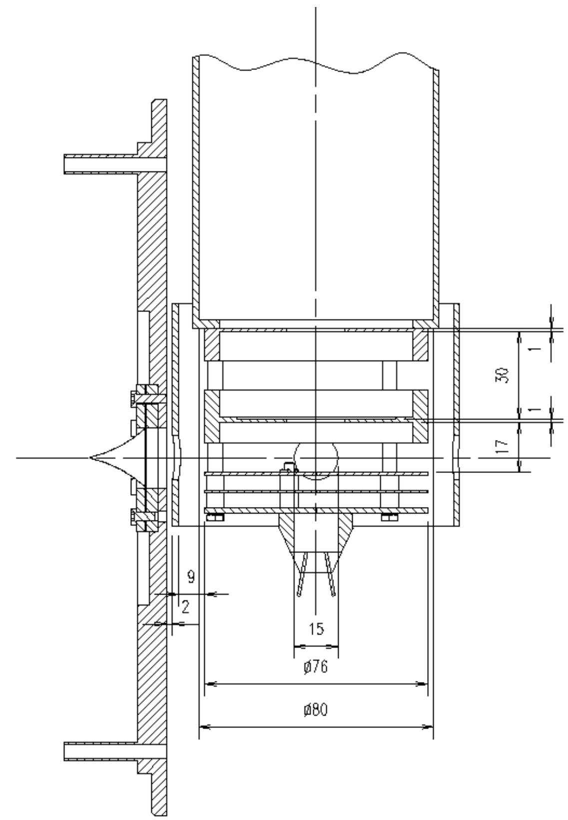
Harm-Geert Muller



Paul et. al. Science 292, 1689 (2001)

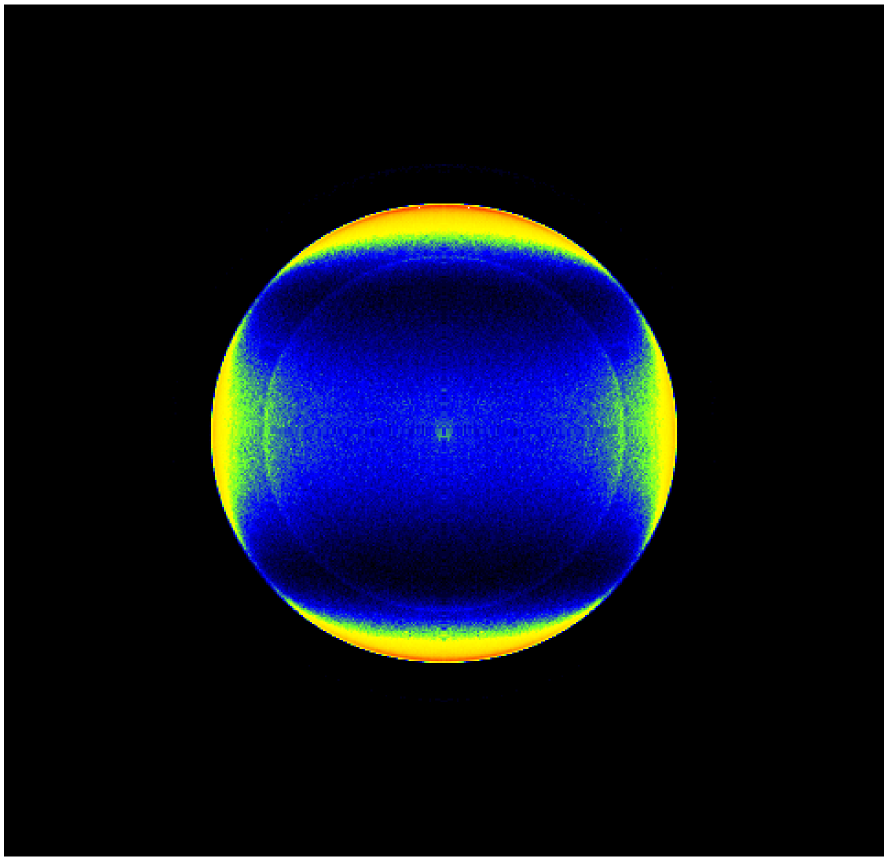
Velocity map ion & photoelectron imaging



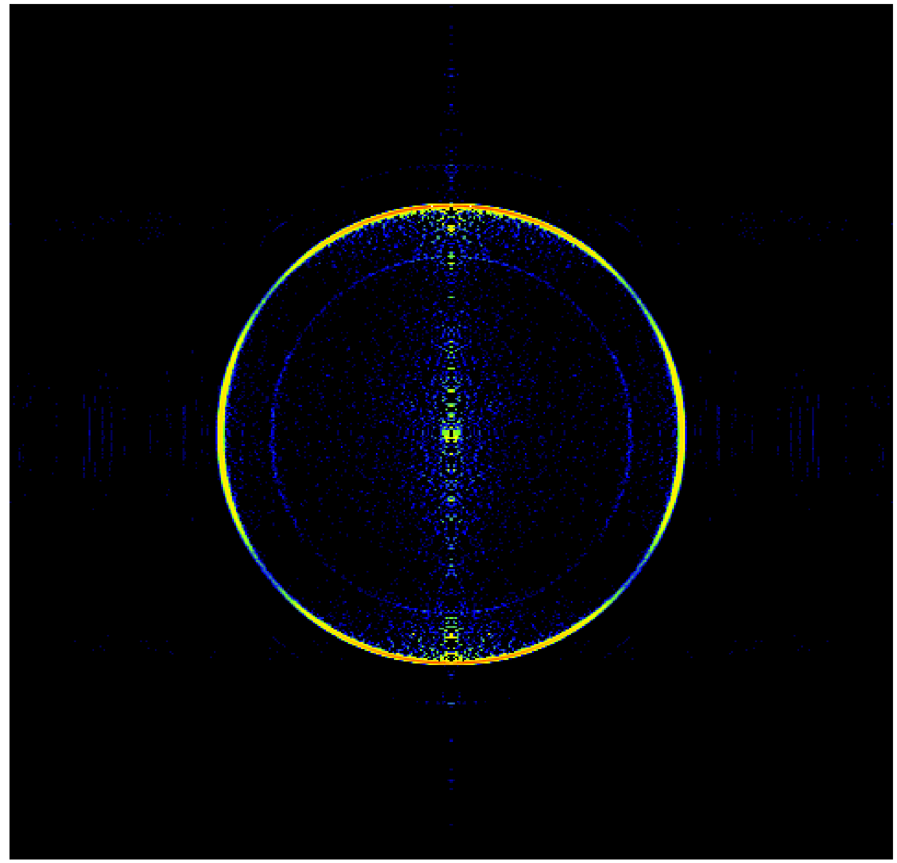


A. T. J. B. Eppink, D. H. Parker, *Review of Scientific Instruments* 68, 3477 (1997).

Extraction of the energy and angular distribution using an iterative procedure

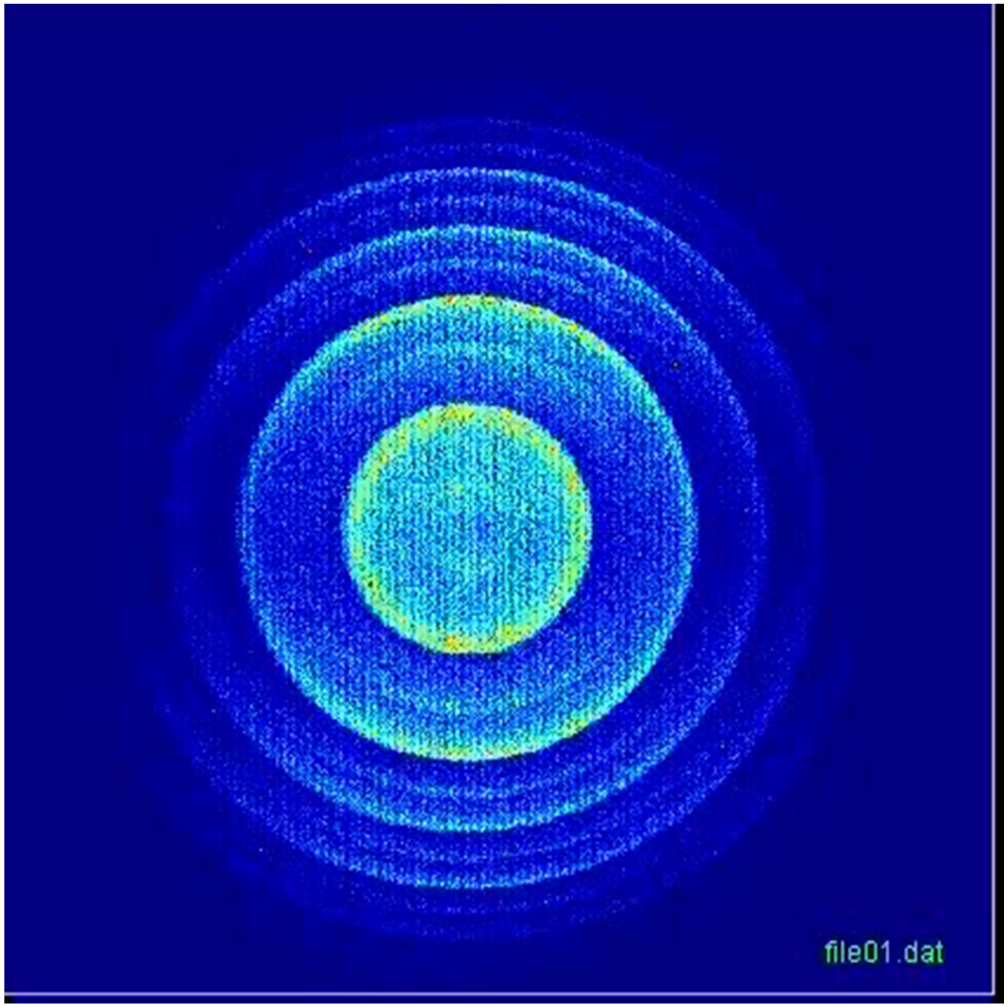


Raw image for 2-photon ionisation of Ar by 532 nm light



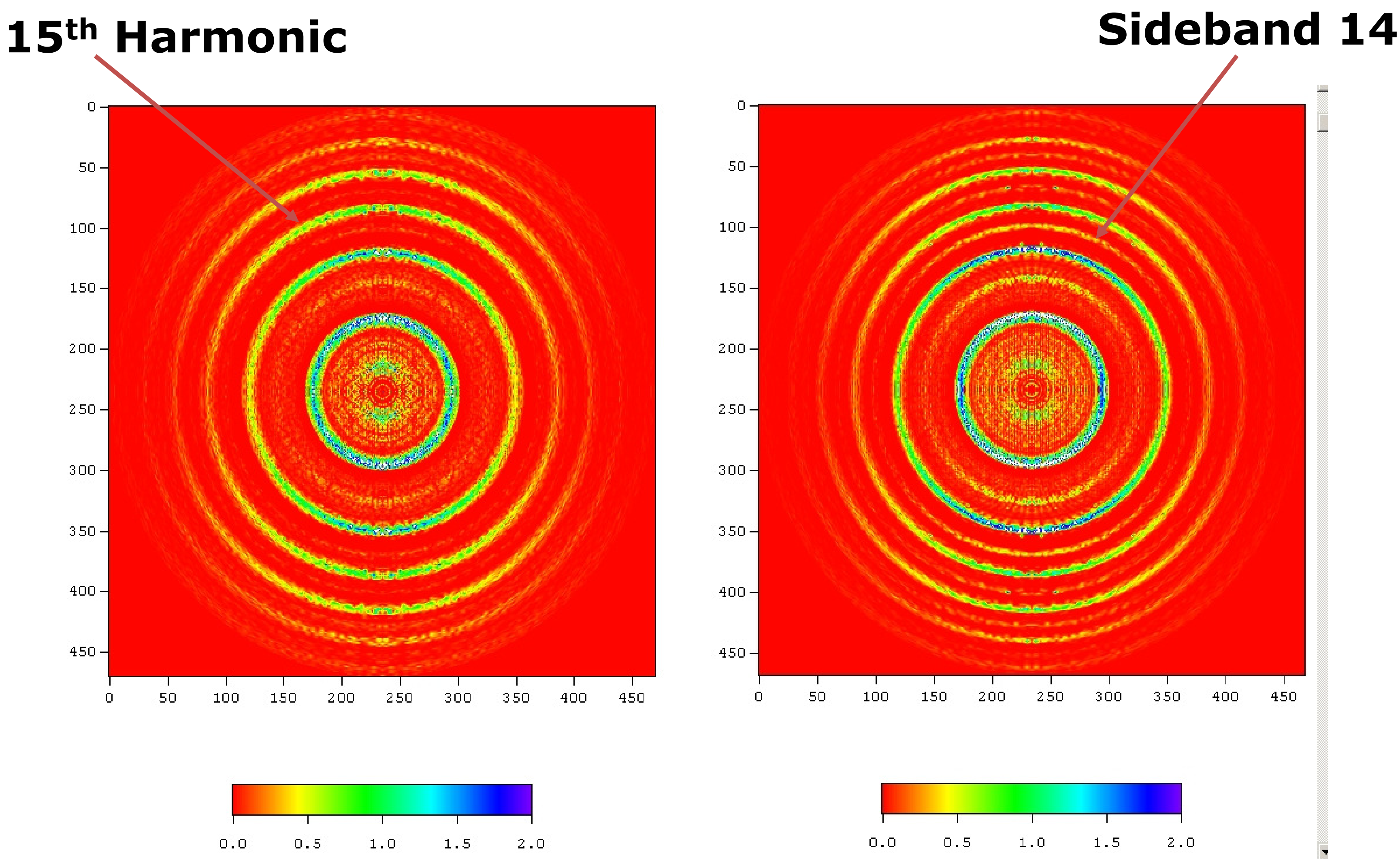
Slice through the 3D velocity distribution, obtained by Abel inversion of the image $\Delta v/v = 1\%$

Raw Photoelectron Images vs Time-delay



Aseyev et.al., Phys. Rev. Lett. 91, 223902 (2003)

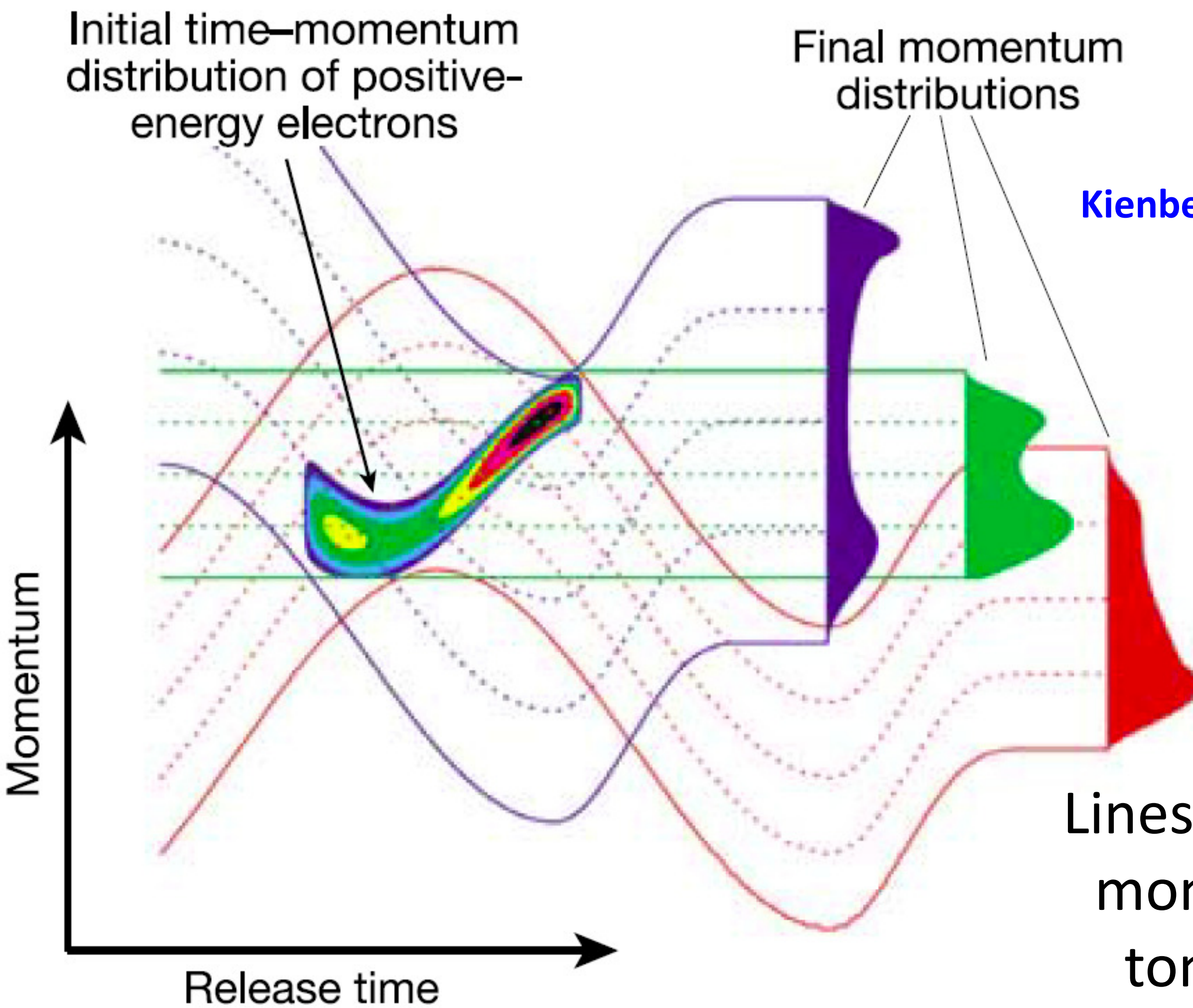
Inverted Photoelectron Image



Aseyev et.al., Phys. Rev. Lett. 91, 223902 (2003)

Characterizing Isolated Attosecond Pulses

In a laser field the conserved quantity is the canonical momentum $\mathbf{p}=\mathbf{k}(t)+qA(t)=\mathbf{k}(t)-A(t)$

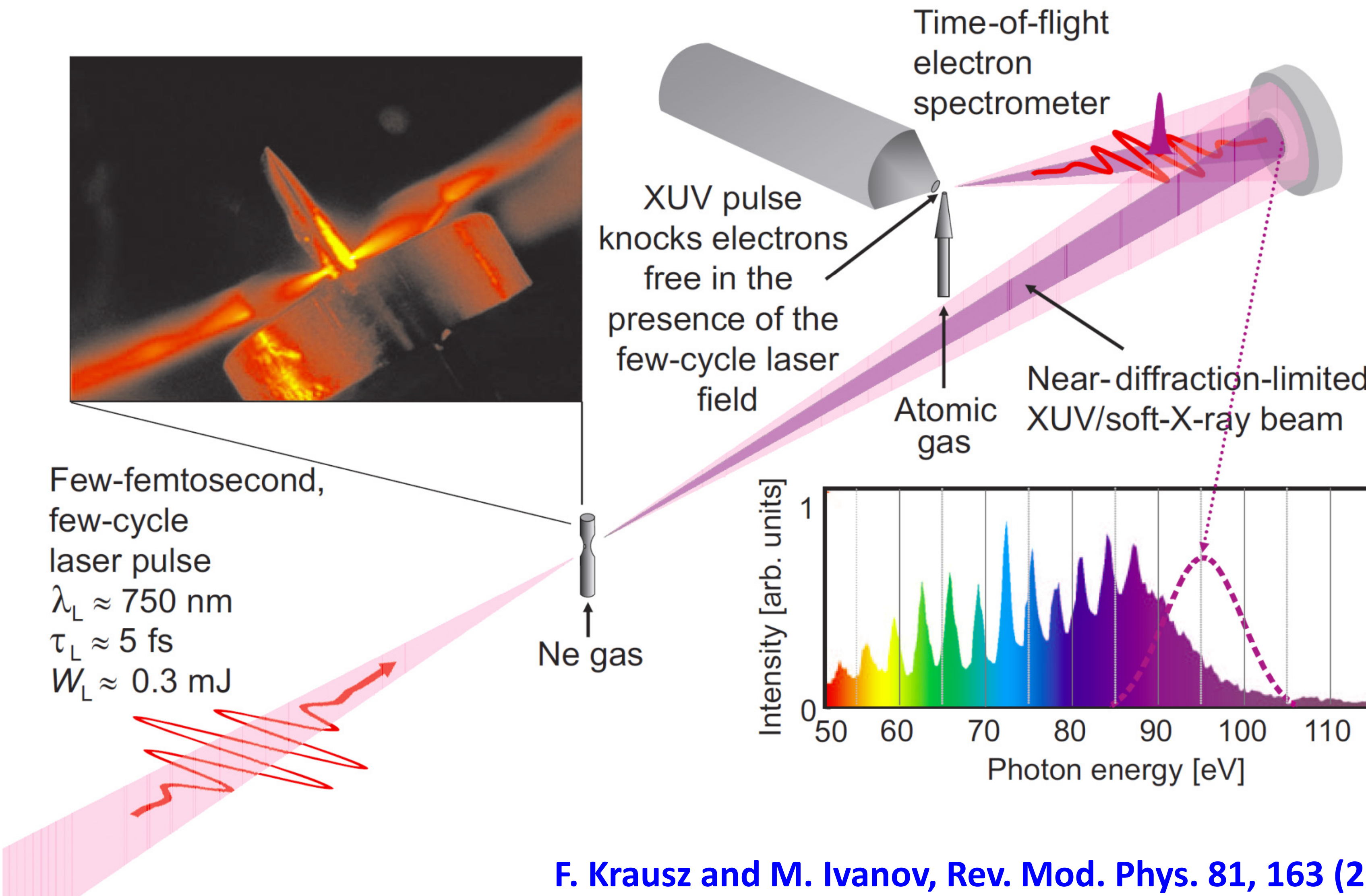


Kienberger et. al. Nature 427, 817 (2004)

Blue and **red**:
Lines of constant canonical momentum give different tomographic projections

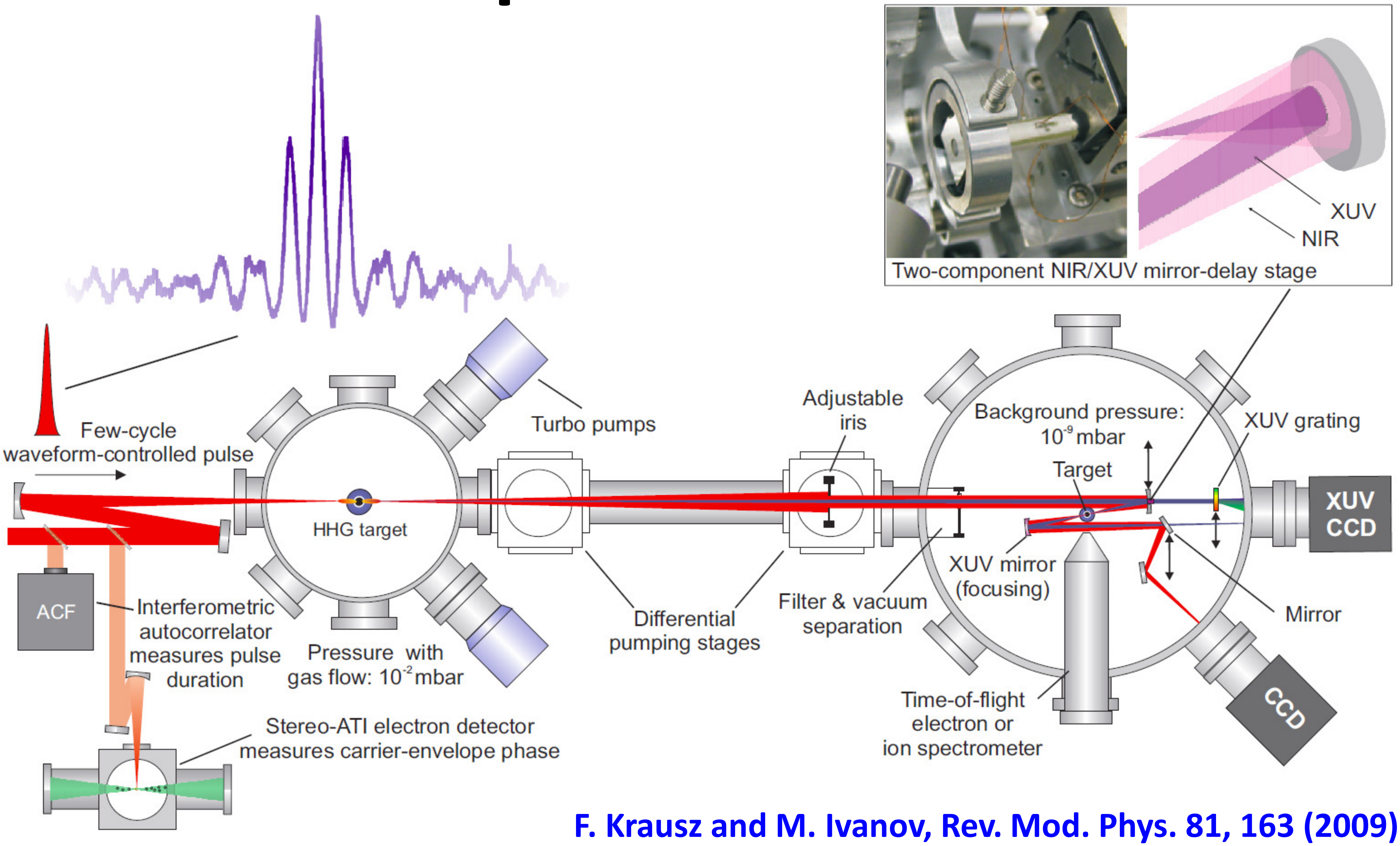
Attosecond pump-probe spectroscopy: atoms

Configuring attosecond pump-probe experiments - 1



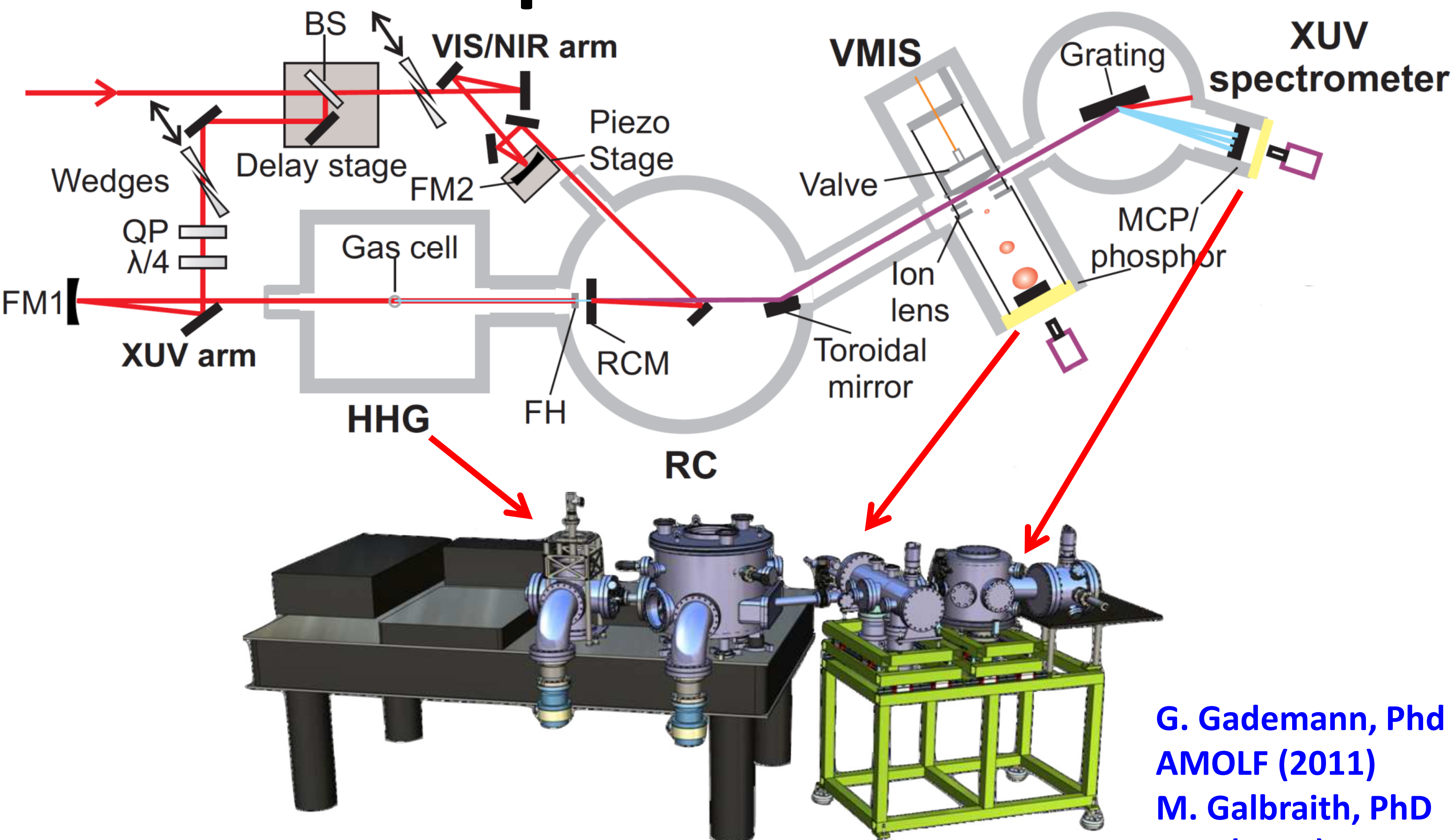
F. Krausz and M. Ivanov, Rev. Mod. Phys. 81, 163 (2009)

Configuring attosecond pump-probe experiments - 2



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

Configuring attosecond pump-probe experiments - 3

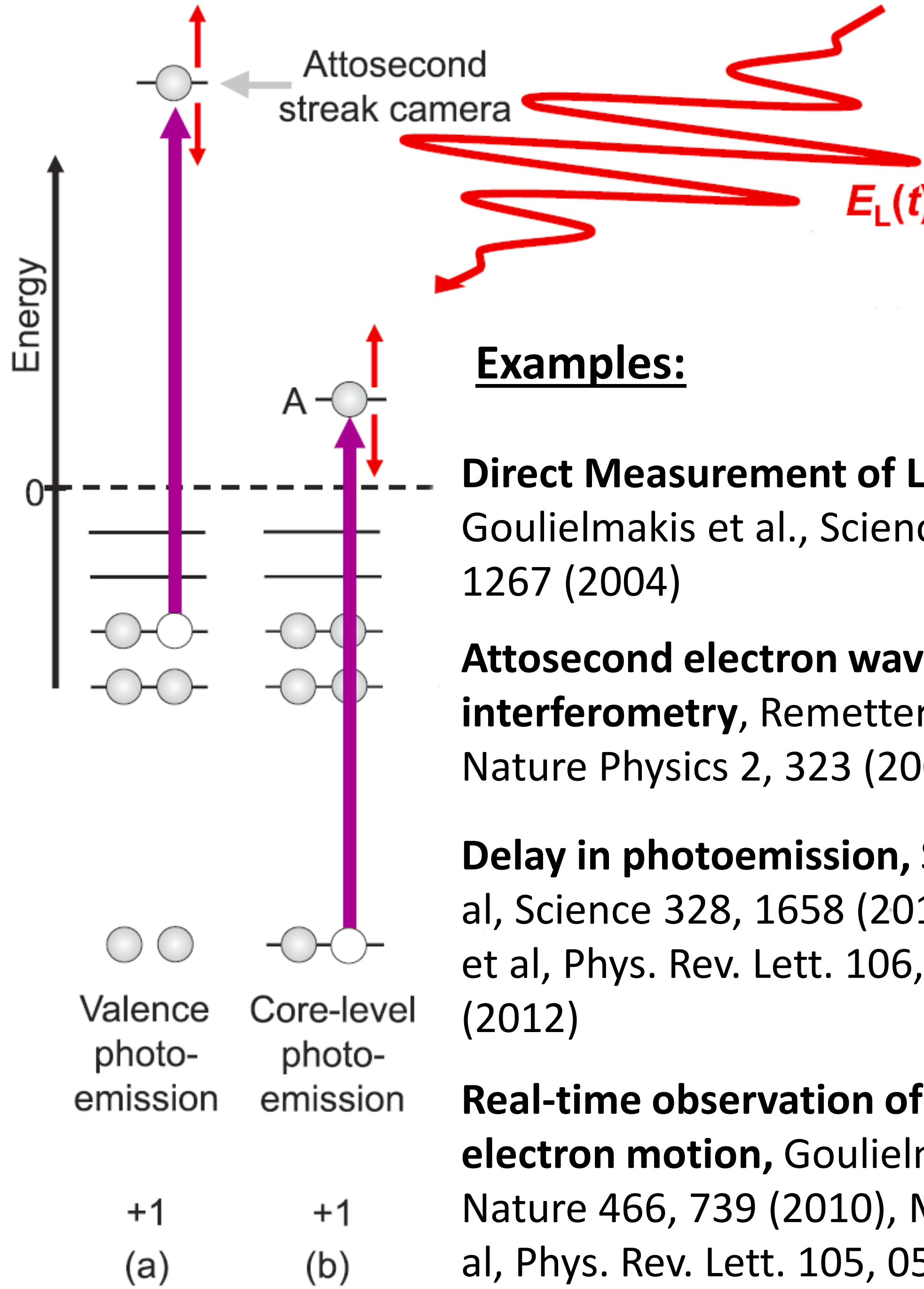


G. Gademann, Phd
AMOLF (2011)
M. Galbraith, PhD
MBI (2016)

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)

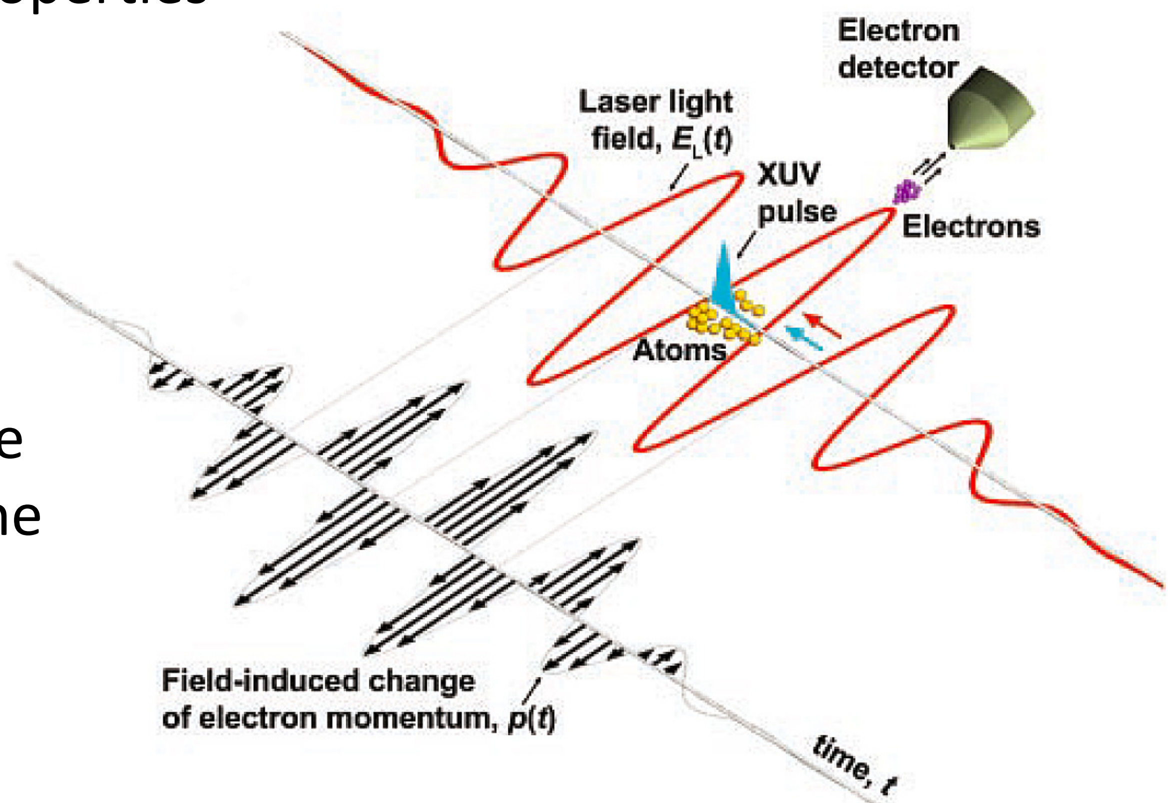
Real-time observation of valence electron motion, Goulielmakis et al., Nature 466, 739 (2010), Mauritsson et al, Phys. Rev. Lett. 105, 053001 (2010)

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



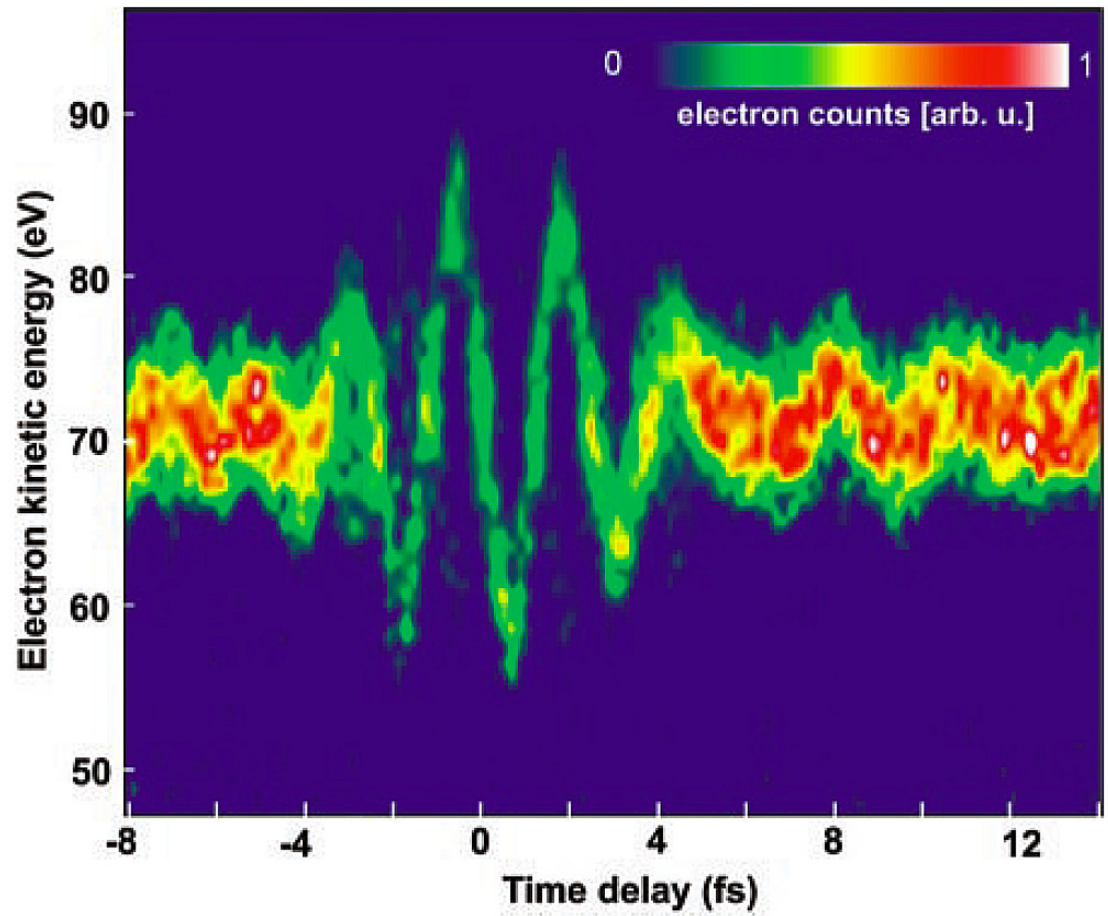
Goulielmakis et al., Science 305, 1267 (2004)

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

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

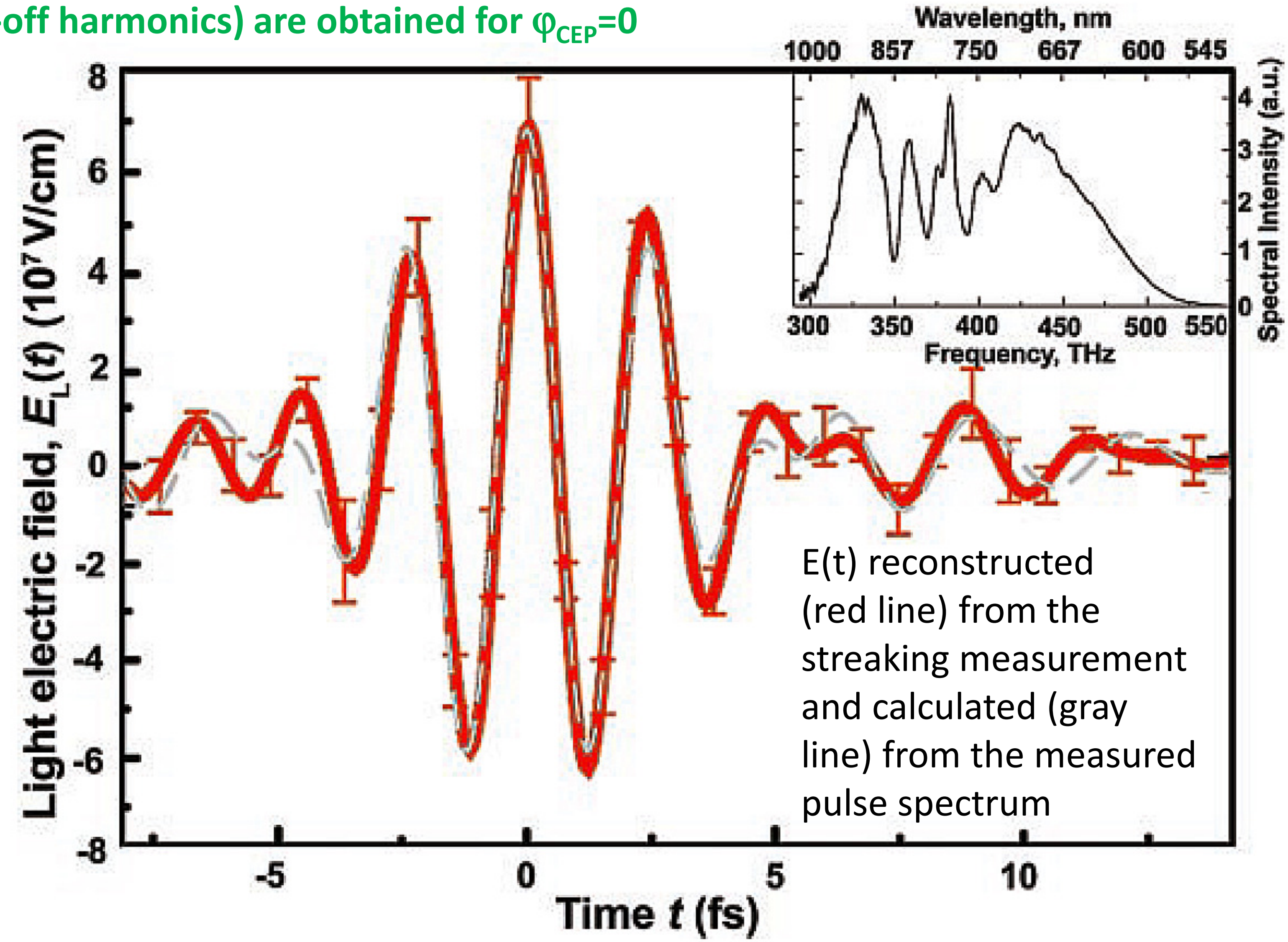
$$E(t) = -\frac{\partial A(t)}{\partial t}$$



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

Direct Measurement of Light Waves

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



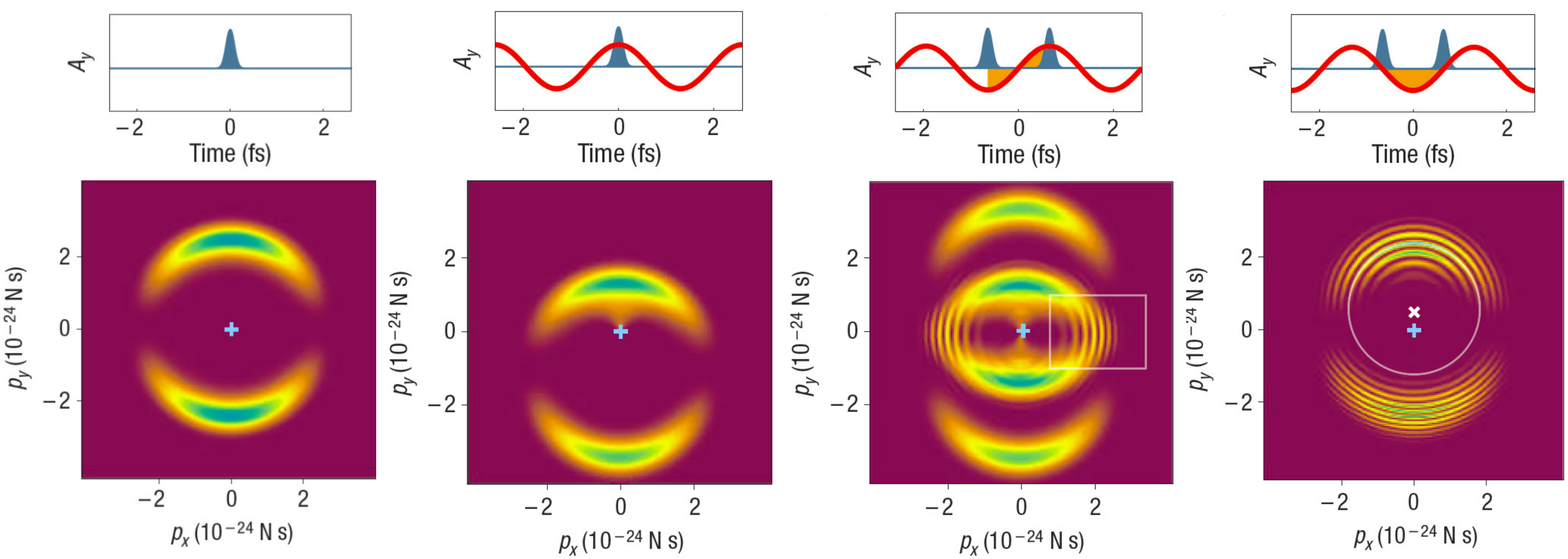
E(t) reconstructed (red line) from the streaking measurement and calculated (gray line) from the measured pulse spectrum

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

Attosecond electron wave packet interferometry

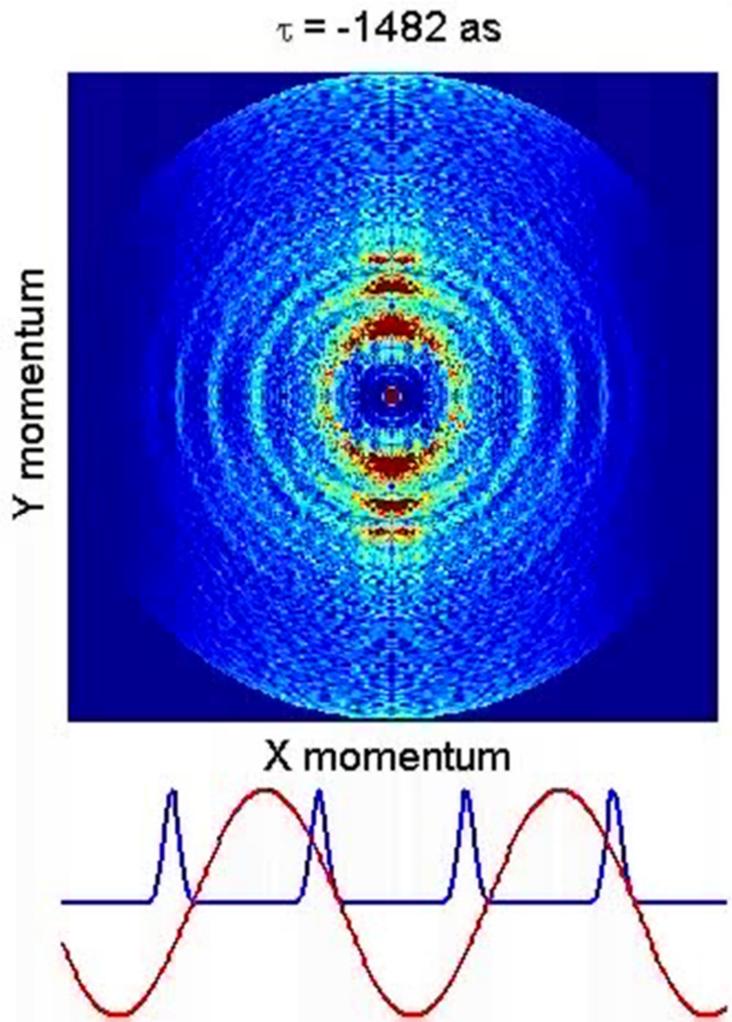
Extension of streaking spectroscopy to attosecond pulse trains
(or: RABBITT with non-perturbative fields)

Red line: A(t)



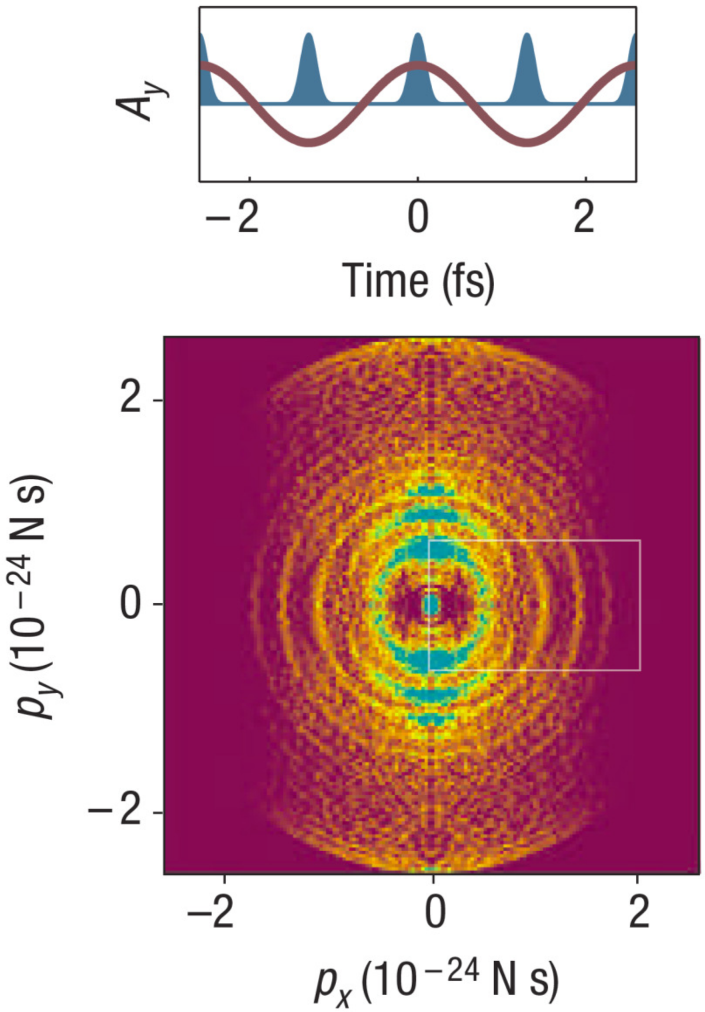
Remetter et al., Nature Physics 2, 323 (2006)

Attosecond electron wave packet interferometry



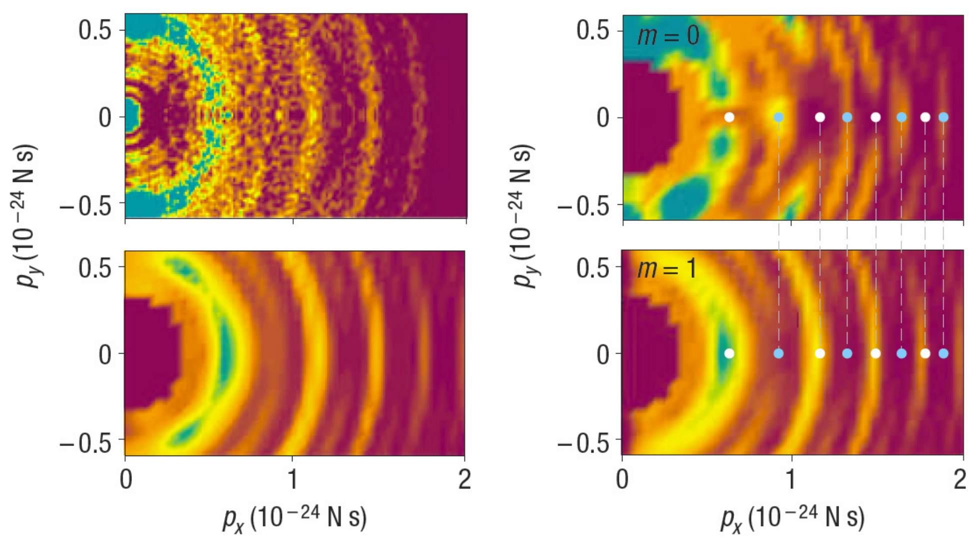
Remetter et al., Nature Physics 2, 323 (2006)

Attosecond electron wave packet interferometry



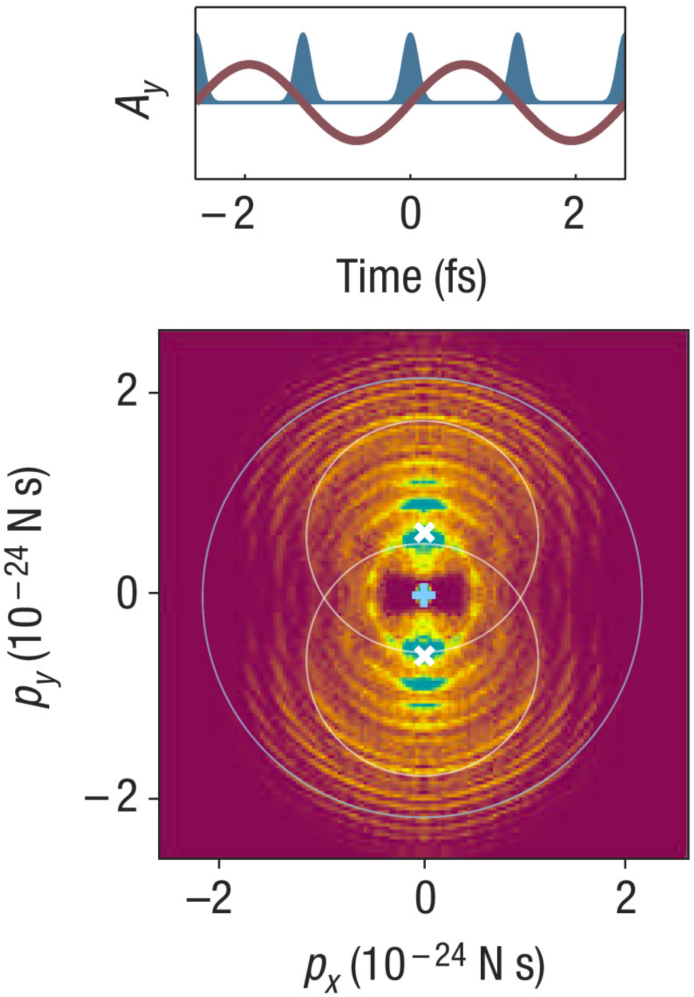
Attosecond pulses synchronized to maxima of the laser vector potential:

- **Opposite** momentum shifts for consecutive attosecond pulses
- Photoelectrons produced by consecutive attosecond pulses interfere



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

Attosecond electron wave packet interferometry



Attosecond pulses synchronized to zero-crossings of the laser vector potential:

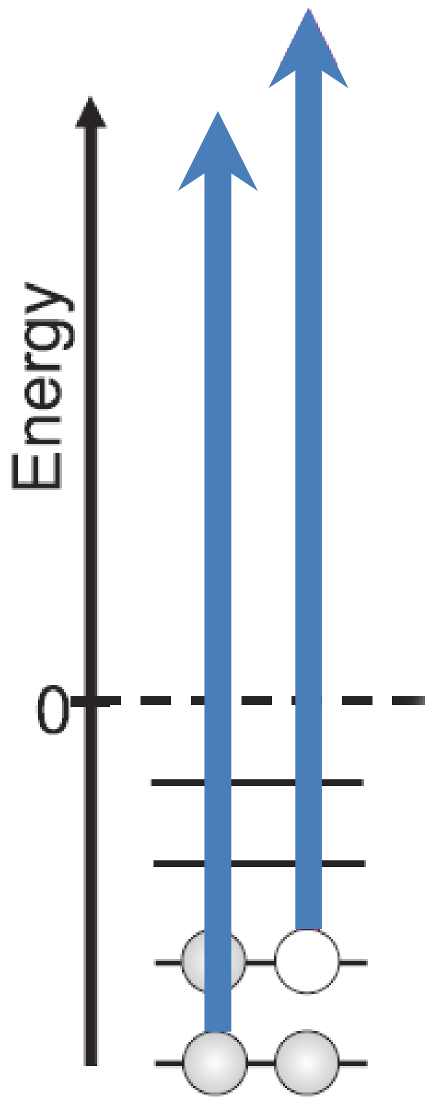
- **No** momentum shift
- Phase shift because of propagation in laser field; constructive interference

$$\frac{1}{2m}\left[p_x^2 + \left(p_y + (-1)^k \frac{2eA_{laser,0}}{\pi}\right)^2\right] =$$

$$(2n+1)\frac{h}{2\pi}\omega_{laser} - IP + \left(\frac{8}{\pi^2} - 1\right)U_p$$

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?

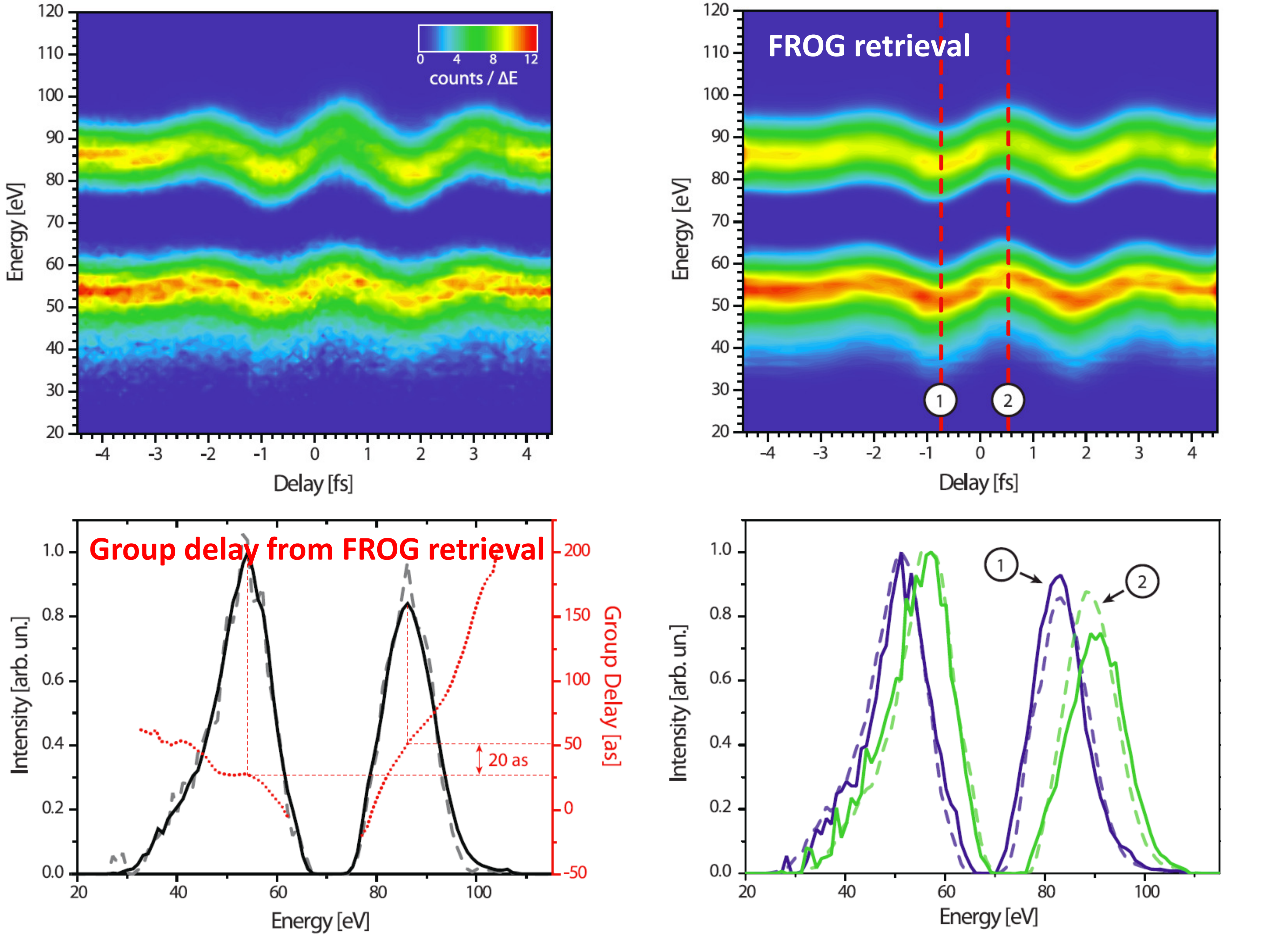
Two experimental approaches:

- 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)



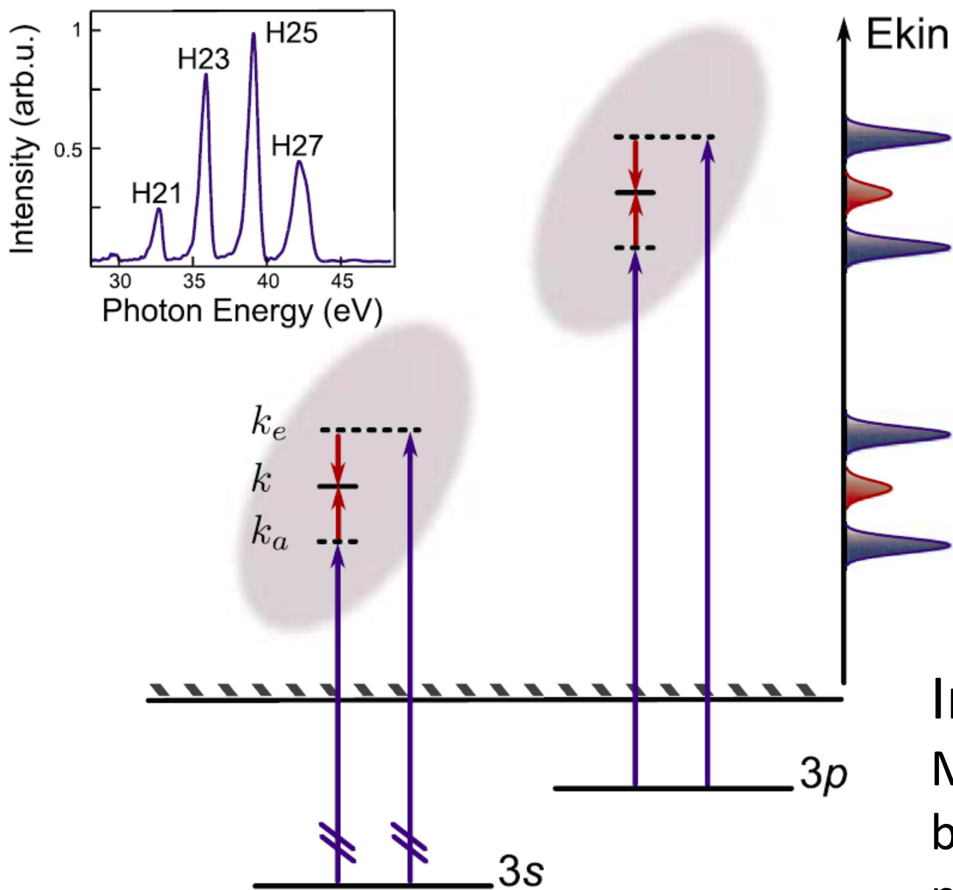
Schultze et al, Science 328, 1658 (2010)

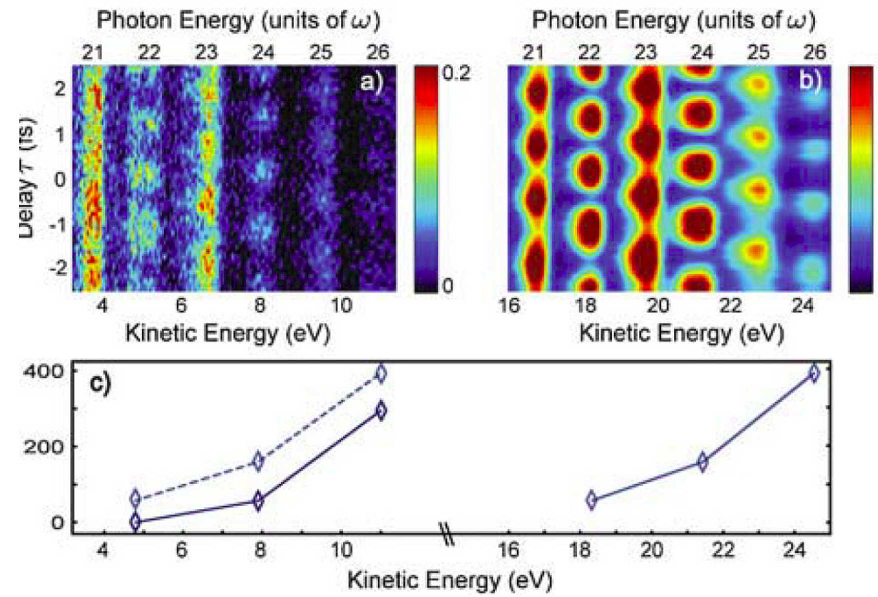
Delay in photoemission

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

τ_A = group delay of the attosecond pulses

τ_I = atomic delay two-color ionization

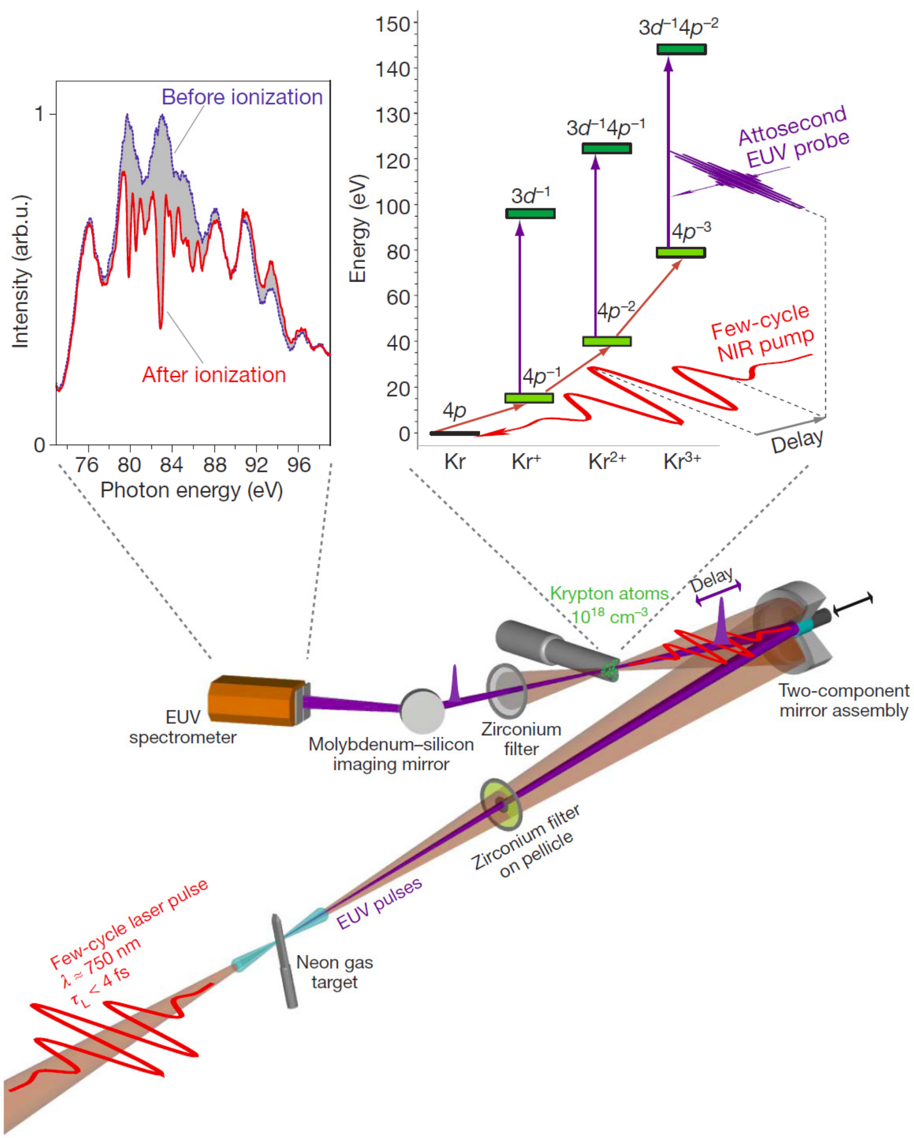




Important:

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)

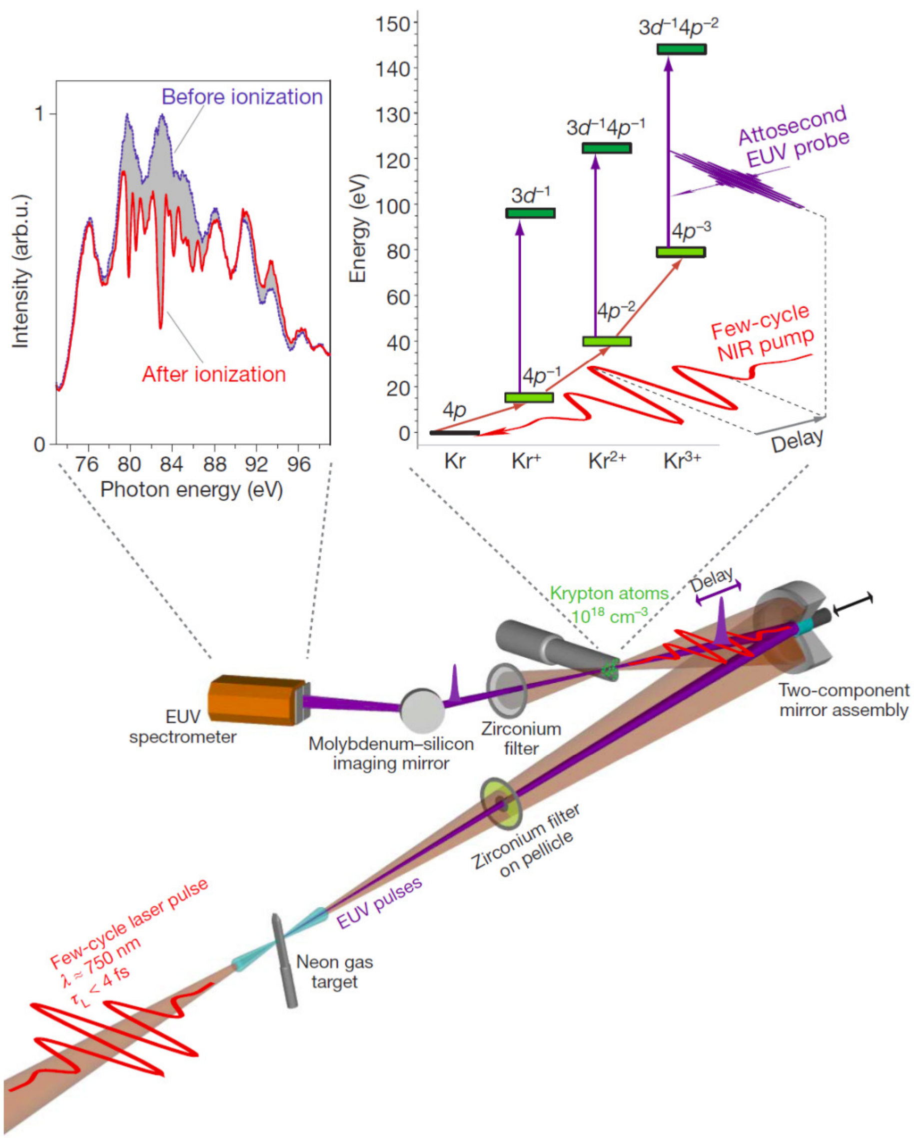


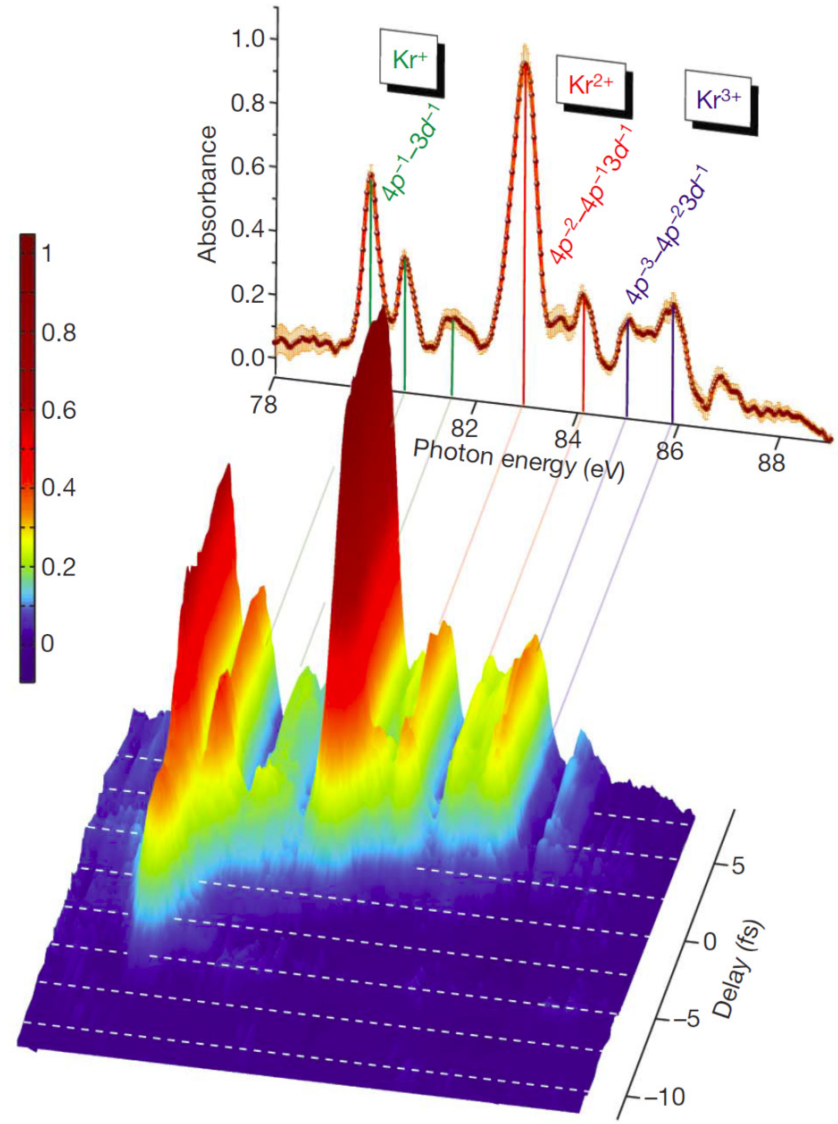
Goulielmakis et al., Nature 466, 739 (2010)

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





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

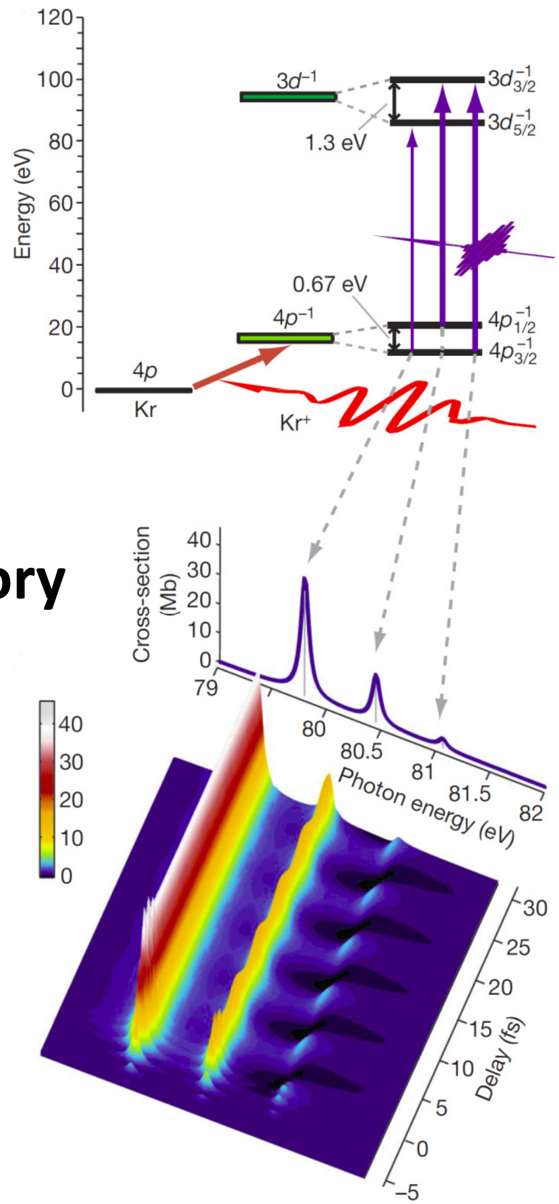
- stepsize formation of different ionic states

Observation of electronic coherence

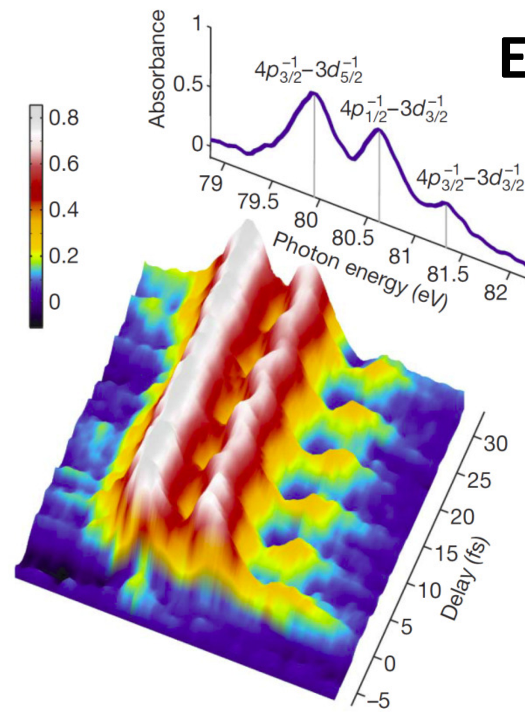
After ionization the Kr^+ ion is in a $4p_{1/2}$ or $4p_{3/2}$ state

Both configuration can be excited to a $3d_{3/2}$ state of the ion → interference

Theory

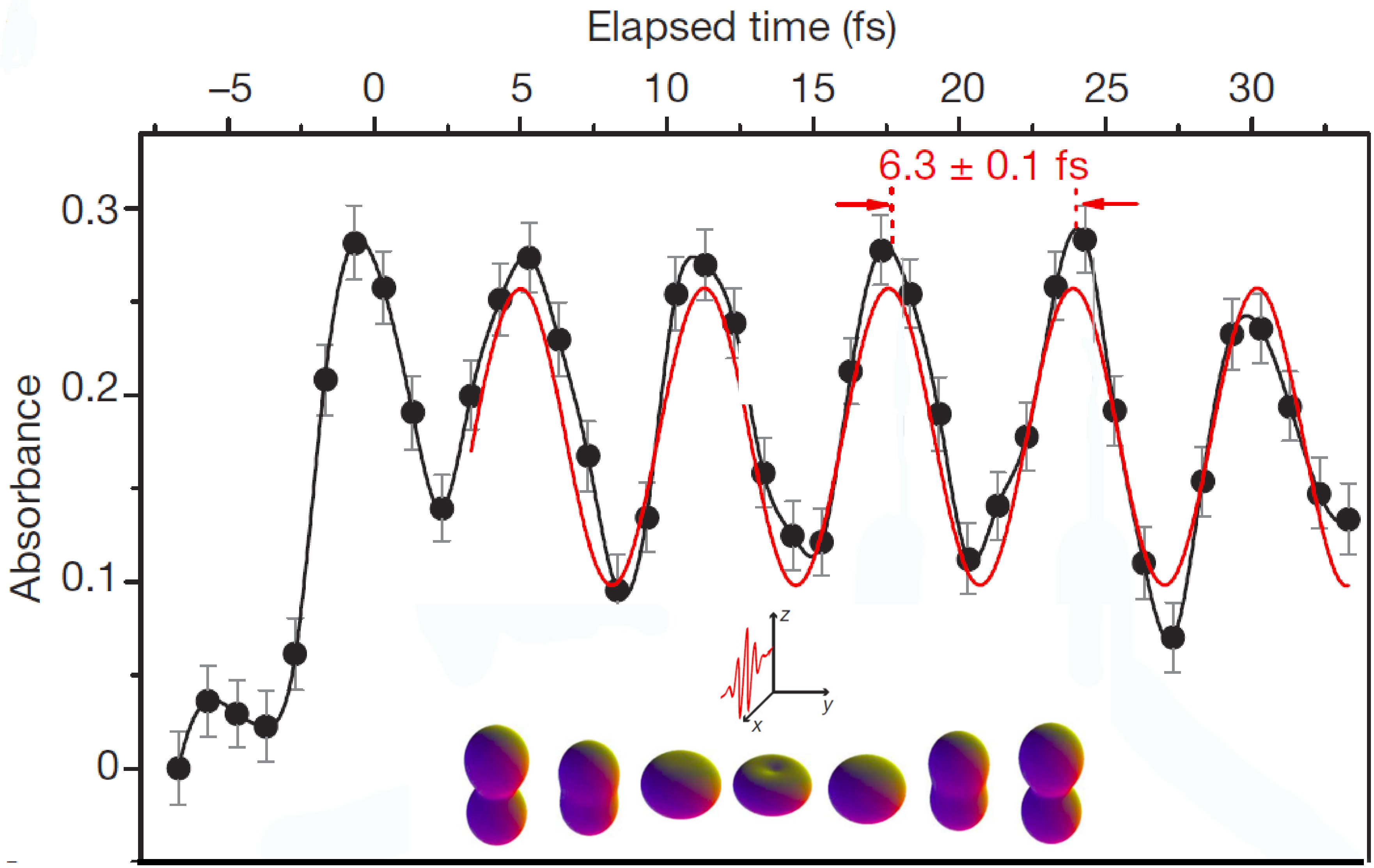


Experiment



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

Observation of electronic coherence



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

Useful materials for further reading:

P. Agostini and L. Dimauro, „The physics of attosecond light pulses", Rep. Prog. Phys. 67, 813 (2004).

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

+ several chapters in „Attosecond and XUV Physics" (ed. by M.J.J. Vrakking and Th. Schultz, Wiley, december 2013)