LUND UNIVERSITY

LLC

LUND LASER CENTRE

Photoionization dynamics on the attosecond time scale

Anne L'Huillier
Lund University
Sweden

Outline

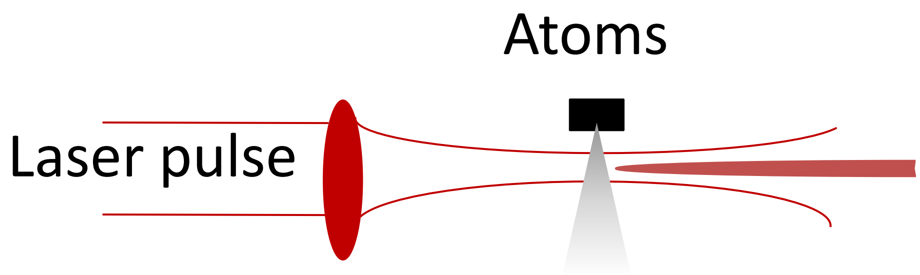
- **Extreme nonlinear optics**
  - Brief history/tutorial
  - Attosecond sources

Attosecond light sources

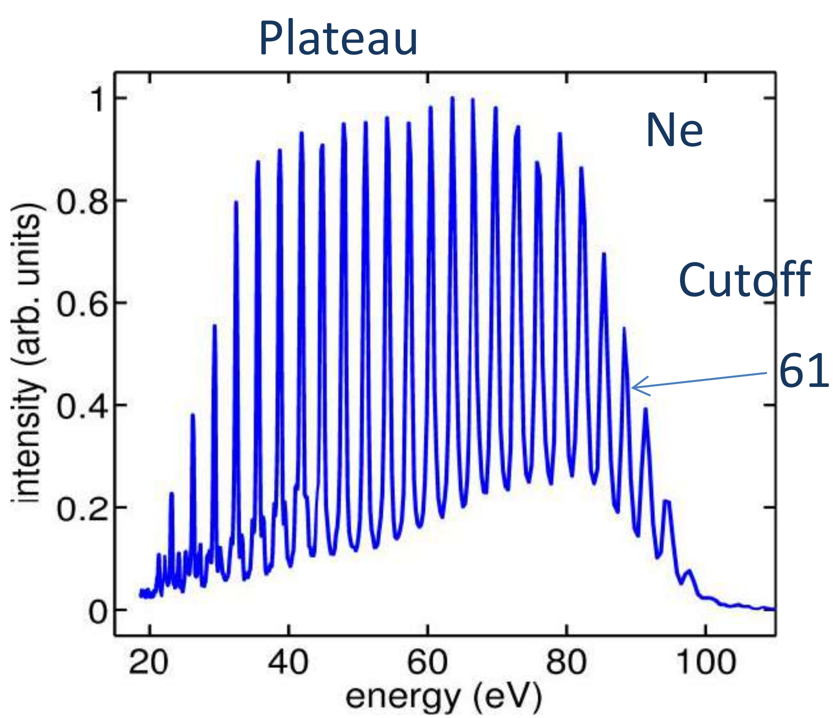
- **Ultrafast atomic physics**
  - Ionization time delays
  - Resonant photoionization

Attosecond physics

Extreme nonlinear optics

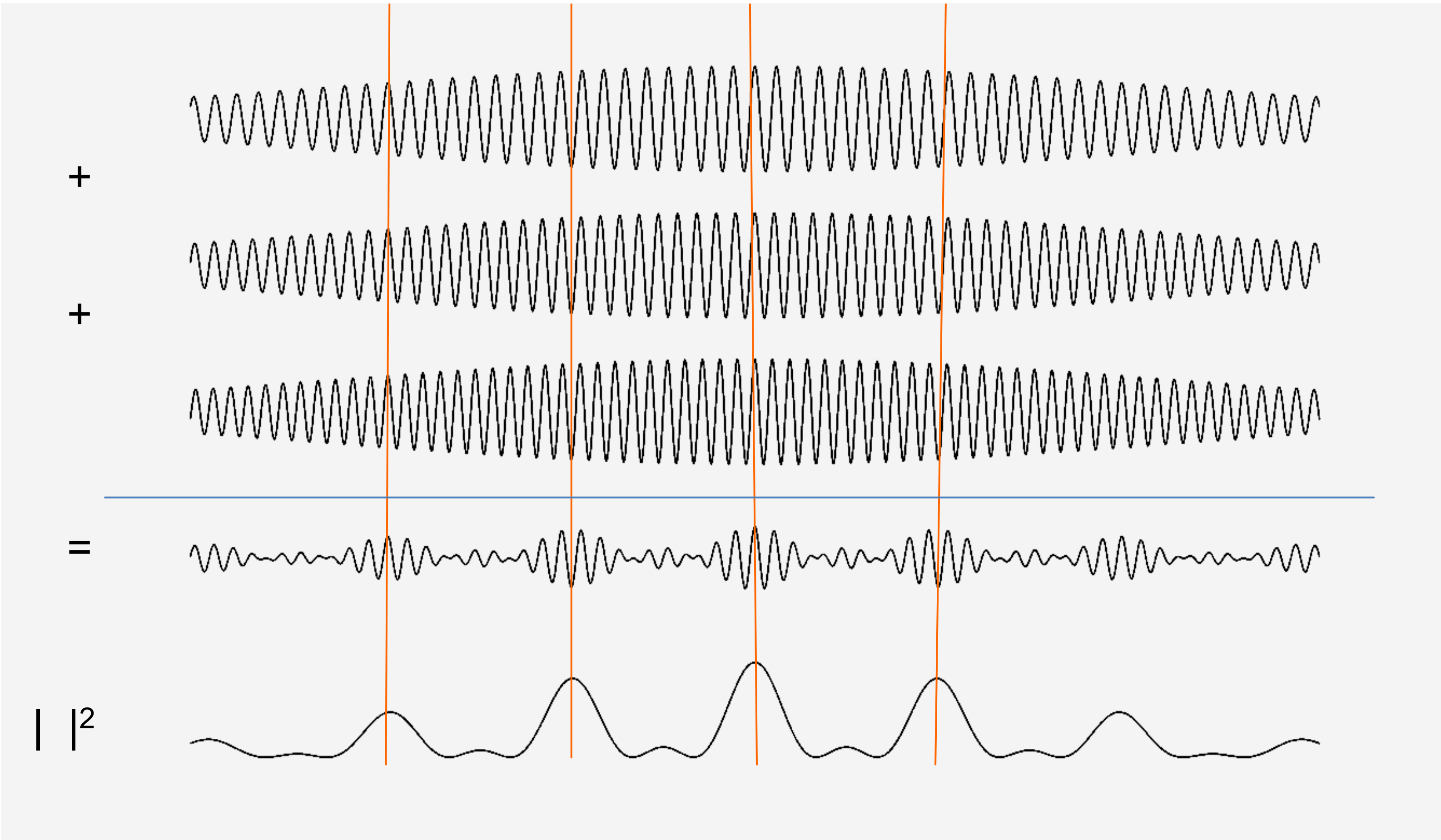


High-order harmonics



Ferray et al. J.Phys.B 1988
McPherson et al. JOSA B 1987

Attosecond pulses ?

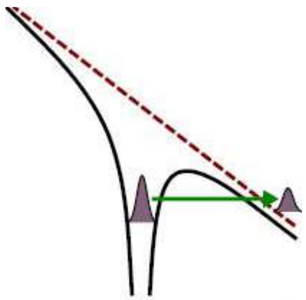


"If the harmonics are appropriately phased, this bandwidth corresponds to temporal pulses on the order of 5 X 10^{-17} s, and thereby motivates a search for a new regime of short-pulse generation."

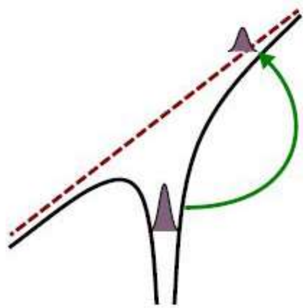
Farkas and Toth, Phys. Lett., 1992, Hänsch, Harris, Opt. Comm. 1993

Single atom response: One half cycle

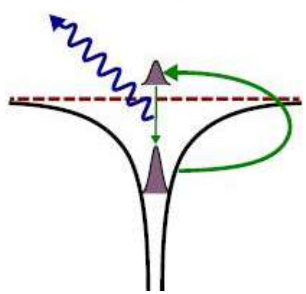
$$i\hbar\frac{\partial\Psi}{\partial t} = -\frac{\hbar^2}{2m}\nabla^2\Psi + \left[-\frac{e^2}{4\pi\epsilon_0 r} + eE_0\sin(\omega t)z\right]\Psi$$



Tunneling

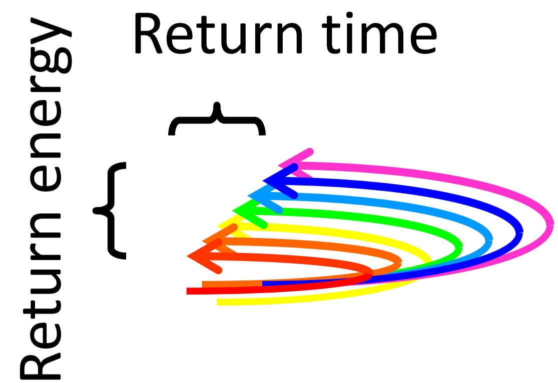


Acceleration

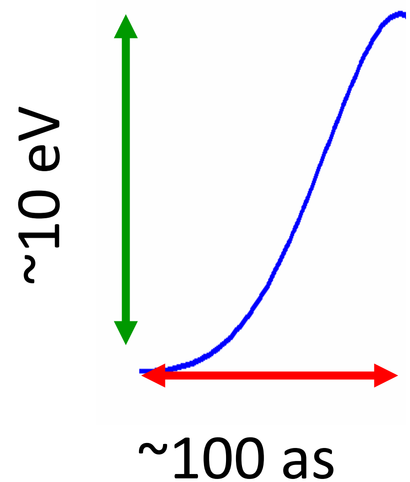


Return and recombination

Photon emission



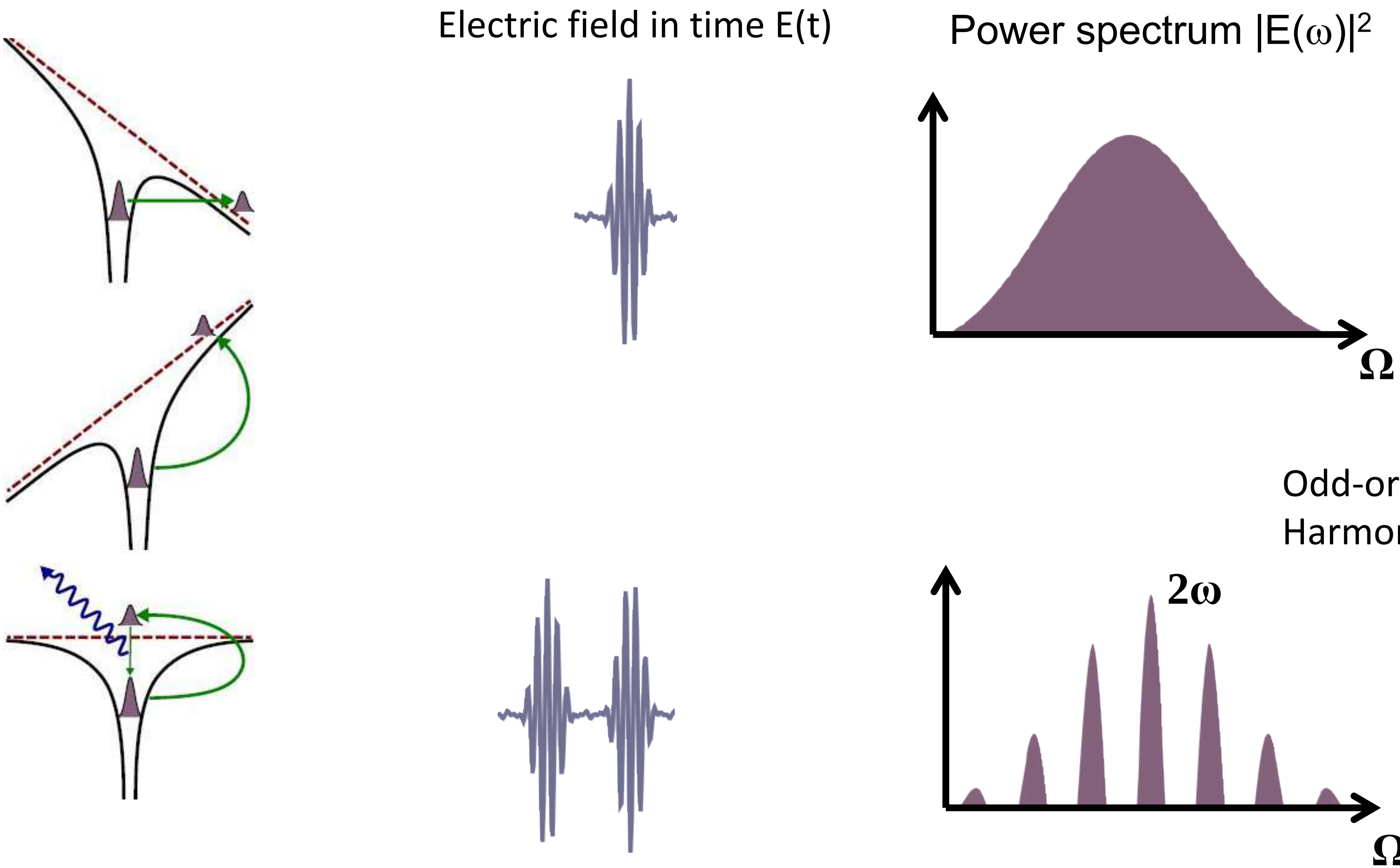
Attosecond electronic wavepacket



Attosecond pulse

Schafer, Kulander PRL 1993, Corkum PRL, 1993, Lewenstein et al. PRA 1994

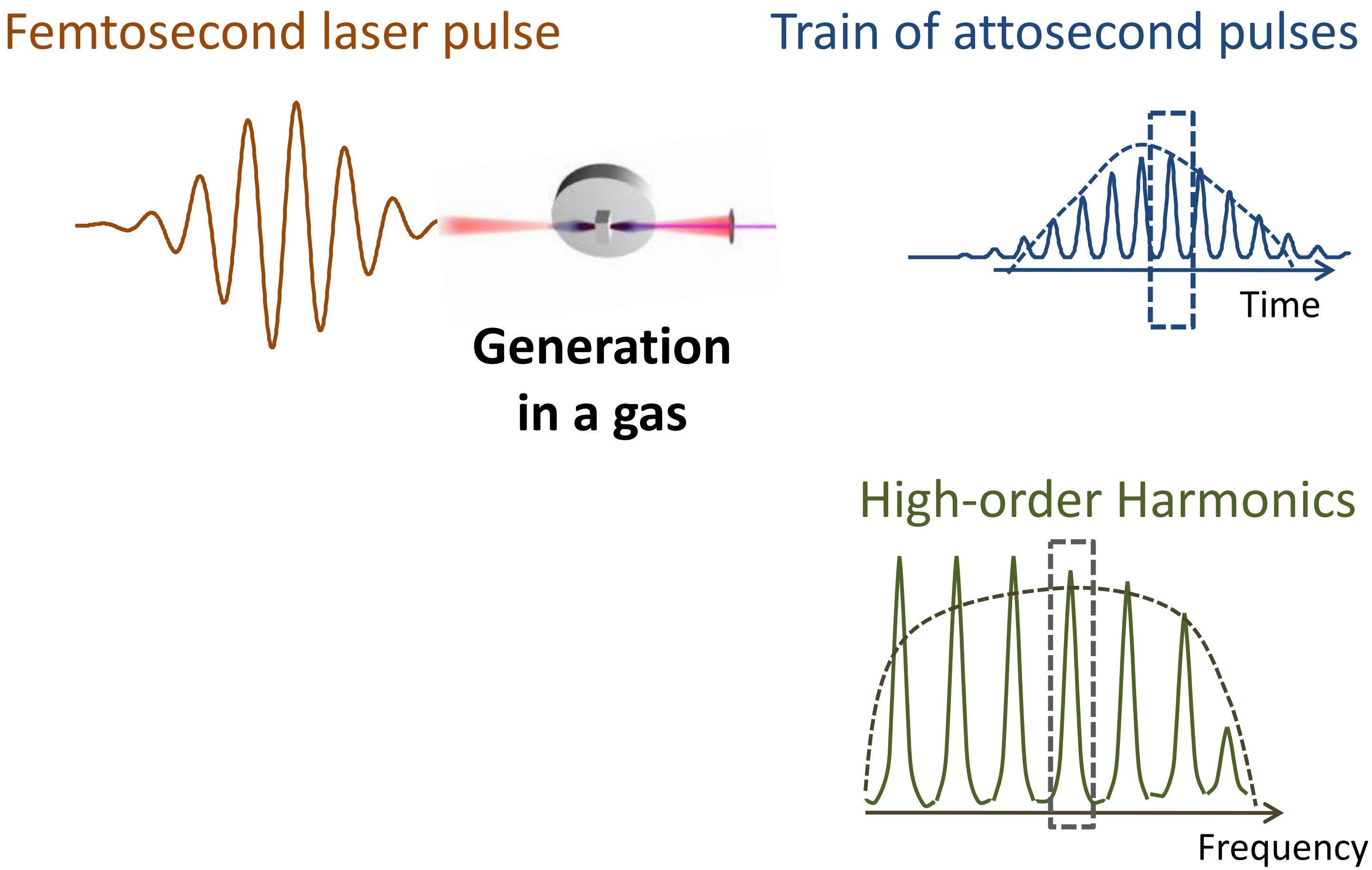
Single atom response: Several half cycles



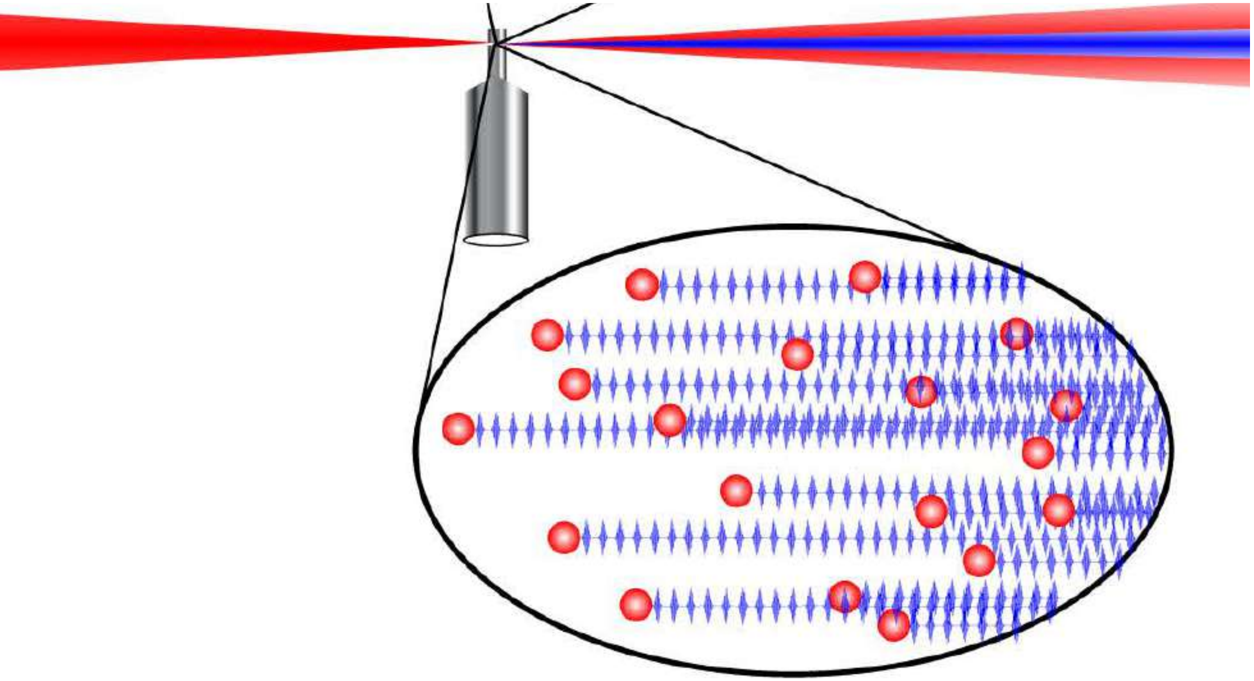
Harmonics = Interferences of attosecond pulses

Attosecond pulses = phase locked harmonics

Light sources based upon high-order harmonic generation in gases



Nonlinear Optics



$$\sum \delta\varphi \approx 0$$

Phase matching

Phase velocity of the fundamental =
Phase velocity of the harmonic fields

Degree of ionization = a few %

Gaarde et al., J. Phys. B, 2002, Heyl et al., J. Phys. B, 2017

Attosecond Pulses

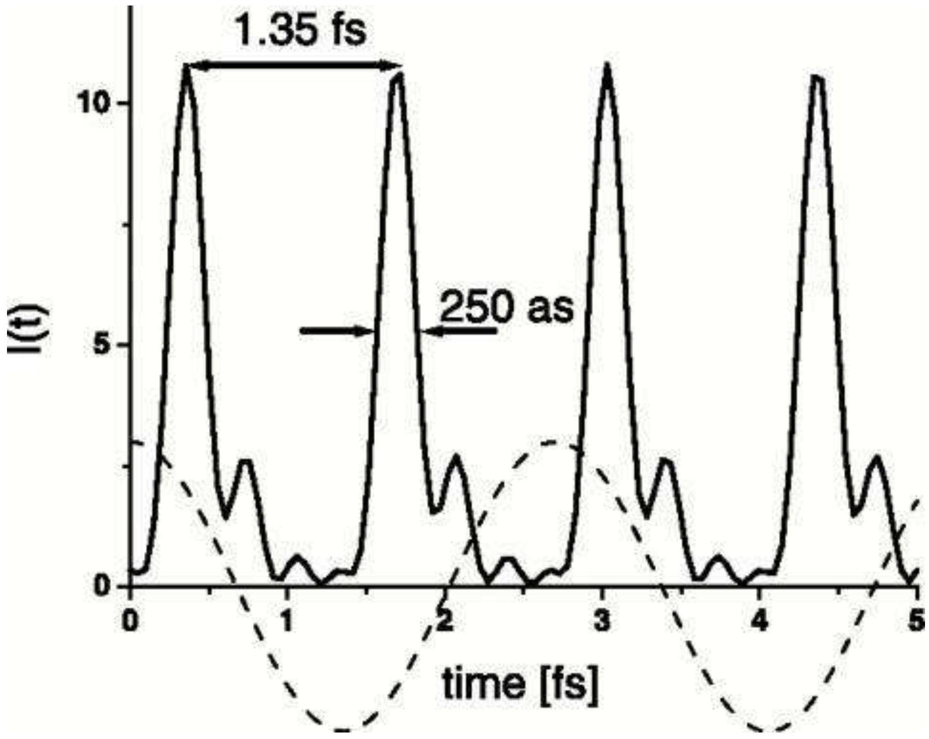
Paris, 2001, 250 as

Vienna, 2001, 450 as

Lund, 2004, 170 as

Milan, 2006, 130 as

Munich, 2008, 80 as



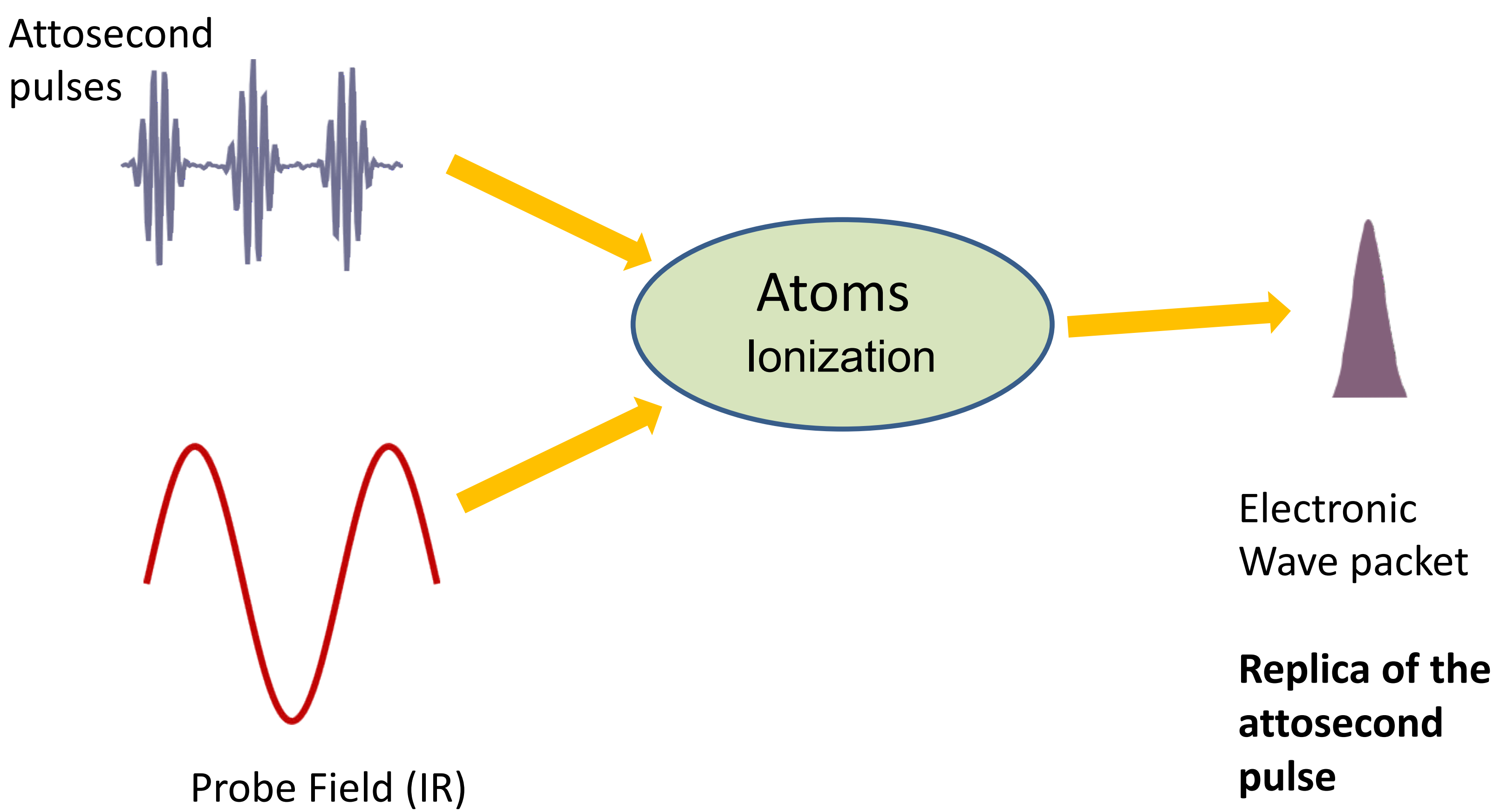
Trains of attosecond pulses – RABBIT

Single attosecond pulses – Streaking

P. Agostini

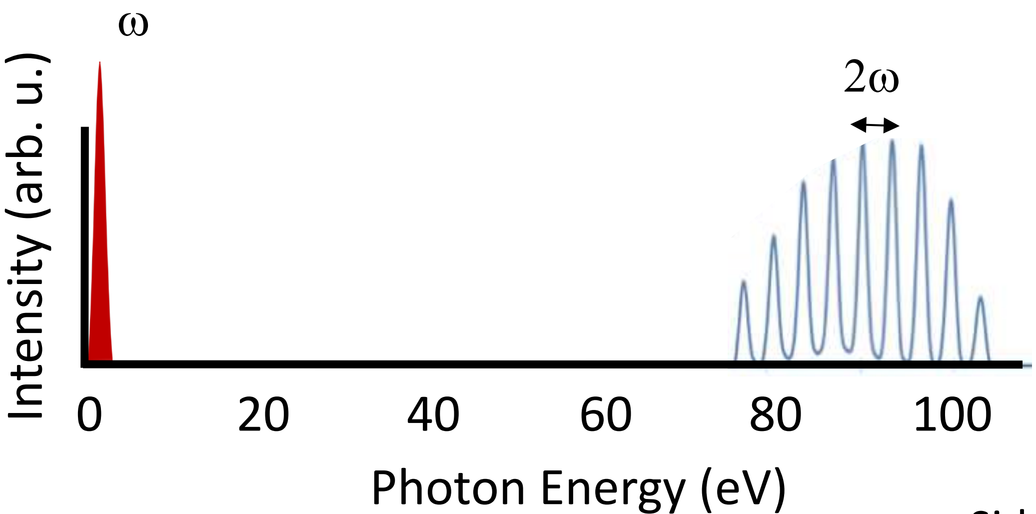
Paul et al., Science, 2001 Henstchel et al., Nature, 2001

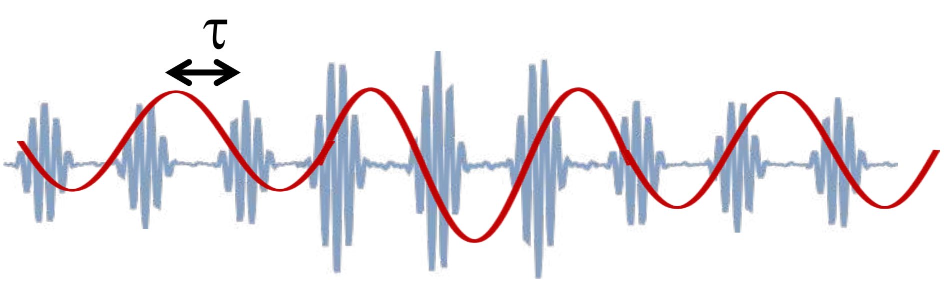
Characterization

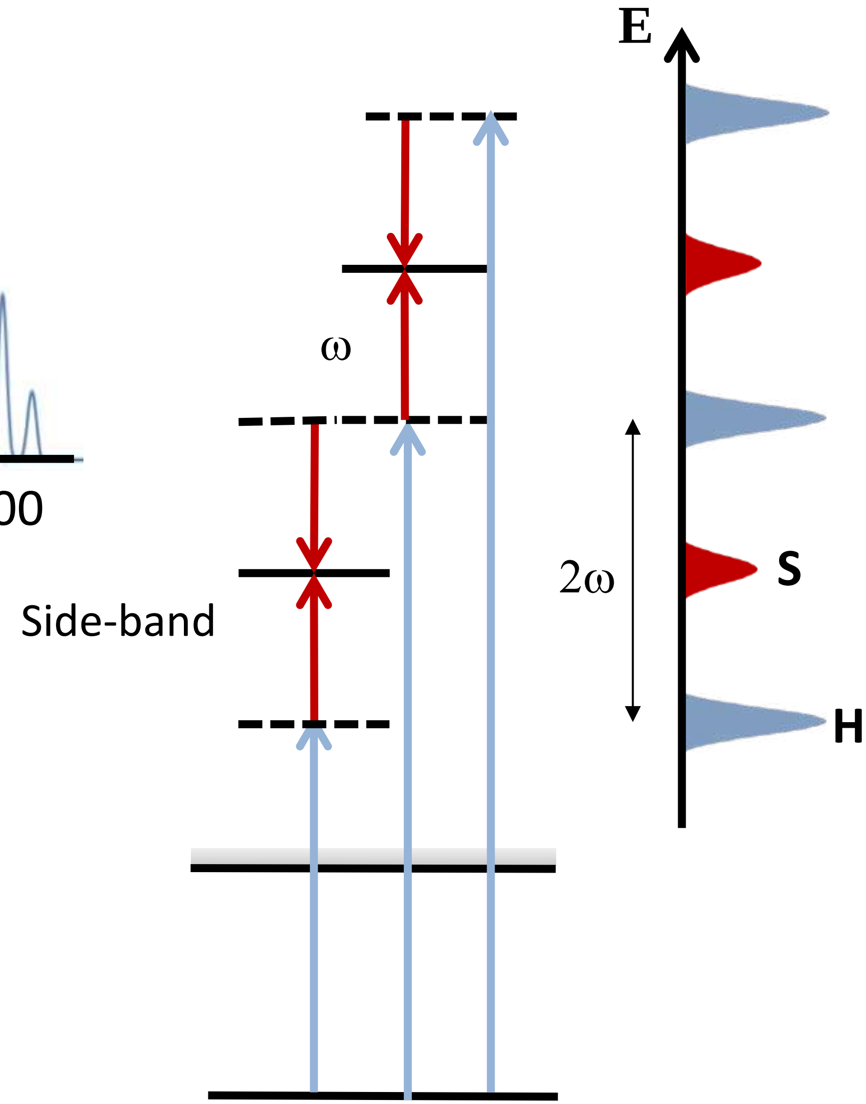


$$|E(t)|, \phi(t) \Leftrightarrow |E(\omega)|, \phi(\omega) \Leftrightarrow |E(\omega)|, \frac{d\phi}{d\omega}$$

RABBIT technique: interferometry

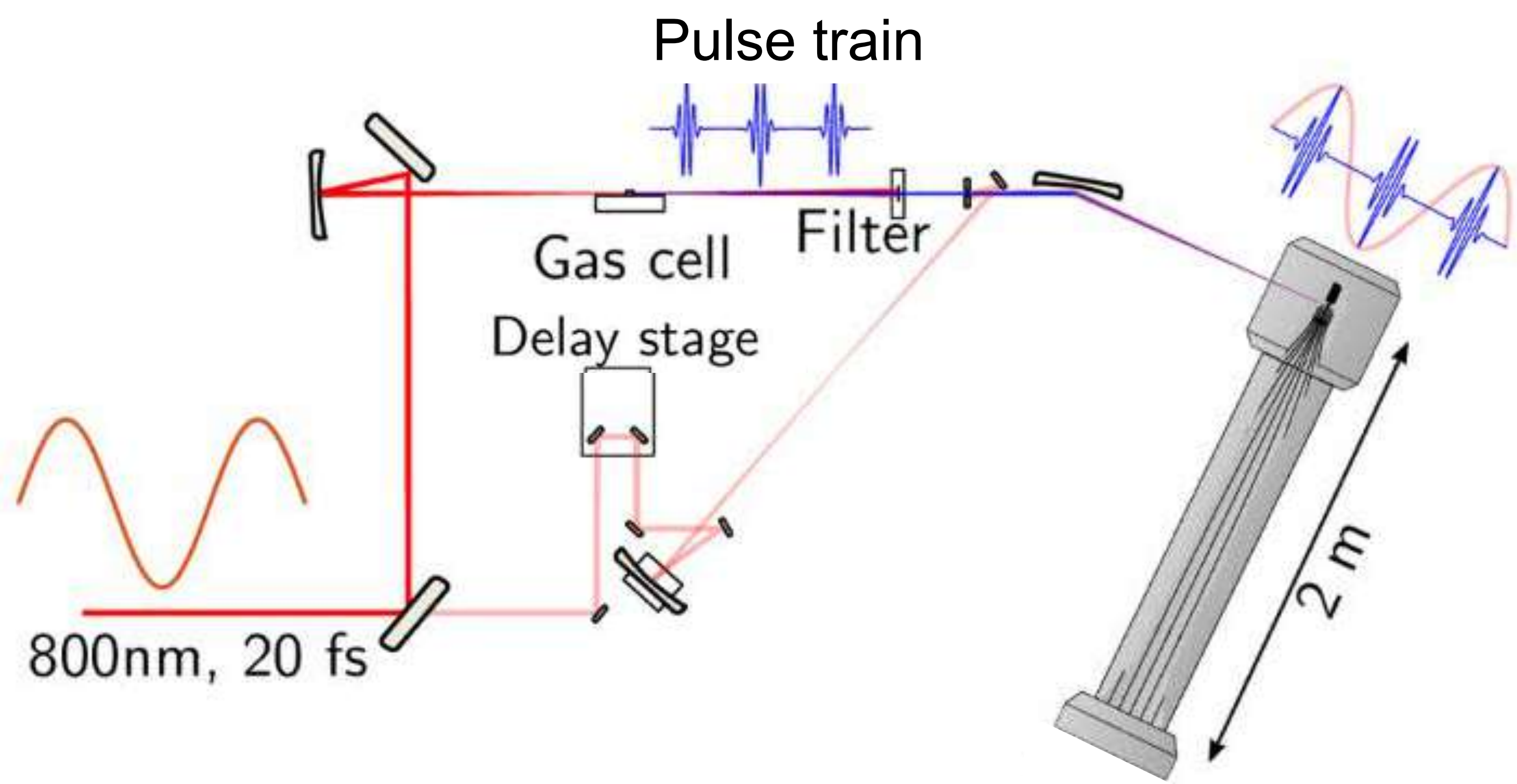






Paul et al., Science, 2001, Véniard et al., Phys. Rev. A 1996

Optical interferometry

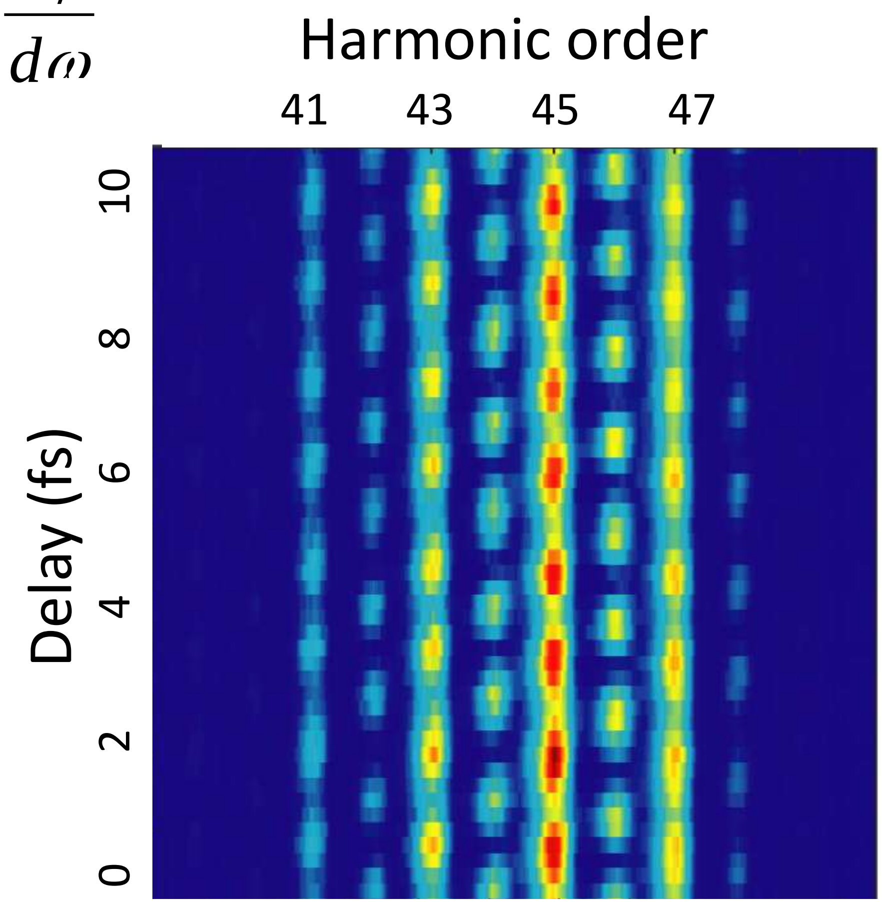


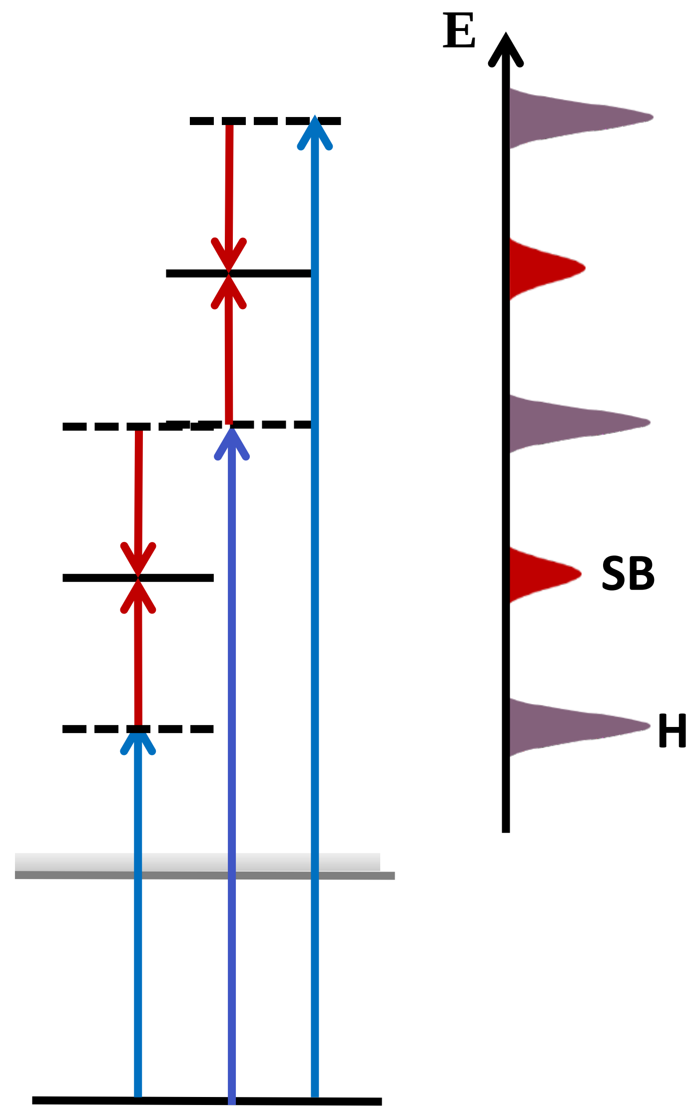
Magnetic bottle electron spectrometer

Collaboration:
R. Feifel, R. Squibb,
Gothenburg

Quantum interferometry

$$|E(\omega)|, \frac{d\phi}{d\omega}$$





$$S \propto A + B\cos(2\omega\tau - \Delta\phi)$$

$$\tau_{XUV} = \frac{\Delta\phi}{2\omega} \approx \frac{d\phi}{d\omega}$$

Paul et al., Science, 2001,
Mairesse et al., Science, 2003

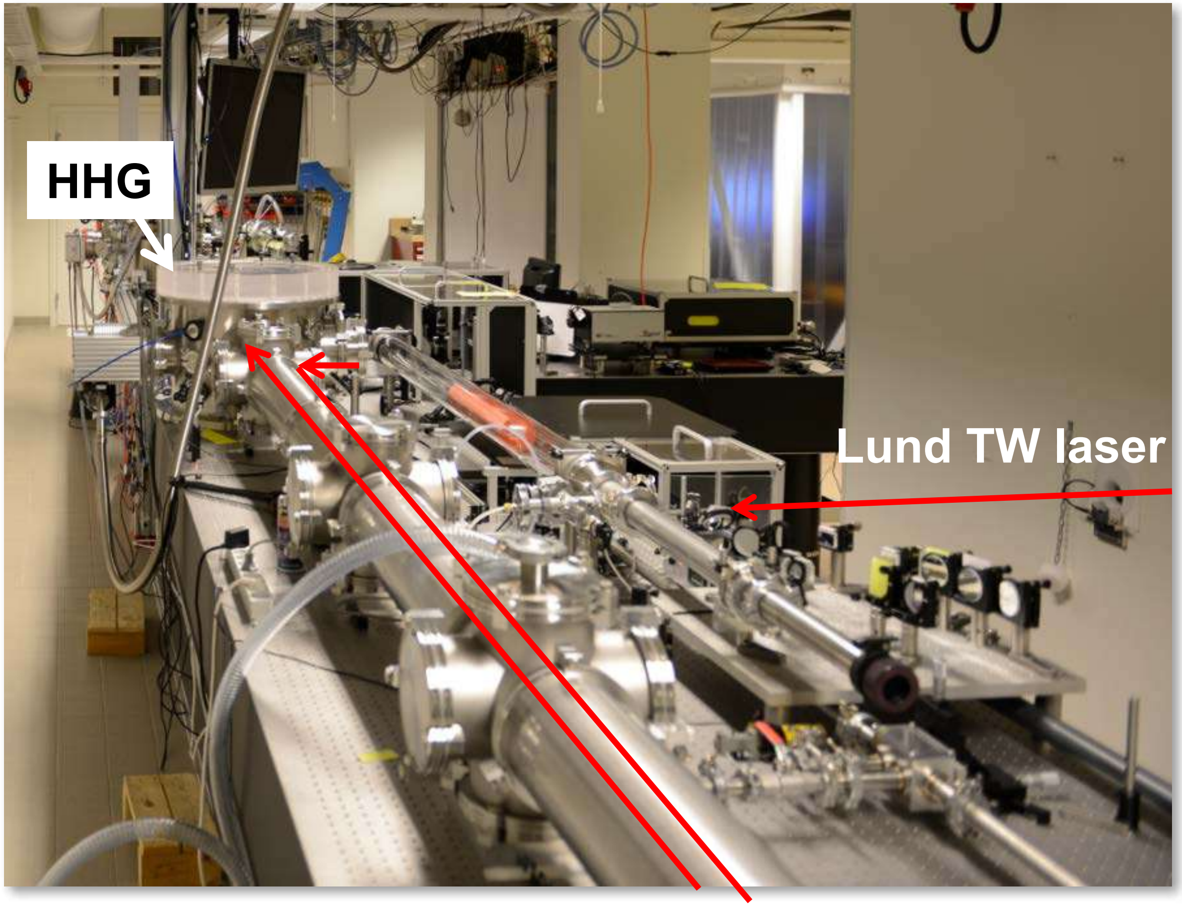
Development of attosecond sources

Application-specific

Laser	800 nm, **50 mJ**, 10 Hz- 1kHz	850 nm, 100 µJ, **200 kHz**	**2-4 µm**, 0.1– 1 mJ 1-100 kHz
Attosecond source	**Intense** pulses (**µJ**) 20-50 eV	**High-repetition rate** pulses 20-100 eV	**X-ray** attosecond pulses **100-1000 eV**
Applications	XUV-XUV pump-probe Nonlinear phenomena	Surface science Coincidence measurements	Ultrafast X-ray science
	Geometrical Scaling (up)	Geometrical Scaling (down)	Wavelength Scaling

Heyl et al., Optica, 2016

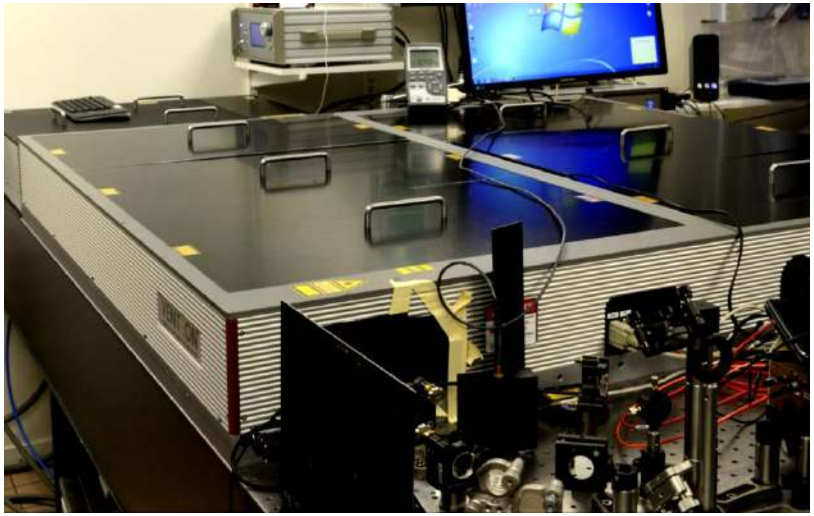
Intense attosecond source



Laser	**Ti:Sapphire, 800 nm, 50 mJ, 35 fs, 10 Hz**
Atto-second source	Intense pulses for XUV-XUV pump-probe 17-50 eV
Appli-cations	Nonlinear phenomena

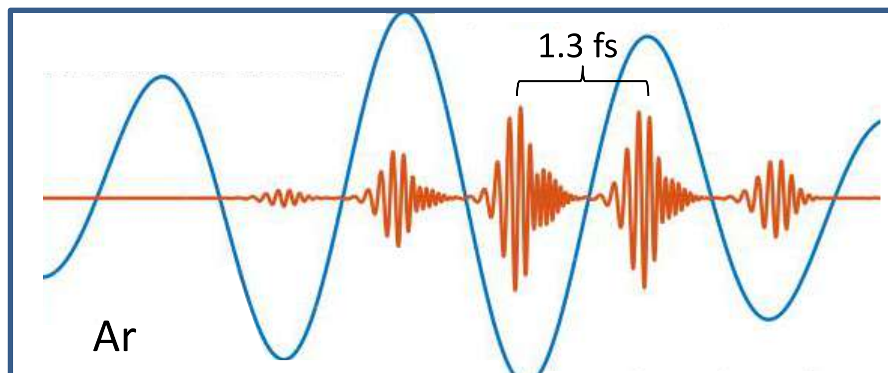
Manschwetus et al., Phys. Rev. A, 2016

High repetition rate attosecond source



Optical Parametric Amplification

Laser	**OPCPA, 850 nm, 5 μJ, 7 fs, 200 kHz, CEP-stable**
Atto-second source	Few attosecond pulses @ 17-50 eV
Application	Surface and AMO science



Harth et al., J. Opt., 2017, Guo et al., J. Phys. B, 2017

Development of attosecond sources

Laser	800 nm, 50 mJ, 10 fs, 10 Hz- 1kHz	850 nm, 100 µJ, 7 fs, 200 kHz	2-4 µm, 0.1– 1 mJ 30 fs, 1-100 kHz
Attosecond source	**Intense** pulses (µJ) 20-50 eV	**High-repetition rate** pulses 20-100 eV	**X-ray** attosecond pulses 20-1000 eV
Applications	XUV-XUV pump-probe Nonlinear phenomena	Surface science Coincidence measurements	Ultrafast X-ray science

Sheehy et al., PRL, 1999, Popmintchev et al. Nat. Phot. 2012

La Physique Attoseconde en France

R. Lopez-Martens
S. Haessler

P. Salières
T. Ruchon
H. Merdji

S. Kazamias

R. Taïeb
J. Caillat
A. Maquet

Bordeaux
E. Cormier
E. Mevel
Y. Mairesse

Lyon
F. Lépine
E. Constant

Strasbourg
J.-Y. Bigot

B. Carré

Outline

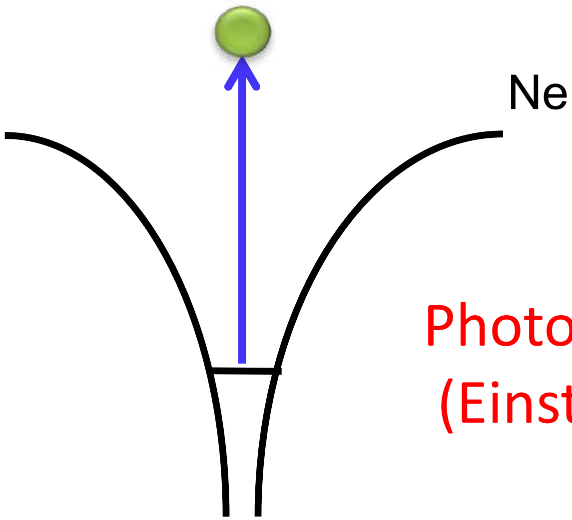
- **Extreme nonlinear optics**
  - Brief history/tutorial
  - Attosecond sources

Attosecond light sources

- **Ultrafast atomic physics**
  - Ionization time delays
  - Resonant photoionization

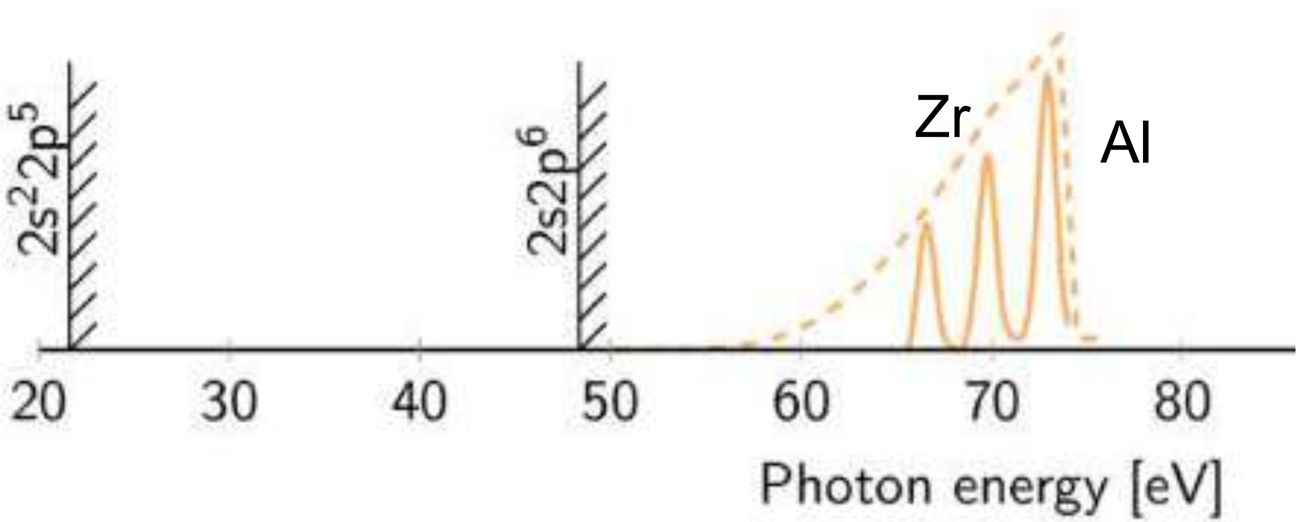
Attosecond physics

Photoionization of neon



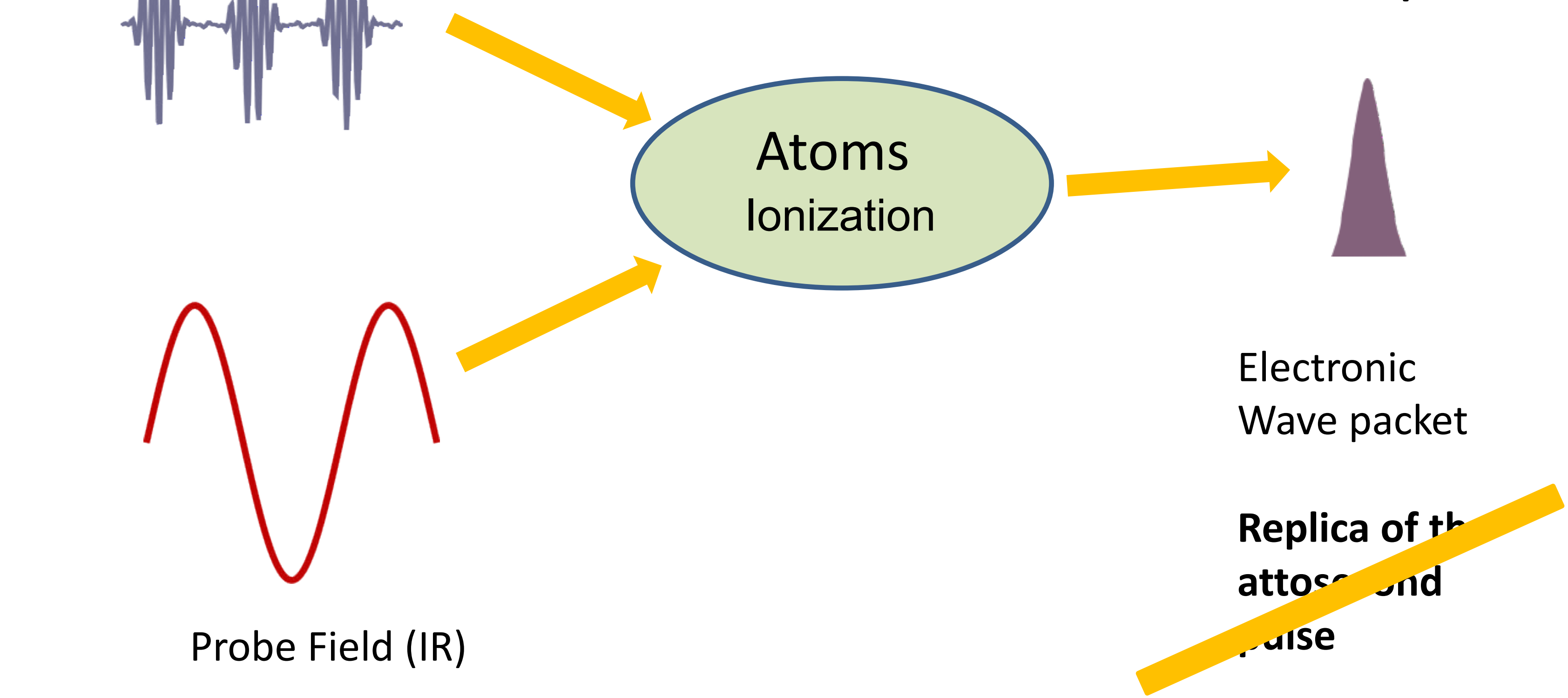
Ne $1s^2 2s^2 2p^6$

Photoelectric effect
(Einstein) 1905



Characterization

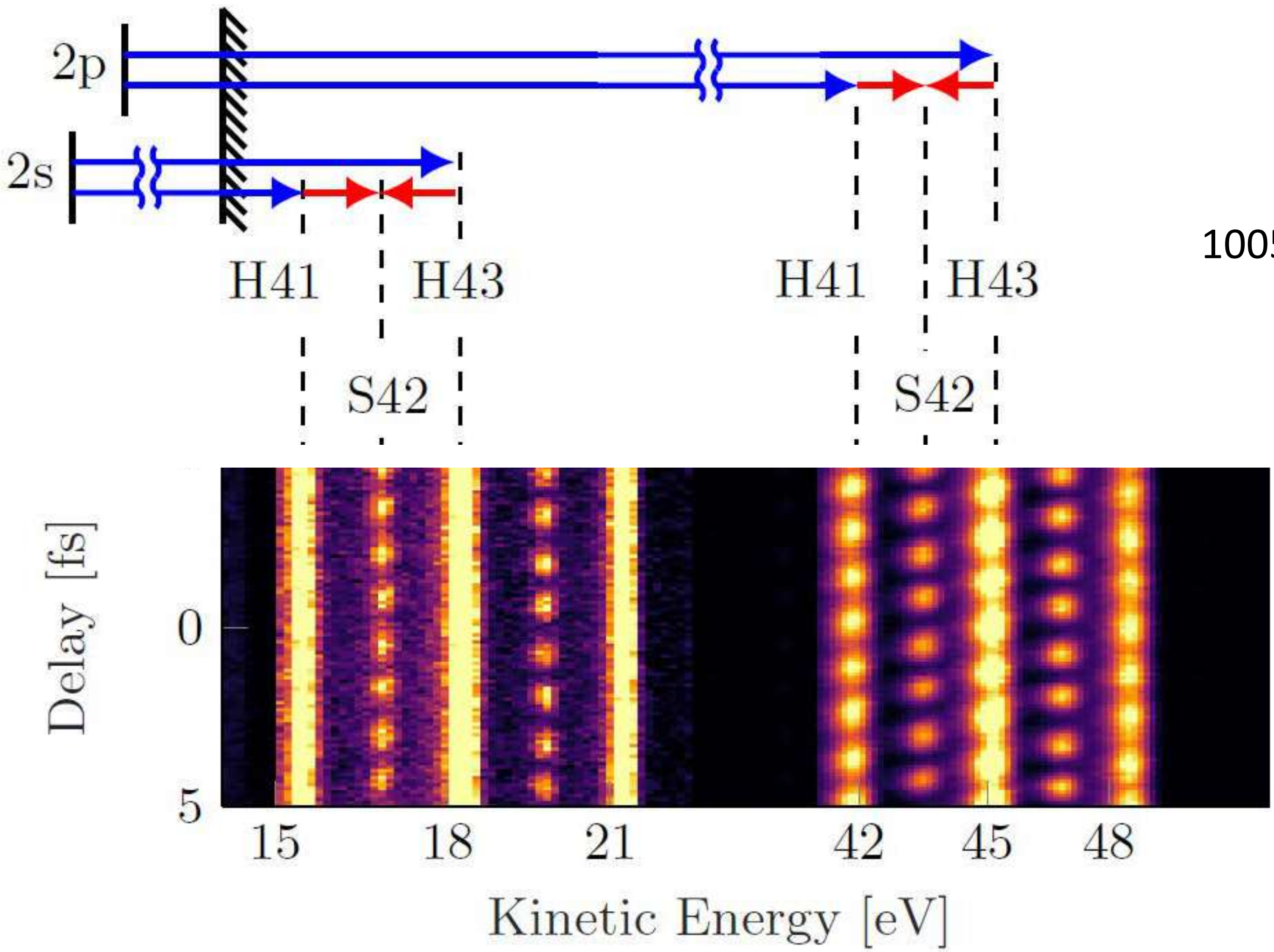
of the attosecond pulses and the ionization process

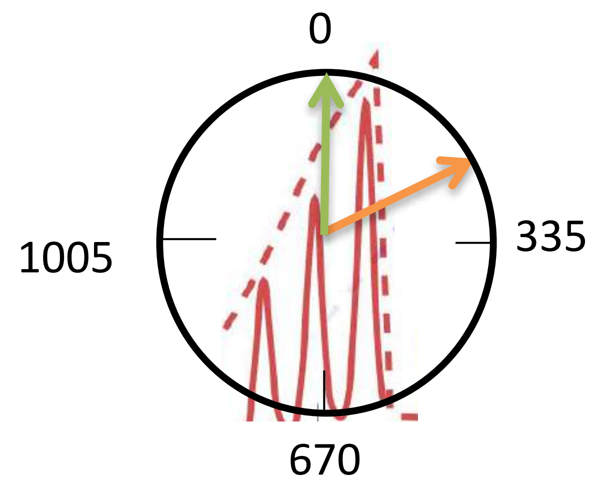


$$S \propto A + B\cos(2\omega\tau - \Delta\phi)$$

$$\frac{\Delta\phi}{2\omega} = \tau_{XUV}$$

Single ionization of Ne in 2s and 2p shells

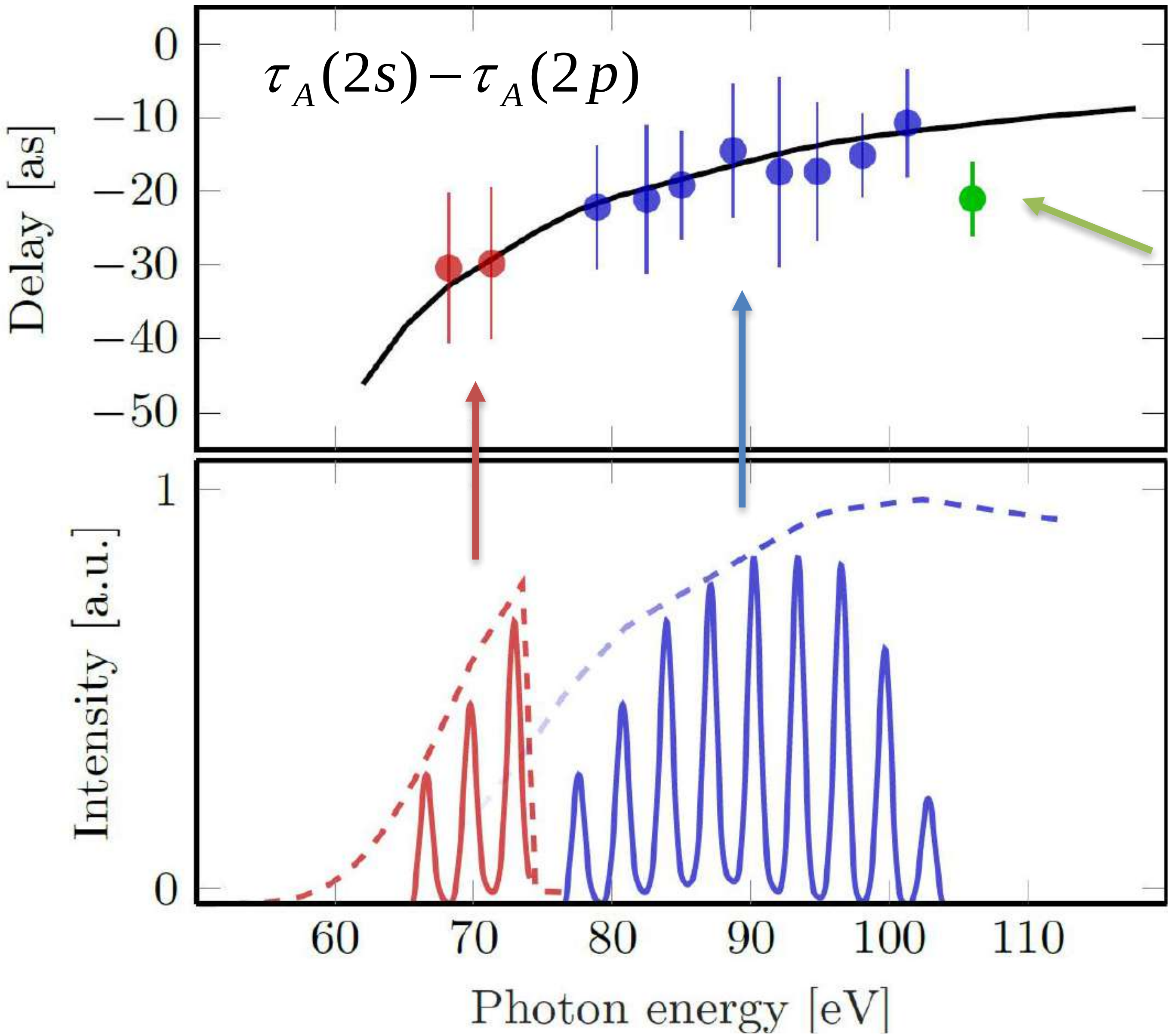




$$\tau_A(2s) - \tau_A(2p)$$

$$\frac{\Delta\phi}{2\omega} = \tau_{XUV} + \tau_A(2s) \qquad \frac{\Delta\phi}{2\omega} = \tau_{XUV} + \tau_A(2p)$$

Single ionization of Ne in 2s and 2p shells



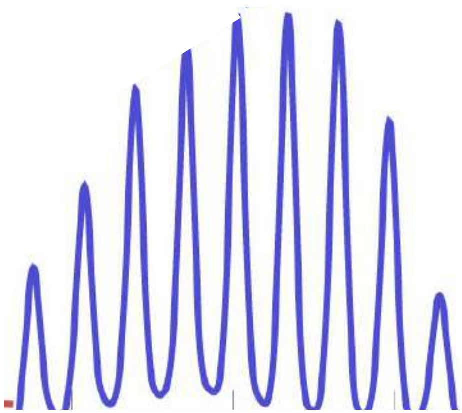
Theory

Schultze et al., 2010
Streaking

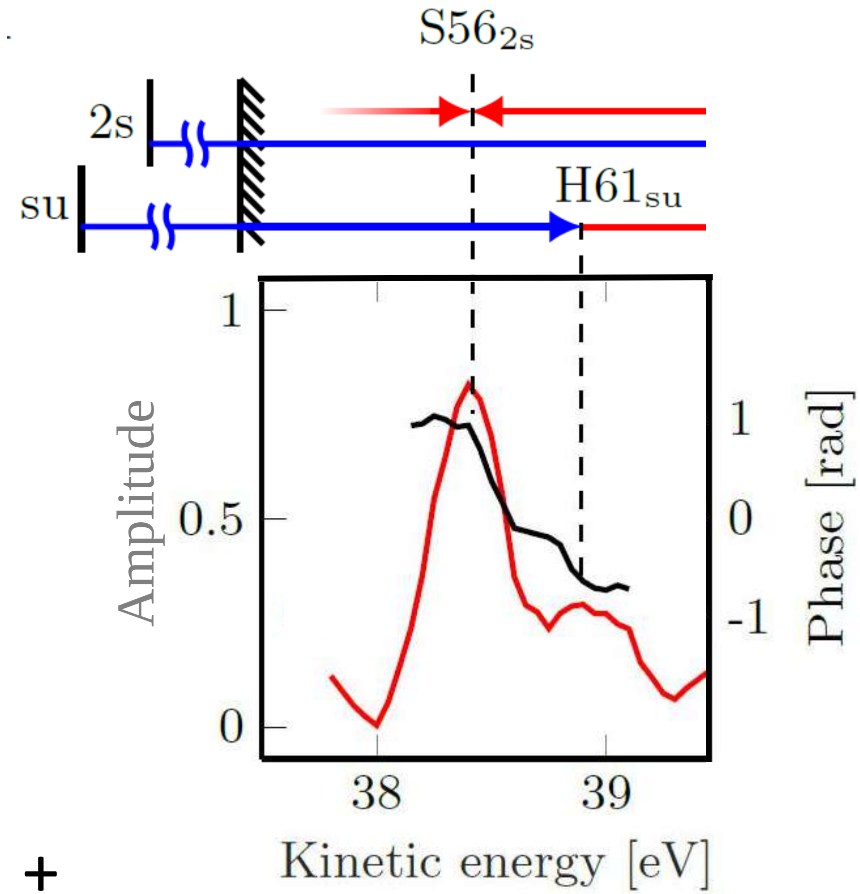
A seven year old puzzle

Isinger et al., Science, 2017

High spectral resolution



- Harmonic bandwidth 200 meV



- Spectrometer resolution 100 meV

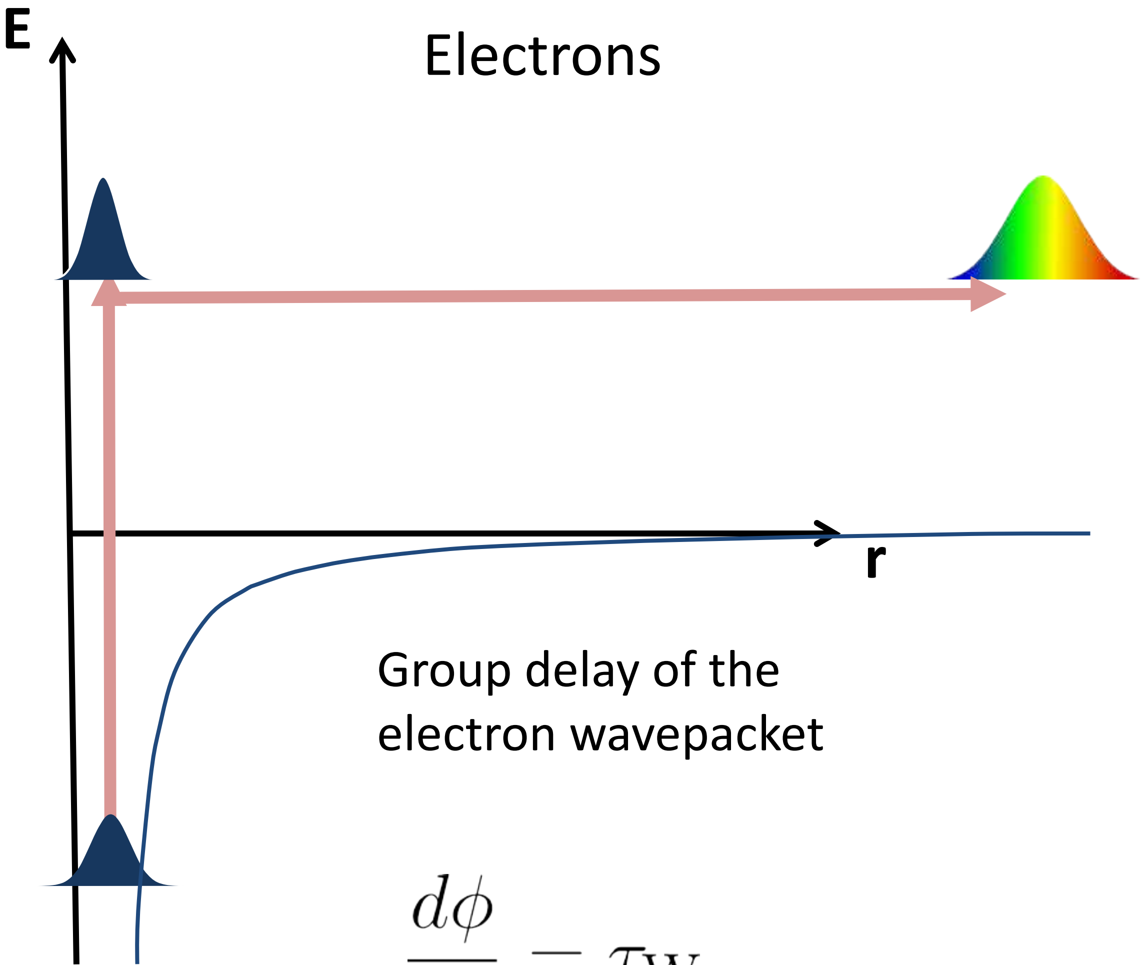
su = shake up
2p ionization +
2p ⟶ 3p

High temporal (20 as) and spectral resolution (200 meV) !

Gruson et al., Science, 2016 Isinger et al., Science, 2017

Interpretation of the atomic delay

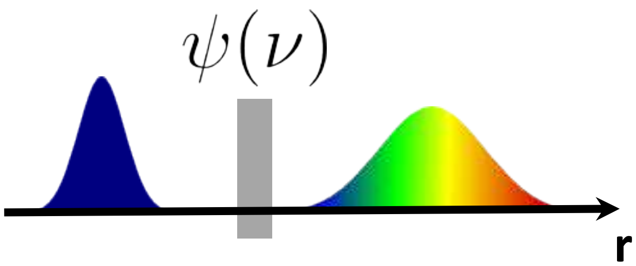
Electrons



Group delay of the electron wavepacket

$$\frac{d\phi}{d\omega} = \tau_{\mathrm{W}}$$

Photons

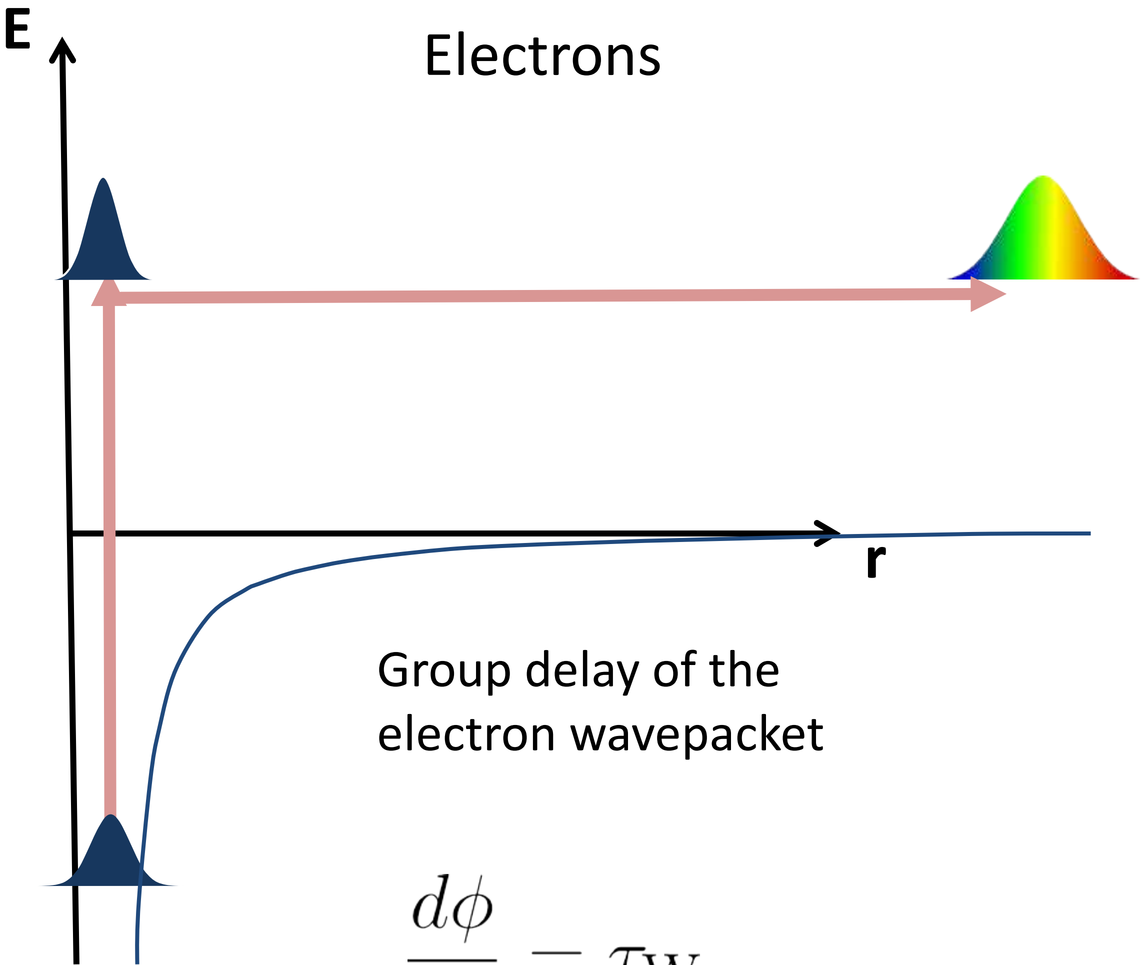


Group delay of light pulse

$$\frac{d\psi}{d\nu} = \tau_{\mathrm{GD}}$$

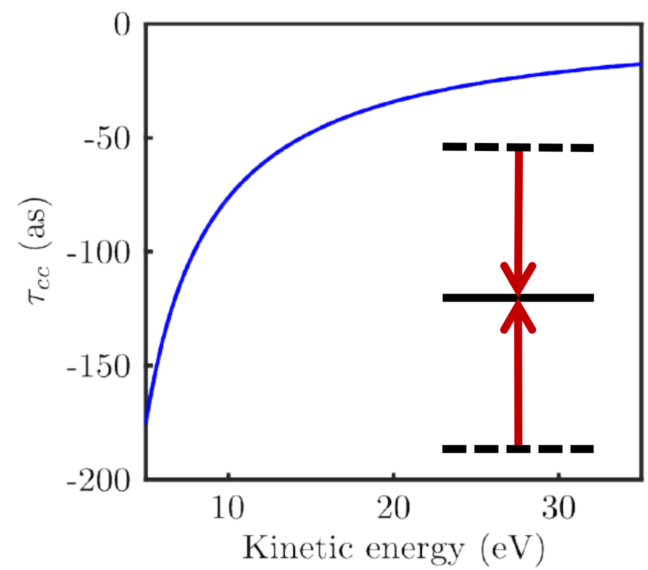
E. P. Wigner, Phys. Rev. 1955

Interpretation of the atomic delay



Group delay of the electron wavepacket

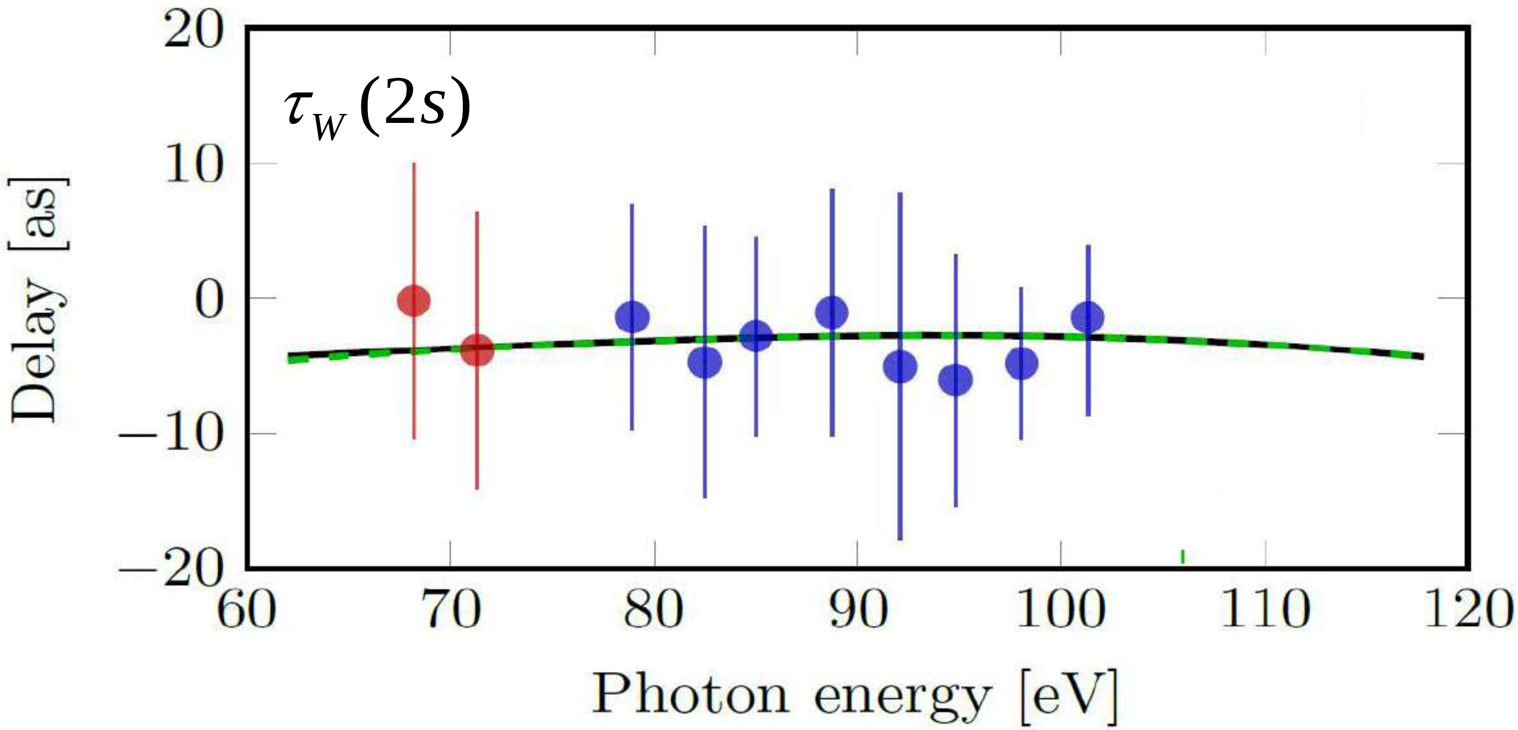
$$\frac{d\phi}{d\omega} = \tau_{\mathrm{W}}$$



Delay induced by the probe field τ_{cc}

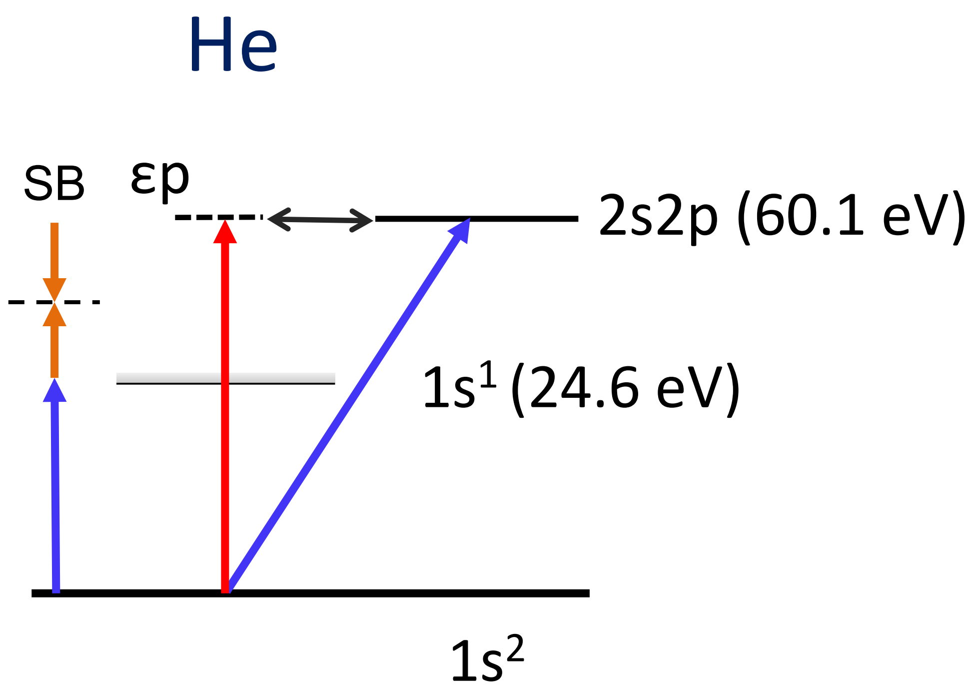
$$\tau_{\mathrm{A}} = \tau_{\mathrm{W}} + \tau_{\mathrm{cc}}$$

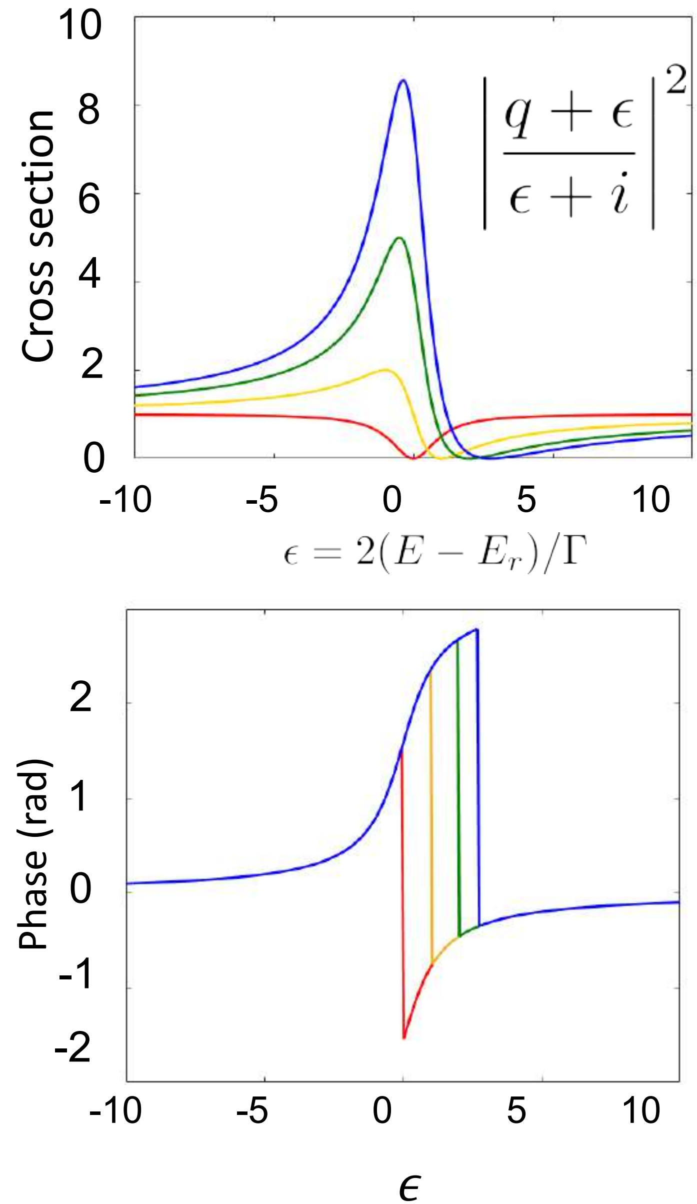
“Absolute” Photoionization time delays



Ossiander et al., Science, 2016

Fano resonance

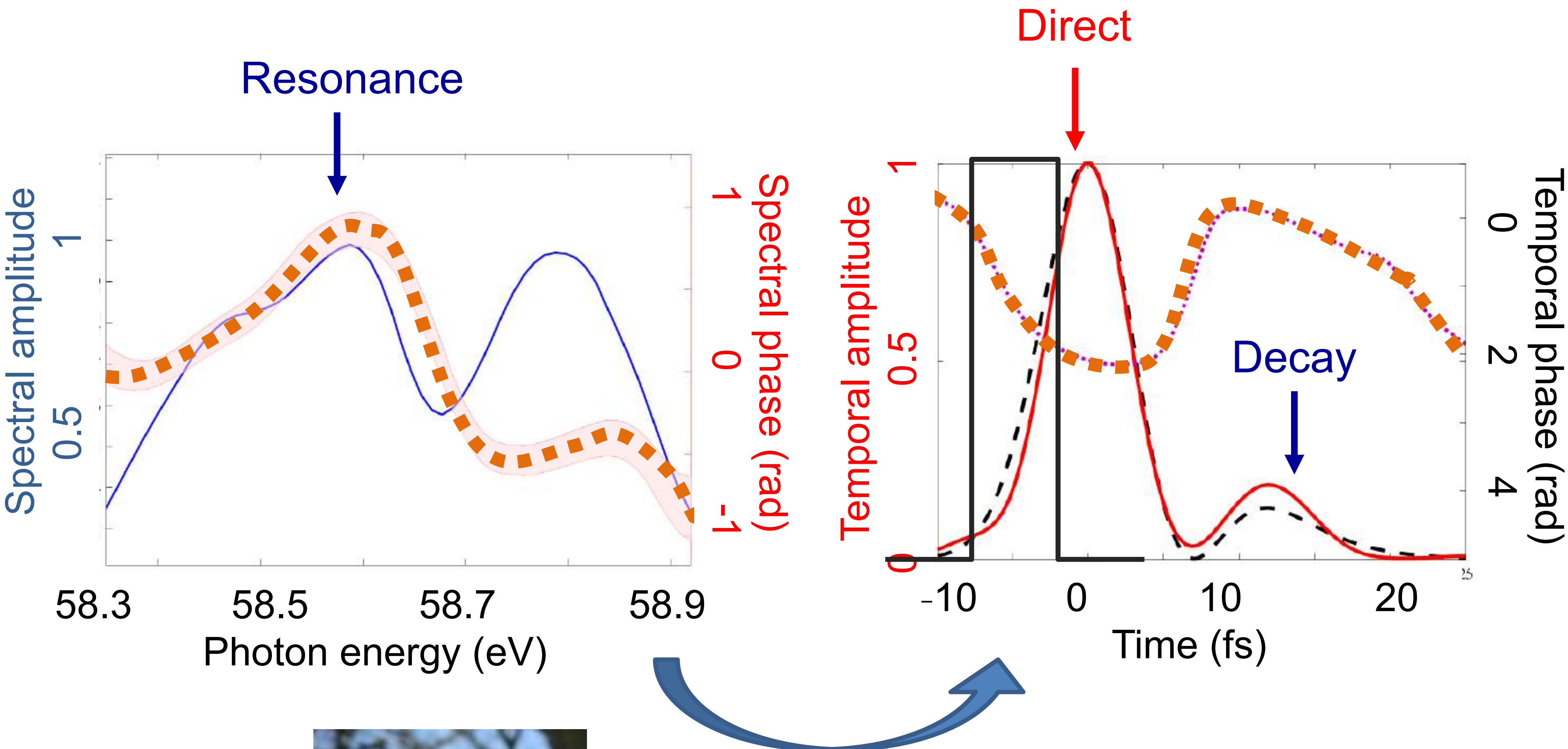




- $q = 0$
- $q = -1$
- $q = -2$
- $q = -2.75$

Fano, Phys. Rev. 1961
Drescher et al., Nature, 2002
Ott et al., Science, 2013

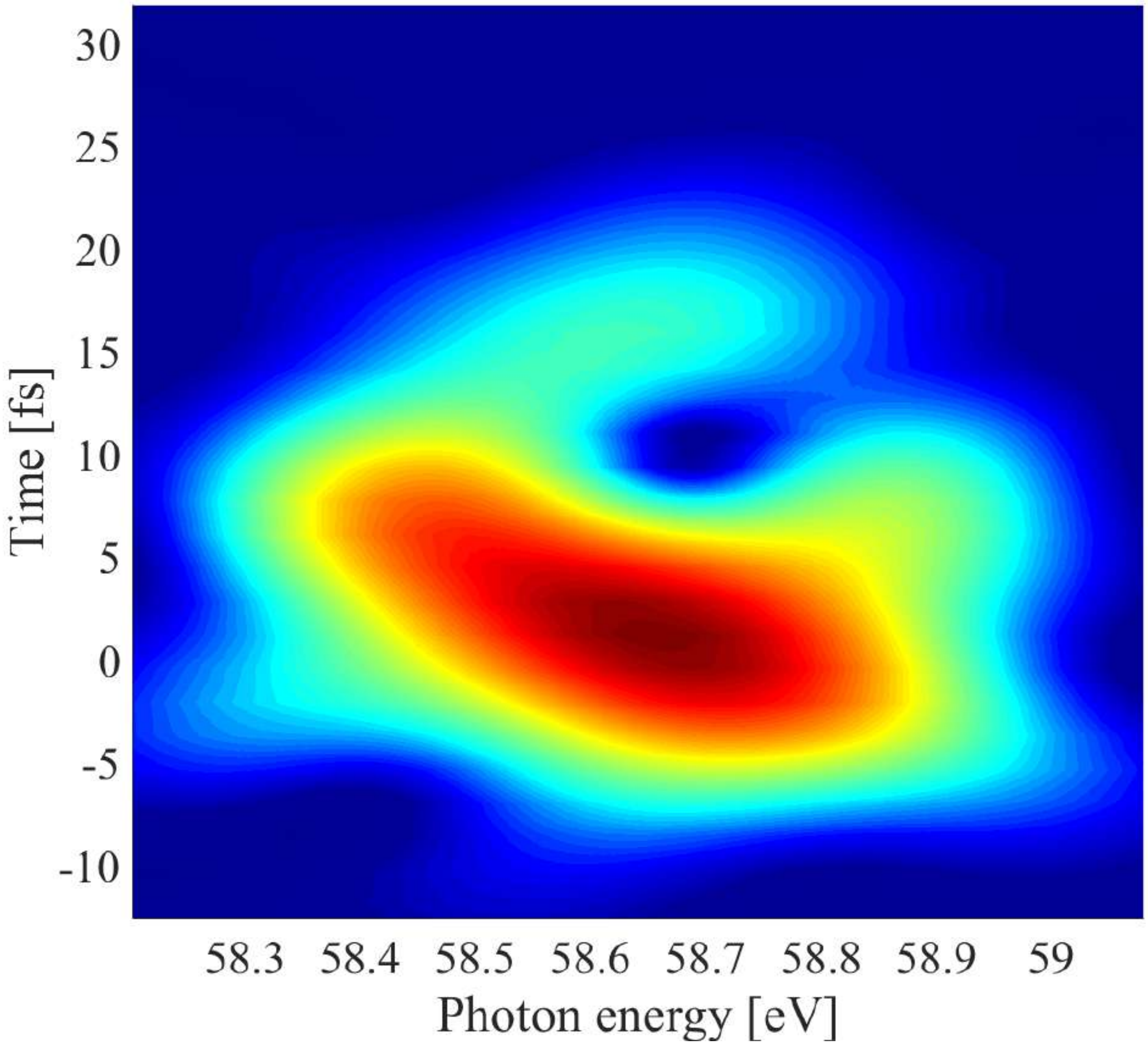
Ultrafast atomic physics

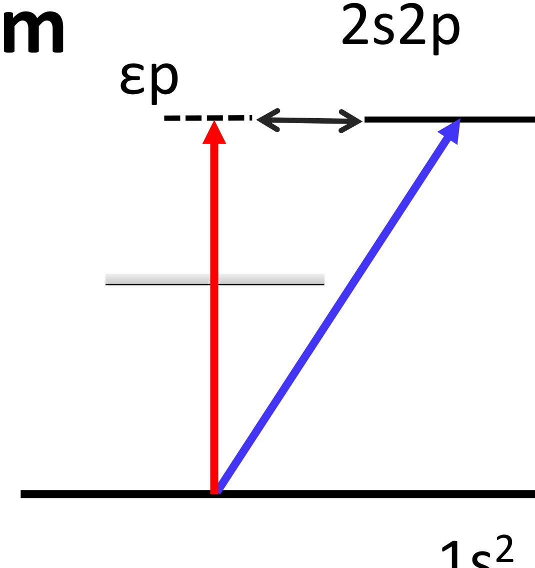


Collaboration
P. Salières,
F. Martin

Gruson et al. Science, 2016
Kaldun et al., Science, 2016

Time-frequency representation: Short time Fourier transform

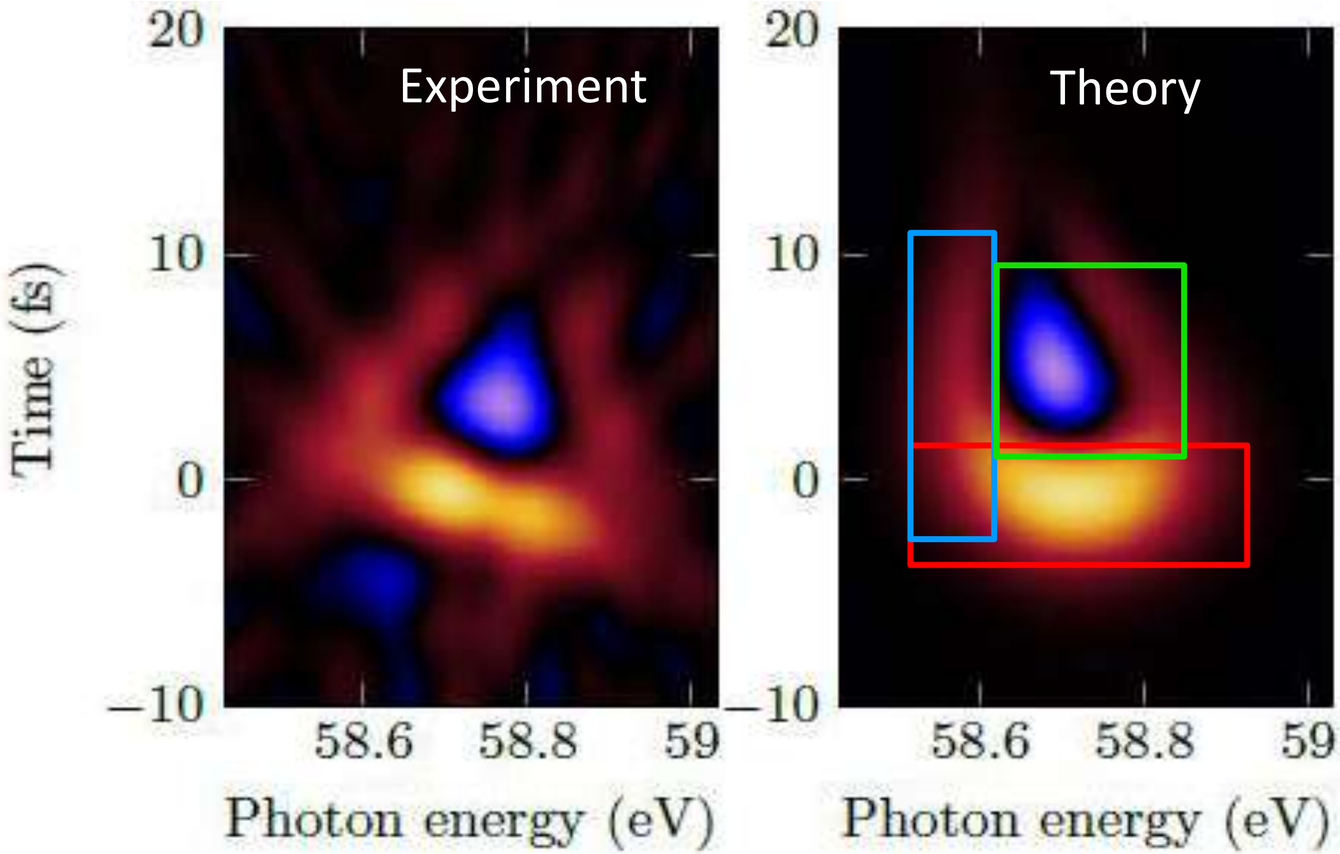


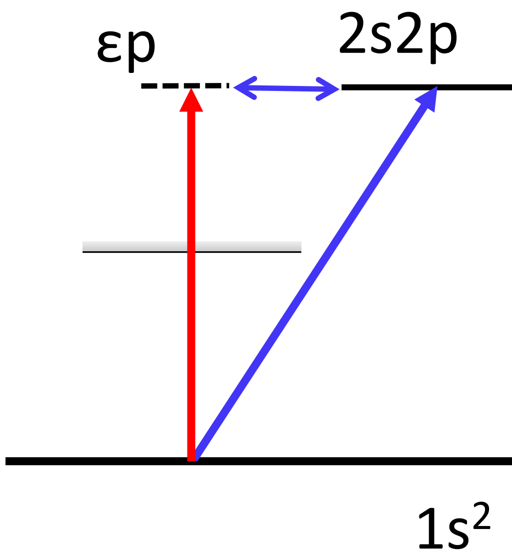


Collaboration
P. Salières
R. Taïeb
F. Martin

Wigner representation of the autoionizing wavepacket

$$W(E,t) = \int A\left(t + \frac{\tau}{2}\right) A^*\left(t - \frac{\tau}{2}\right) e^{-\frac{iE\tau}{\hbar}} d\tau$$



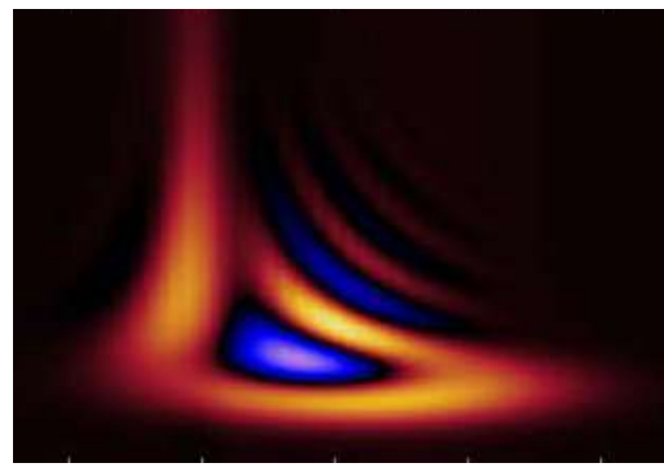


Collaboration
P. Salières
R. Taïeb
F. Martin

Conclusion

Multiple development of attosecond light sources for specific applications

Many-electron physics in the time domain using spectral amplitude and phase measurements



Thank you for your attention

J. Mauritsson, P. Johnsson, M. Gisselbrecht, C.L. Arnold, P. Rudawski, S. Zhong, S. Nandi, C. Guo, H. Coudert-Alteirac, M. Isinger, Y.-C. Cheng, S. Mikaelsson, D. Busto, H. Wikmark, J. Peschel, S. Maclot, L. Neoricic

Theoretical Support

J. M. Dahlström, (Lund) E. Lindroth (Stockholm), L. Argenti, F. Martín (Madrid), A. Maquet (Paris), R. Taïeb (Paris)