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# Atomic photoionization using attosecond pulses

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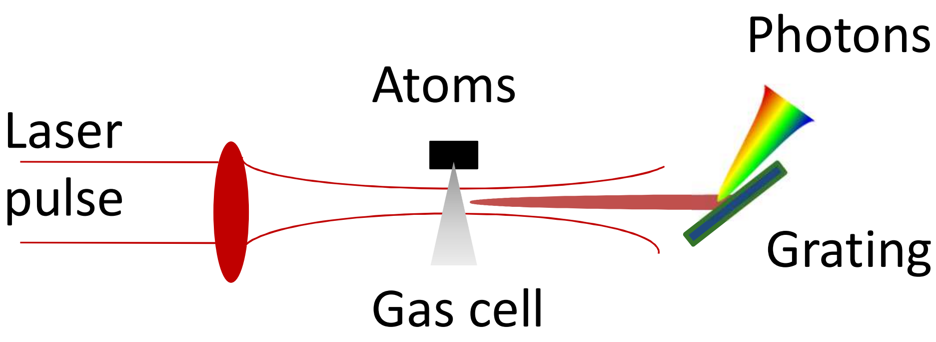
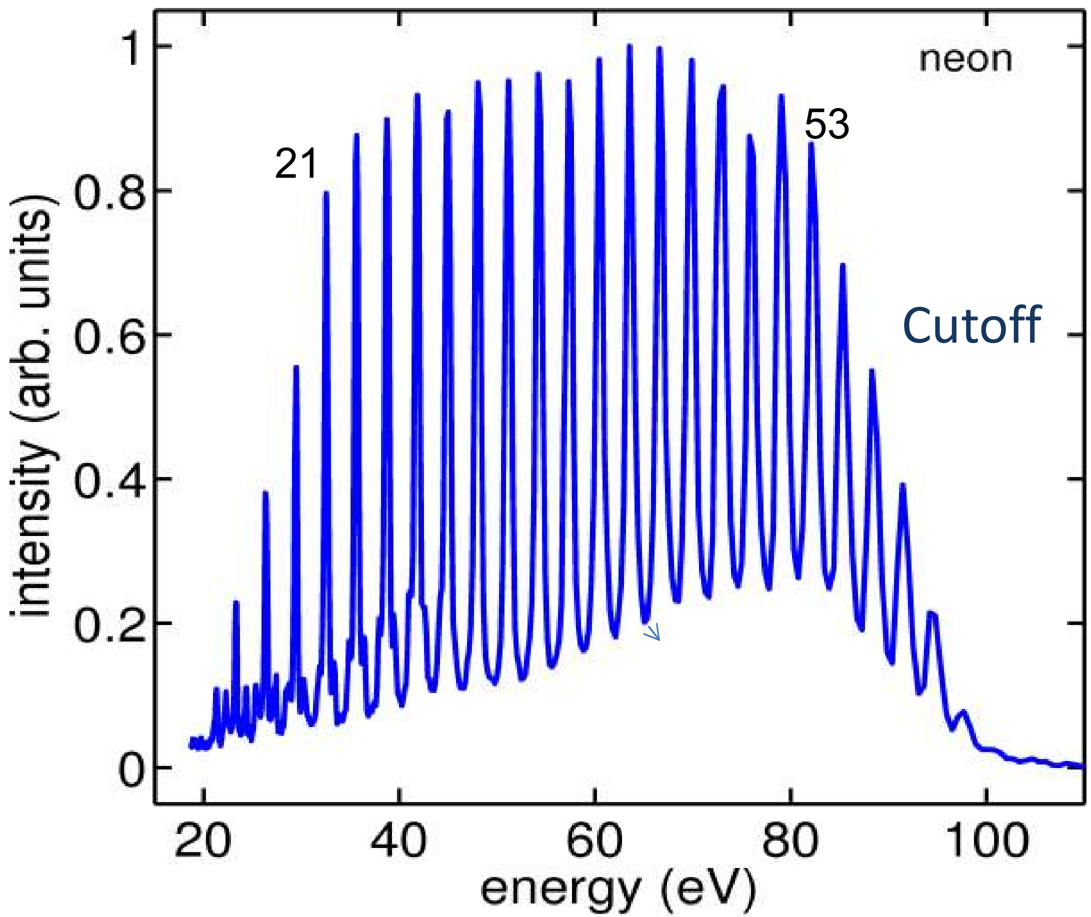
*Lund University*
*Sweden*

# Outline

- **Attosecond pulses**
  - A brief historical introduction
  - Measurement of attosecond pulses
- **Atomic Photoionization**
  - Photoionization time delays
  - Photoionization time delays Ne
  - Photoionization time delays Xe
  - Resonant photoionization He

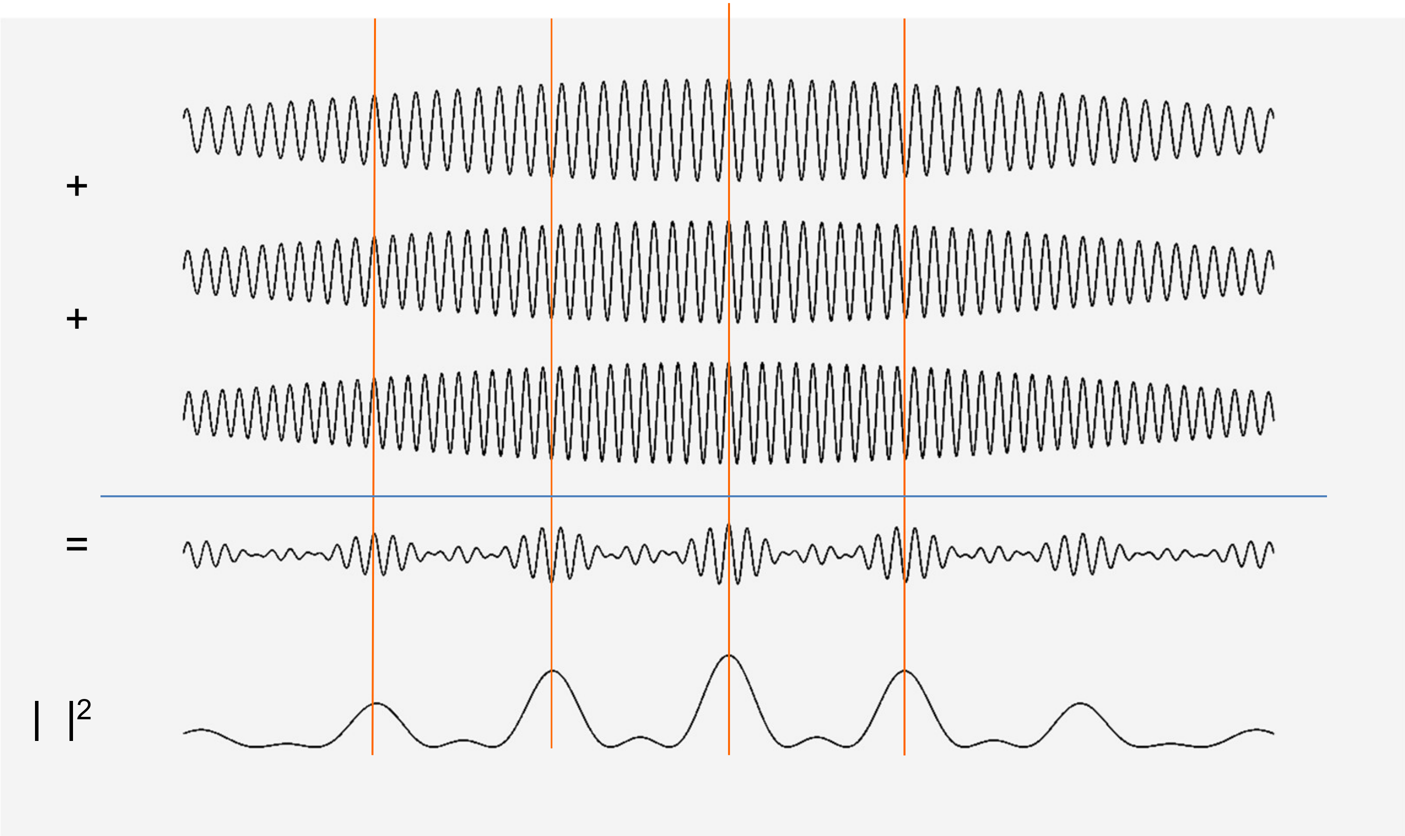
# Atoms in strong laser fields




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

# Attosecond pulses ?

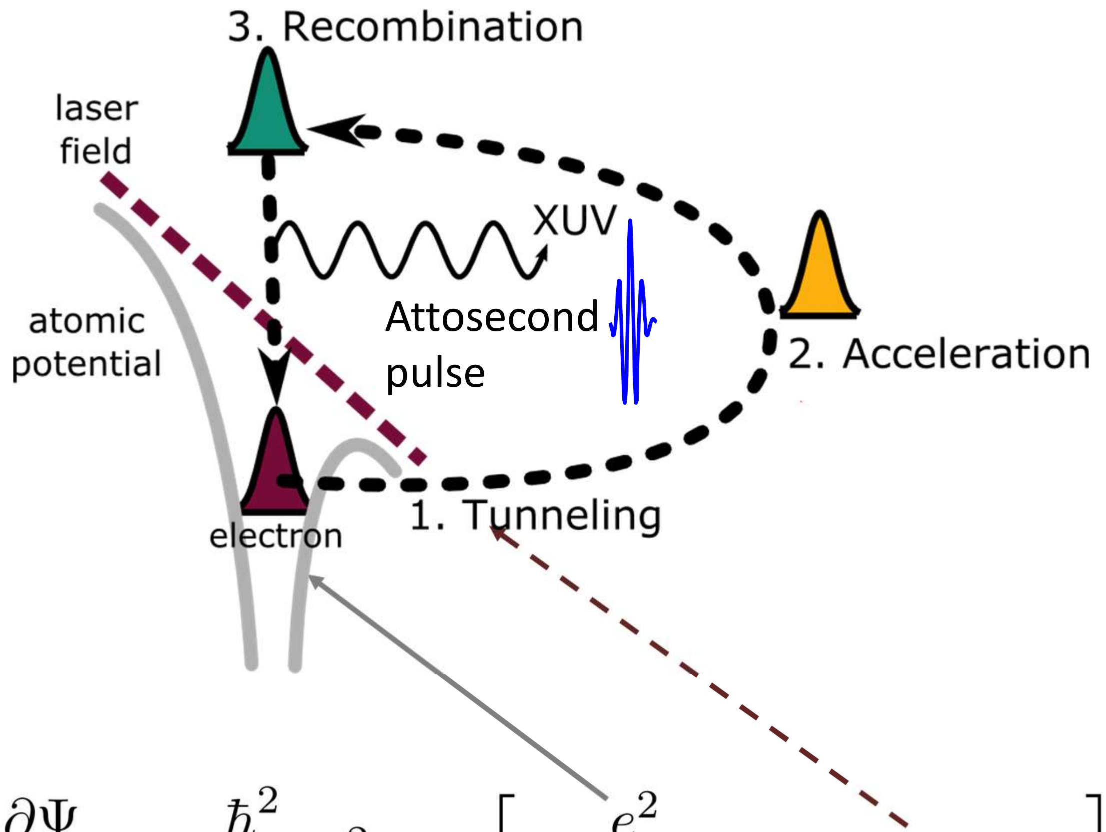



"If the harmonics are appropriately phased, this bandwidth corresponds to temporal pulses on the order of $5 \times 10^{-17}$ s, and thereby motivates a search for a new regime of short-pulse generation."

T. Hänsch, S. Harris

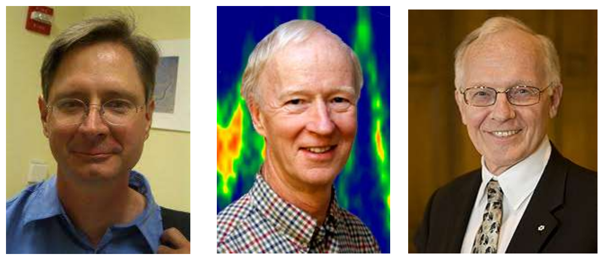
Farkas and Toth, PL, 1992, Hänsch, Harris, Opt. Comm 1993

# Atoms in strong laser fields: half cycle



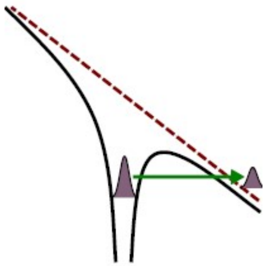

$$i\hbar\frac{\partial\Psi}{\partial t} = -\frac{\hbar^2}{2m}\nabla^2\Psi + \left[-\frac{e^2}{4\pi\epsilon_0 r} + eE_0\sin(\omega t)z\right]\Psi$$

- Numerical solution of the time-dependent Schrödinger equation
- Three-step model

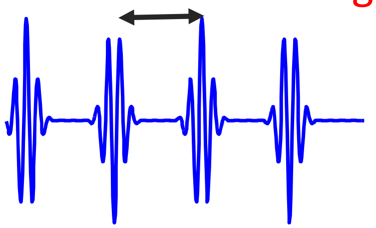


Schafer, Kulander, PRL 1993
Corkum, PRL 1993

# Atoms in strong laser fields: several cycles

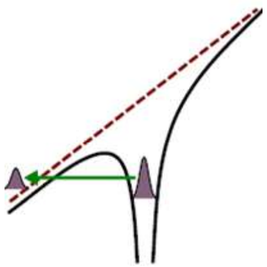


Time domain

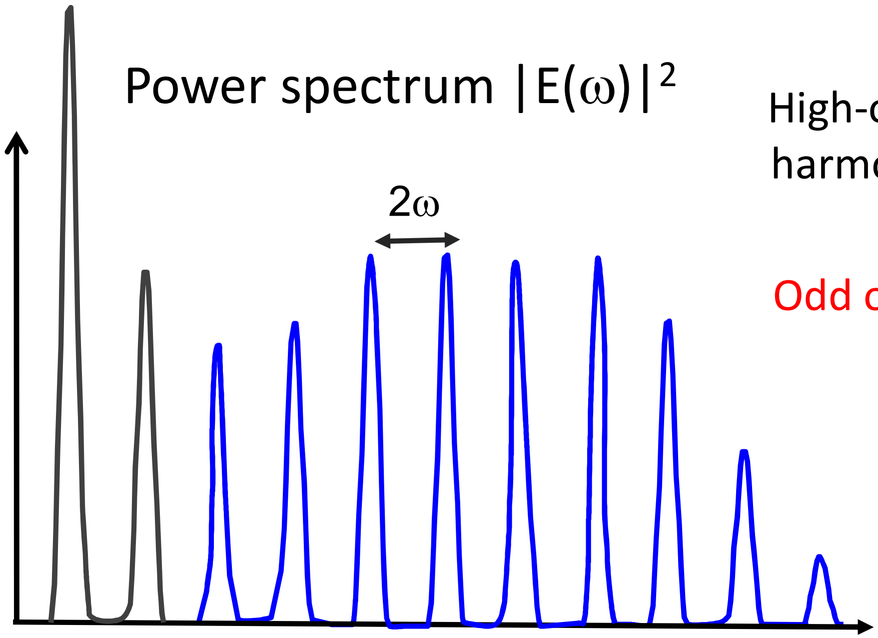



Sign flip

Electric field in time E(t)



Power spectrum $|E(\omega)|^2$




High-order harmonics

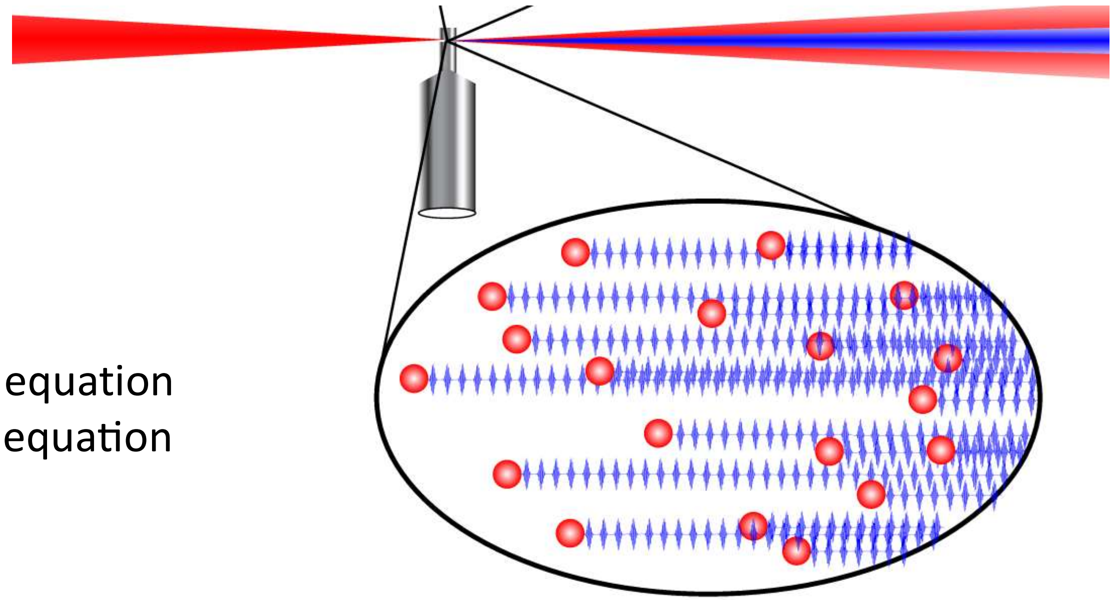
Odd order

Photon energy

**Harmonics = Interferences of attosecond pulses**

**Attosecond pulses= Sum of phase-locked high-order harmonics**

# Nonlinear Optics



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

Maxwell equation
→ Wave equation

Propagation | Diffraction | Dispersion | Nonlinear polarization

$$\left[\frac{\partial}{\partial z} - \frac{i}{2k(\Omega)}\Delta_\perp + i\left\{k(\Omega) - \Omega/c\right\}\right]\hat{\mathcal{E}} = \frac{i\Omega^2}{2k(\Omega)c^2\epsilon_0}\hat{\mathcal{P}}$$

Gaarde et al., 2002, Popmintchev et al., 2012, Heyl et al., 2017

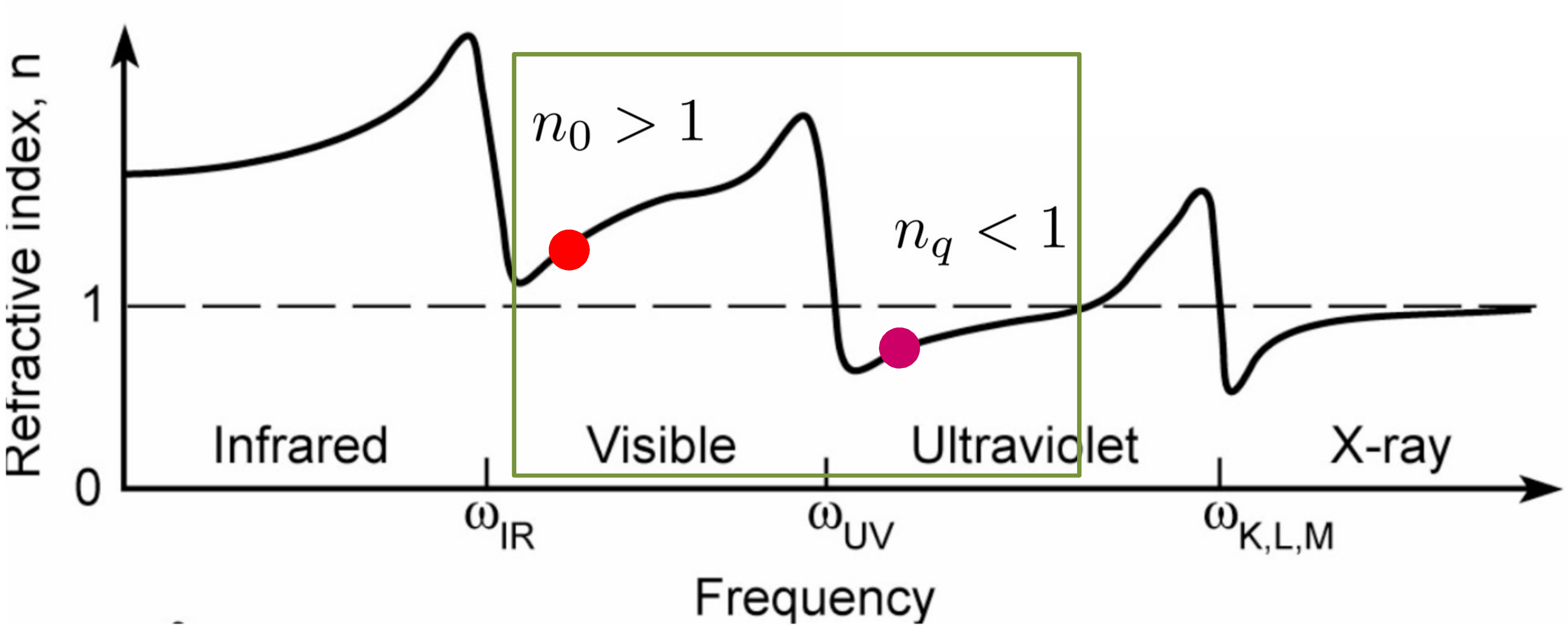
# Phase matching $\Omega = q\omega$

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

$$\frac{c}{n_0} = \frac{c}{n_q}$$

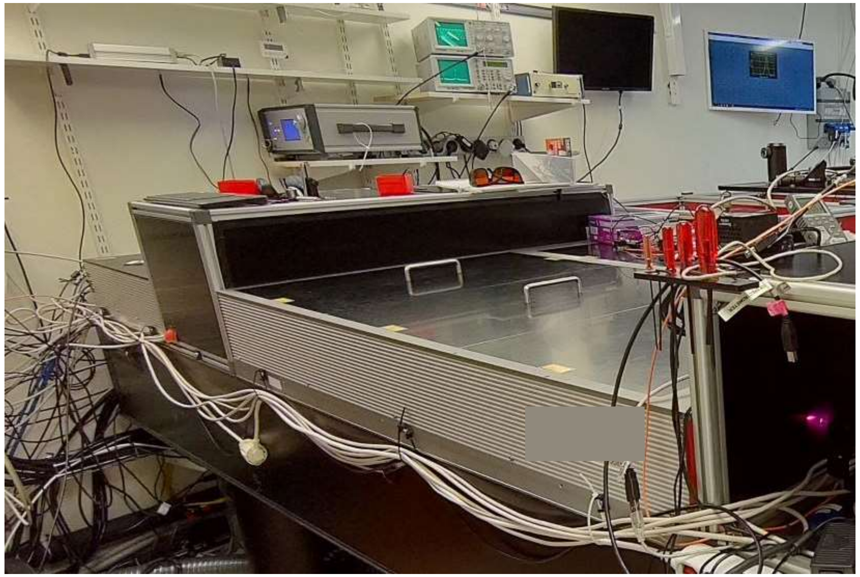
$$n_0 = 1 + \frac{\mathcal{N}_n \alpha}{2\epsilon_0}$$

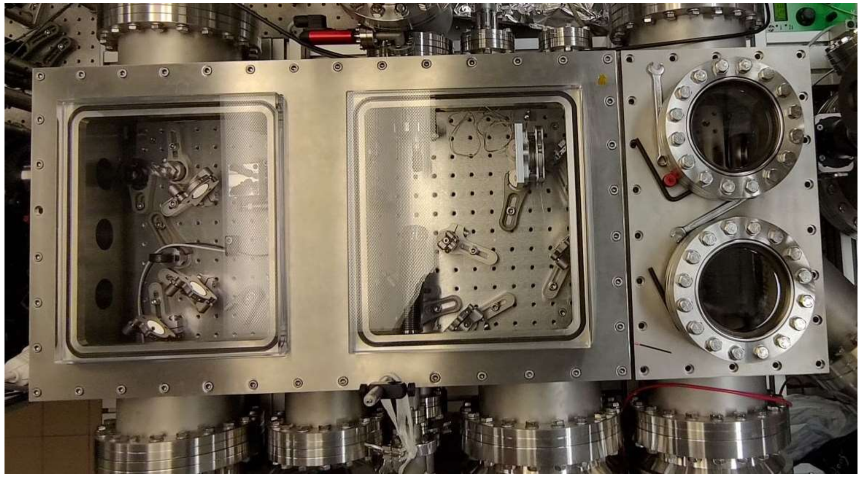
Degree of ionization = a few %



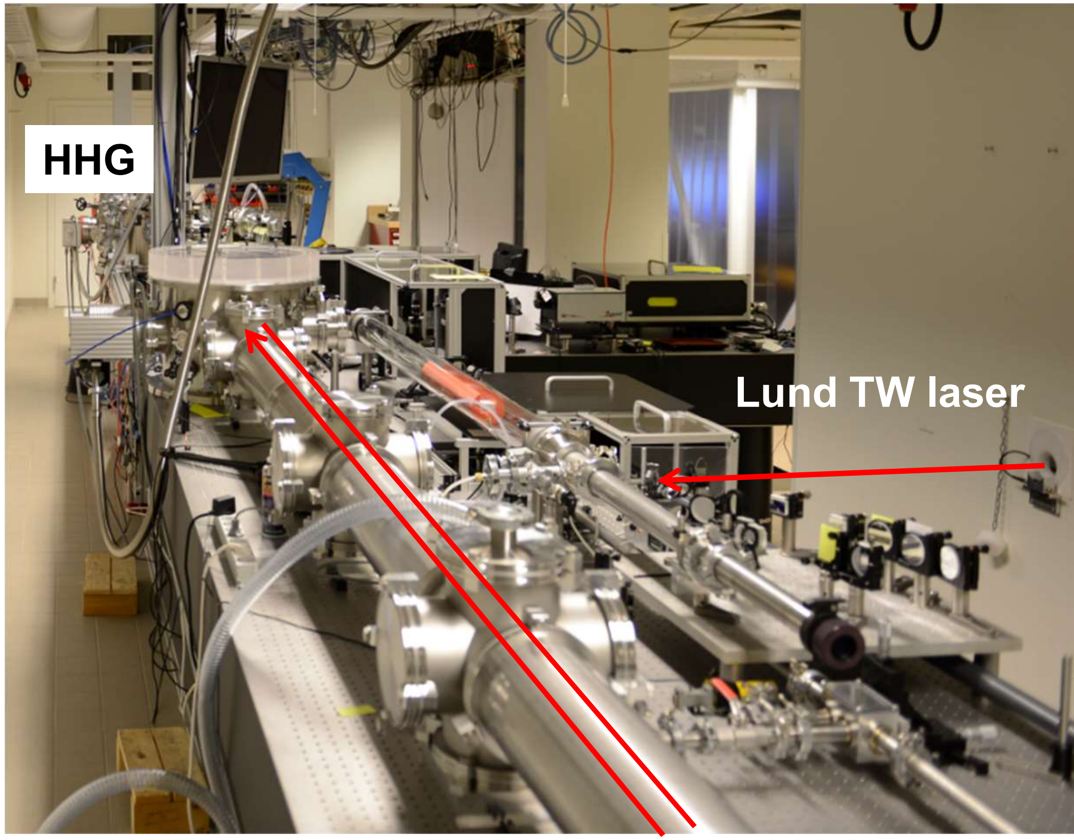

Gaarde et al., JPB, 2008, Heyl et al., JPB, 2017, Attwood, 2017

# Attosecond sources





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

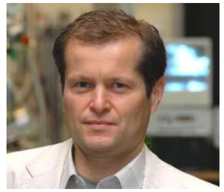



Manschwetus et al., PRA, 2016

# Attosecond Pulses

Paris, 2001, 250 as — RABBIT technique – Interferometry

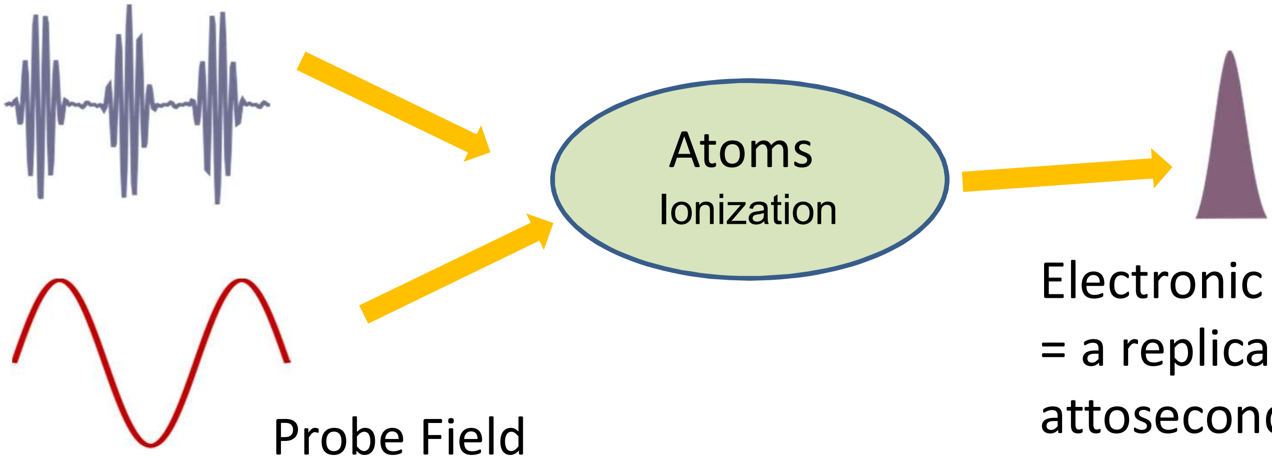
Pierre Agostini



Vienna, 2001, 450 as — Streaking technique

Ferenc Krausz

Attosecond pulses

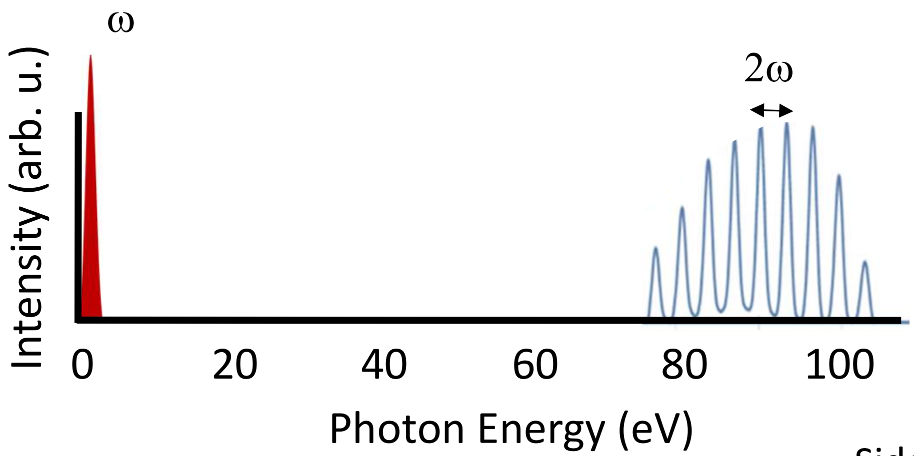


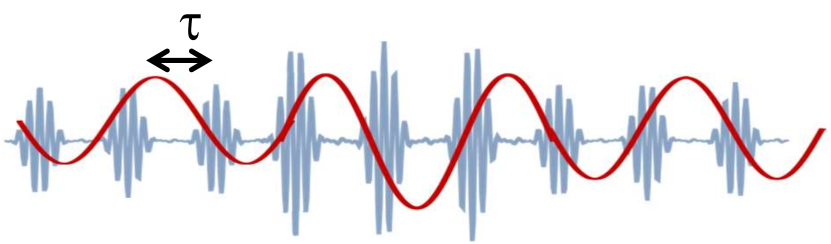

Probe Field (IR)

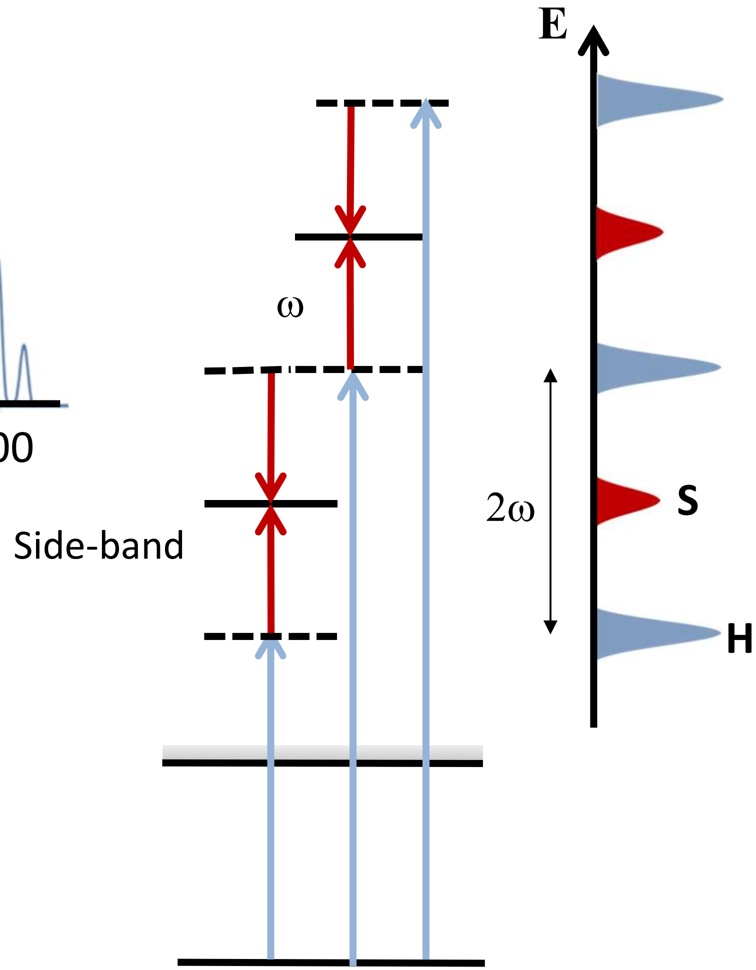
Electronic Wave packet = a replica of the attosecond pulse

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

# RABBIT technique: interferometry

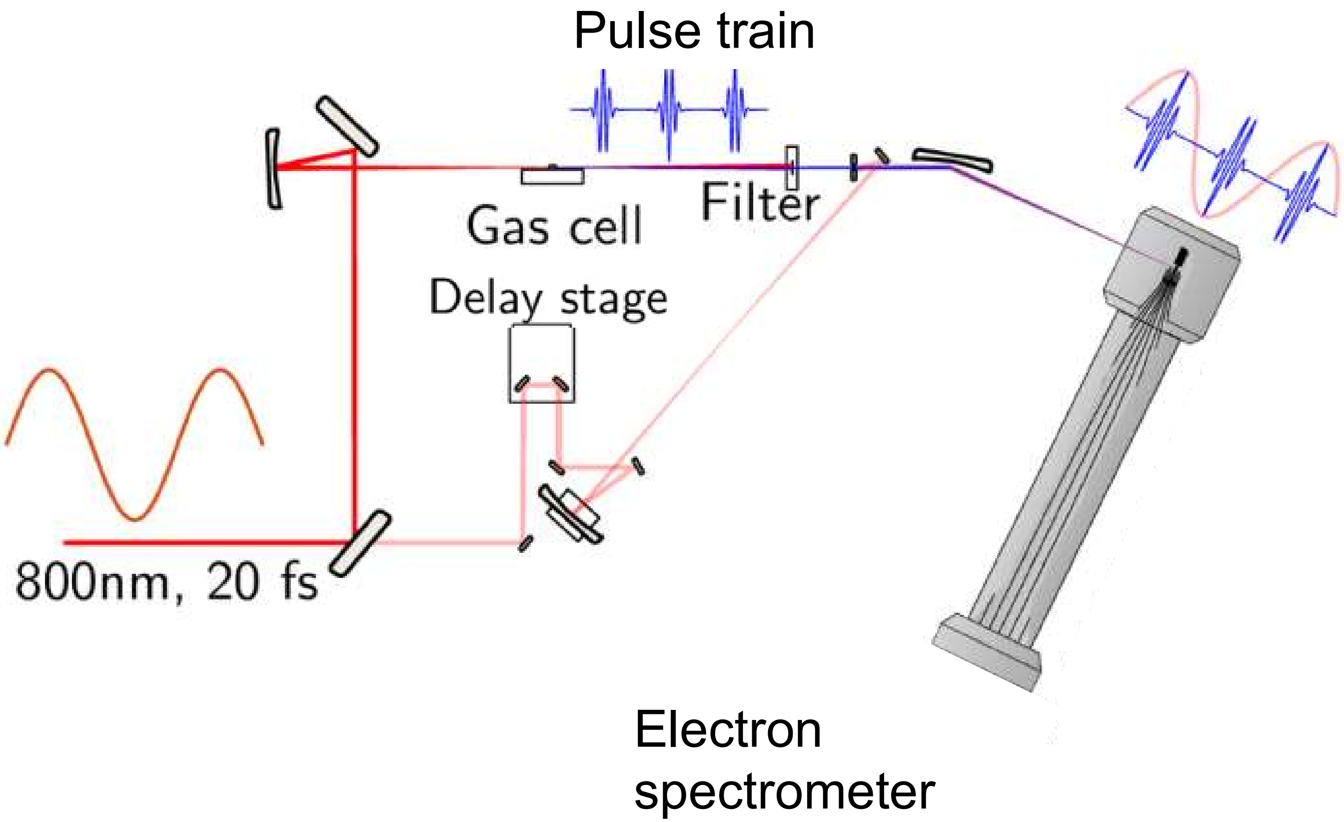


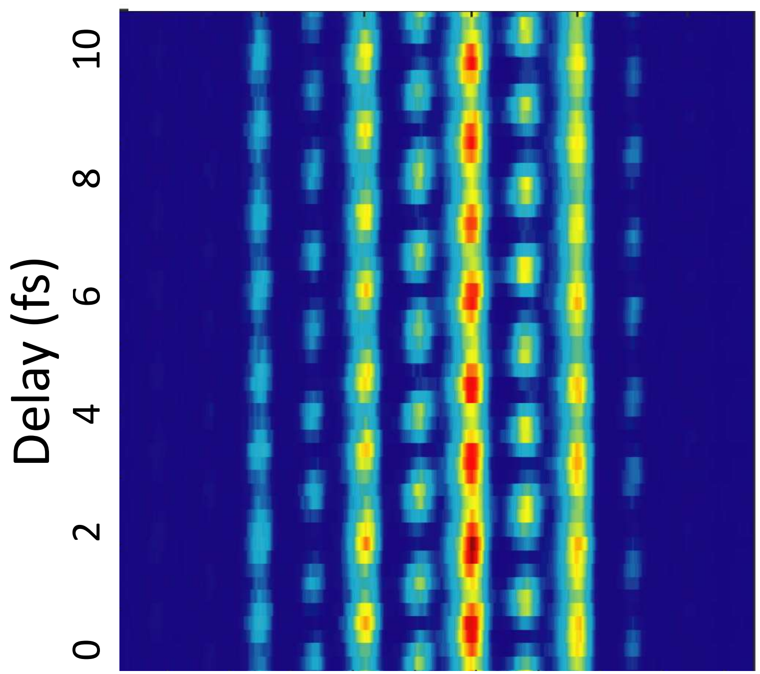



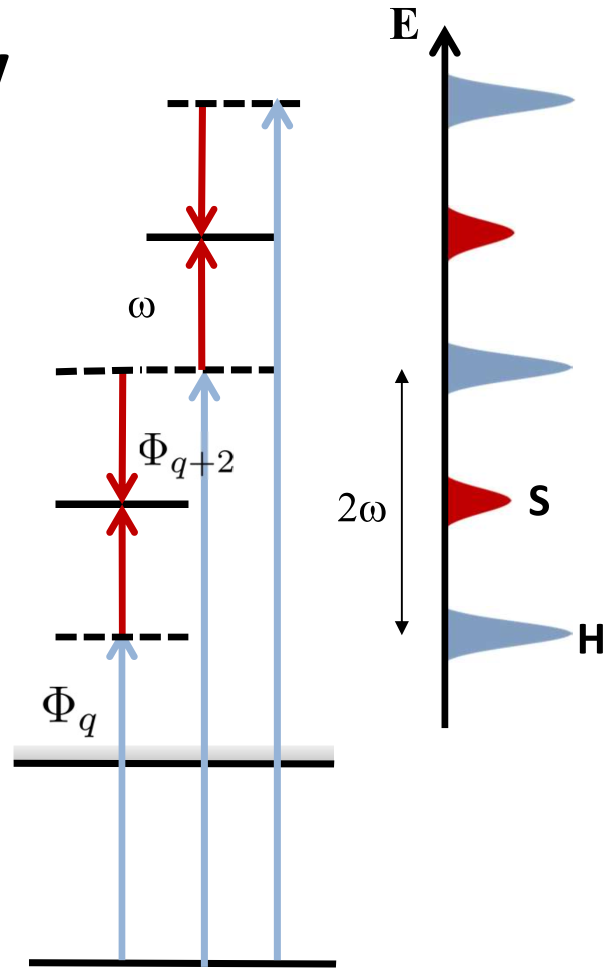

Paul et al., Science, 2001, Véniard et al., Phys. Rev. A 1996

# Optical interferometry



# Quantum interferometry




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

$$\tau_{\rm XUV} = \frac{\Delta\phi}{2\omega} = \frac{\Phi_{q+2} - \Phi_q}{2\omega} \simeq \frac{d\Phi}{d\Omega}$$

$\frac{d\Phi}{d\Omega}, \Phi(\Omega)$

$|E(\Omega)|^2$

FT

→ $E(\Omega)$ → $E(t)$

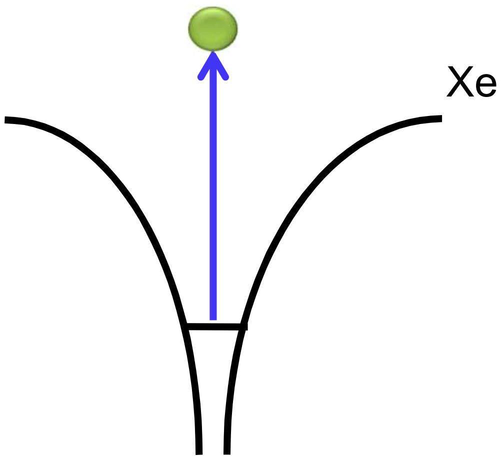
# Outline

- **Attosecond pulses**
  - A brief historical introduction
  - Measurement of attosecond pulses

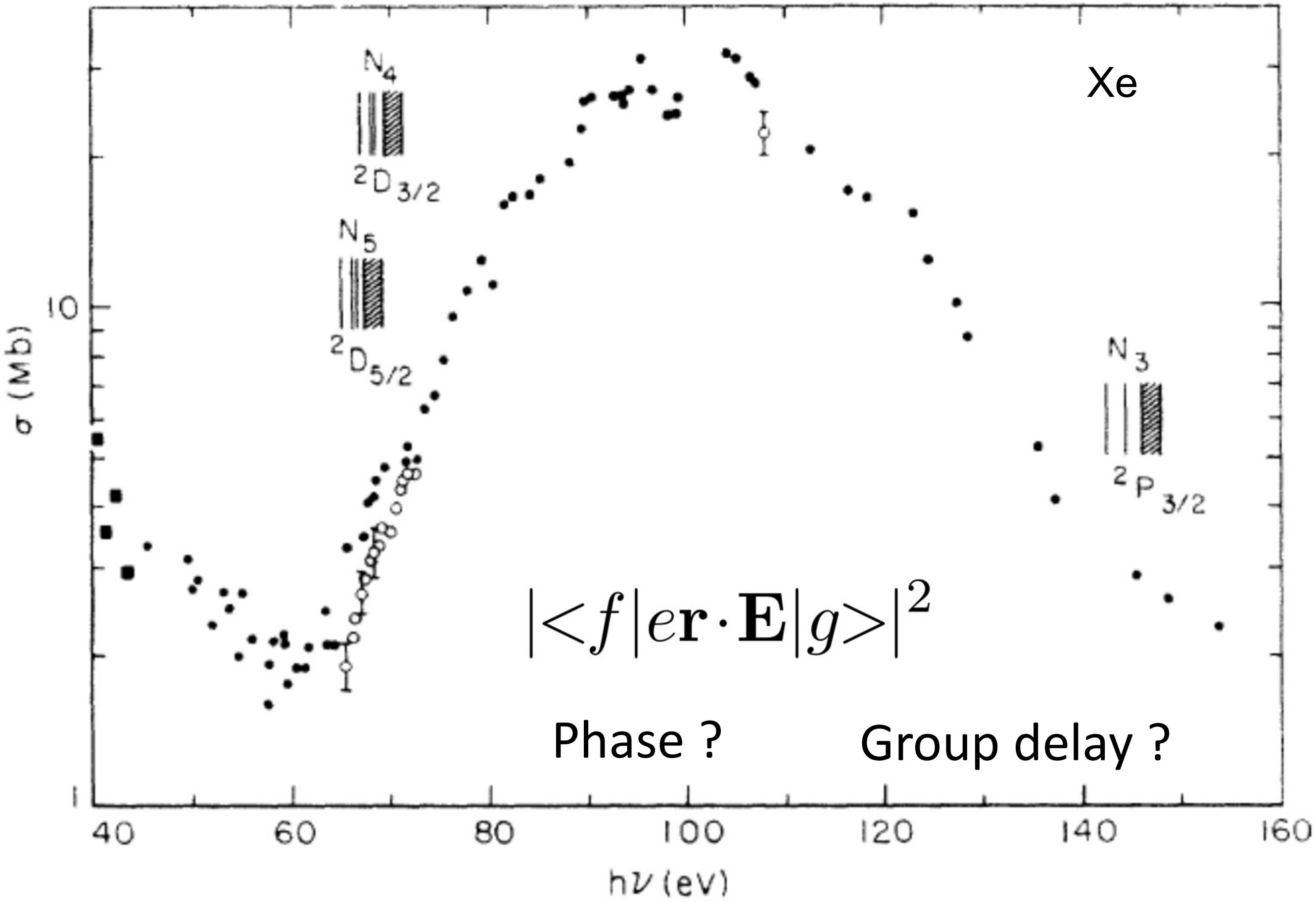
[1] A. L'Huillier, *"Generation of high-order harmonics and attosecond pulses"*, in Current trends in Atomic Physics, Ed. A. Browaeys, T. Lahaye, T. Porto, C. S. Adams, M. Weidemüller and L. F. Cugliando, Oxford University Press, Chap. 8 (2019)

- **Atomic Photoionization**
  - Photoionization time delays
  - Photoionization time delays Ne (attosecond metrology)
  - Photoionization time delays Xe (atomic physics)
  - Resonant photoionization He (quantum optics)

# Photoionization in the time and frequency domain

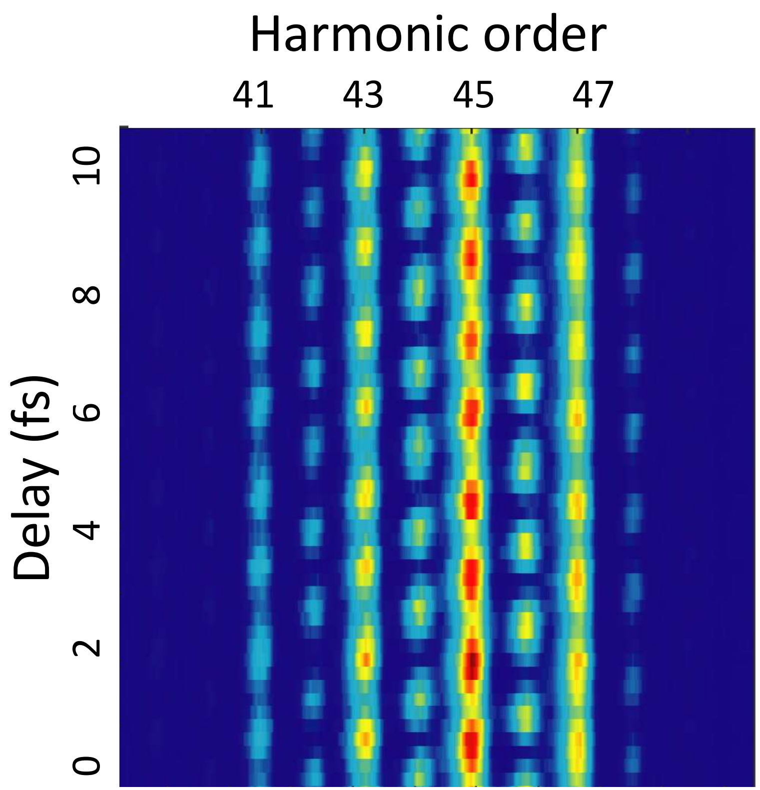


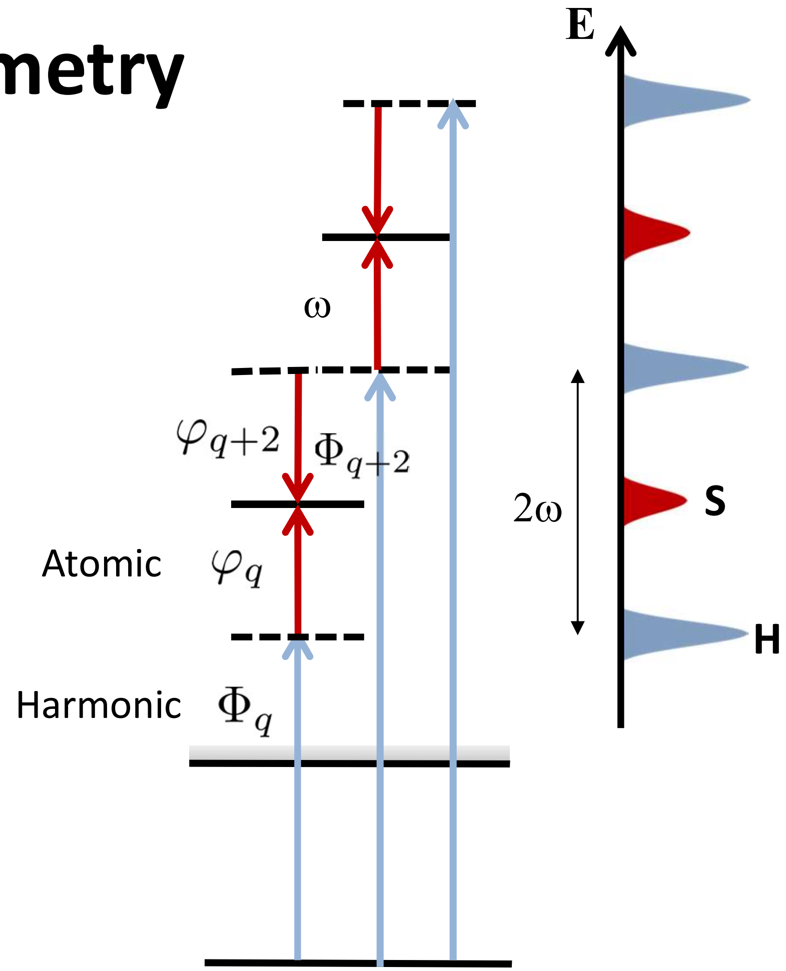

Time for
ionization ?




Ederer, PRL 1964

# Quantum interferometry



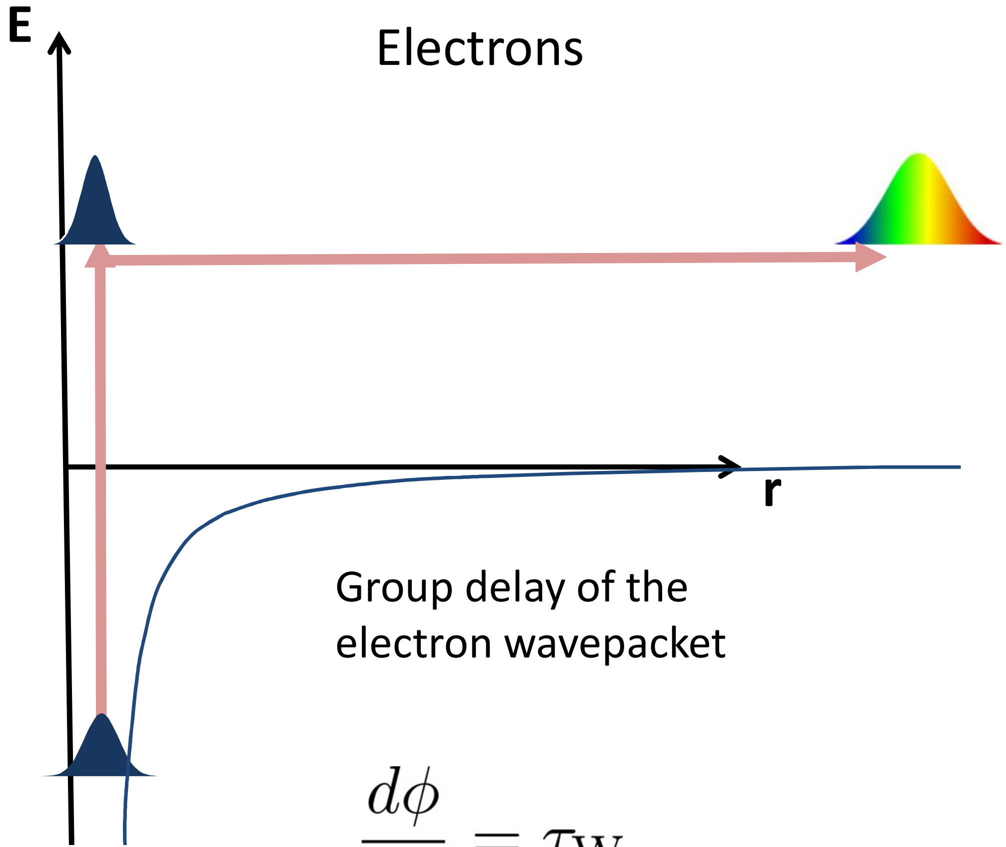

$$S \propto A + B\cos(2\omega\tau - \Delta\phi)$$

$$\frac{\Delta\phi}{2\omega} = \frac{\Phi_{q+2} - \Phi_q}{2\omega} + \frac{\varphi_{q+2} - \varphi_q}{2\omega} = \tau_{\mathrm{XUV}} + \tau_{\mathrm{A}}$$

Physical interpretation of $\tau_{\mathrm{A}}$ ?
How can we distinguish the two contributions to the delay?

# Interpretation of the atomic delay

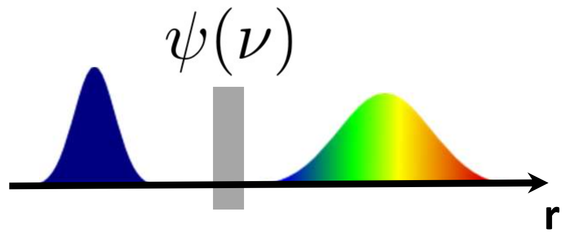
Electrons




Group delay of the electron wavepacket

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

Photons

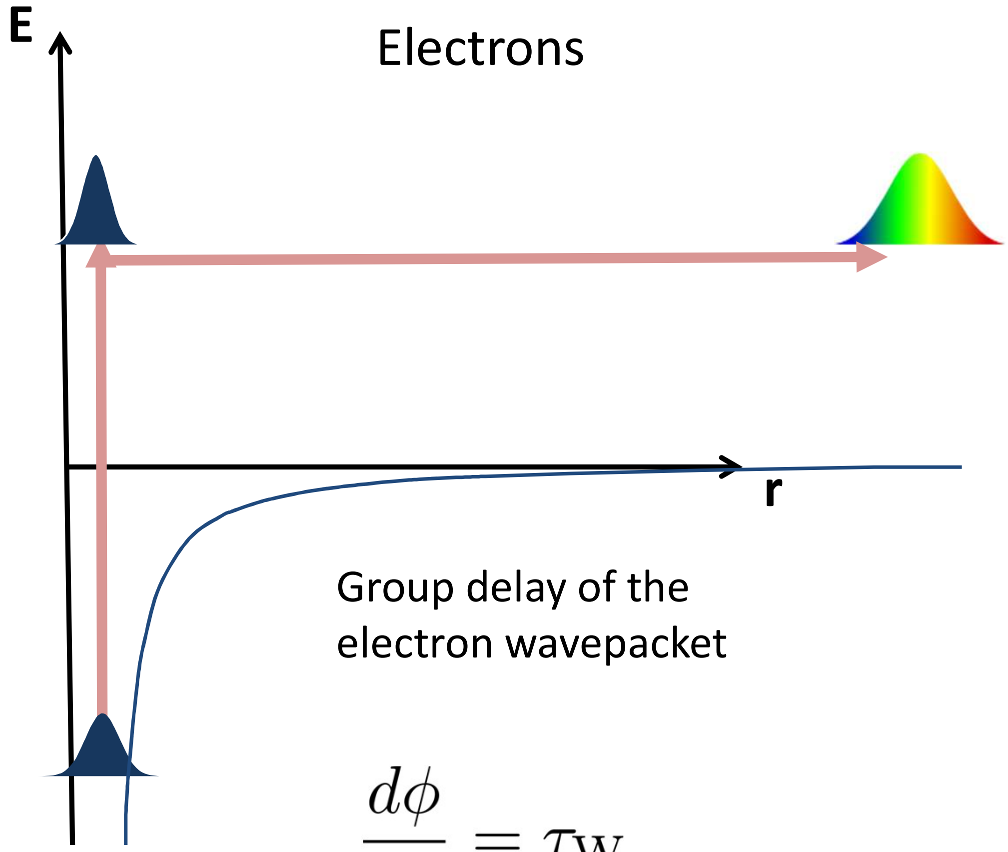



Group delay of light pulse

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

E. P. Wigner, Phys. Rev. 1955

# Interpretation of the atomic delay




Group delay of the electron wavepacket

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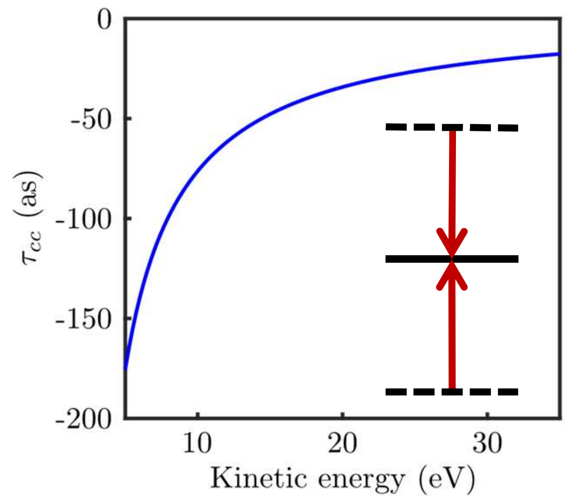
E. P. Wigner, Phys. Rev. 1955




Delay induced by the probe field $\tau_{\mathrm{cc}}$

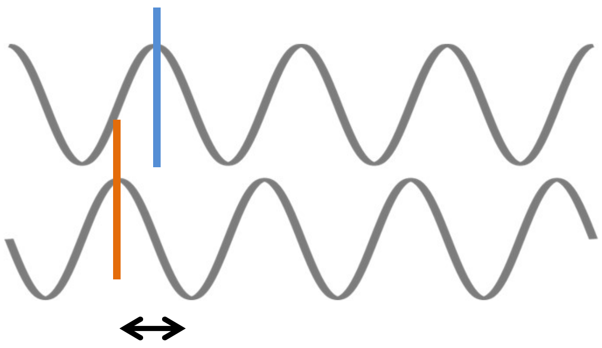
M. Dahlström, A. Maquet

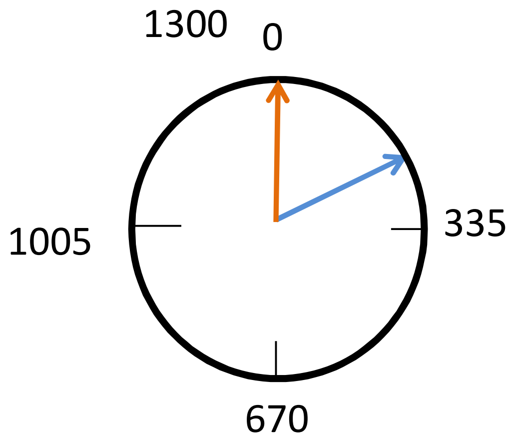
$$\tau_{\mathrm{A}} = \tau_{\mathrm{W}} + \tau_{\mathrm{cc}}$$

# Delay difference measurements

$$\cos[2\omega(\tau - \tau_{\mathrm{XUV}} - \tau_{\mathrm{A}})]$$

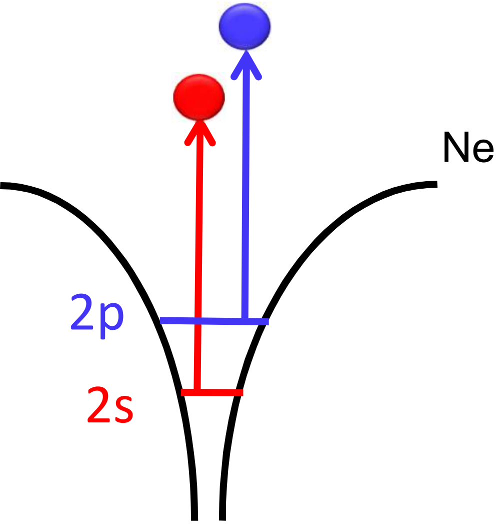
$$\cos[2\omega(\tau - \tau_{\mathrm{XUV}} - \tau_{\mathrm{A}})]$$



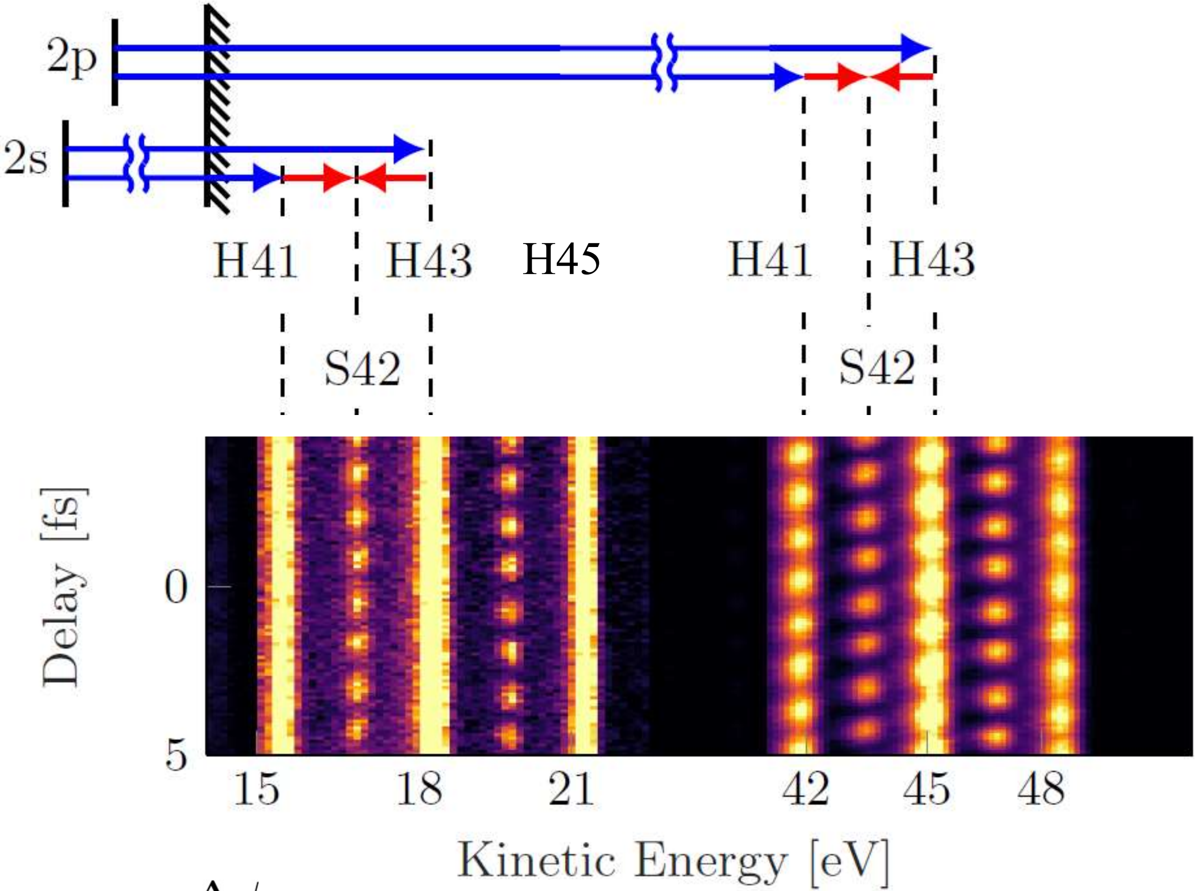



- Comparison of several processes (2s, 2p in Ne)
- Comparison of several species (Ne, Xe)

# Single ionization of Ne in 2s and 2p shells




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

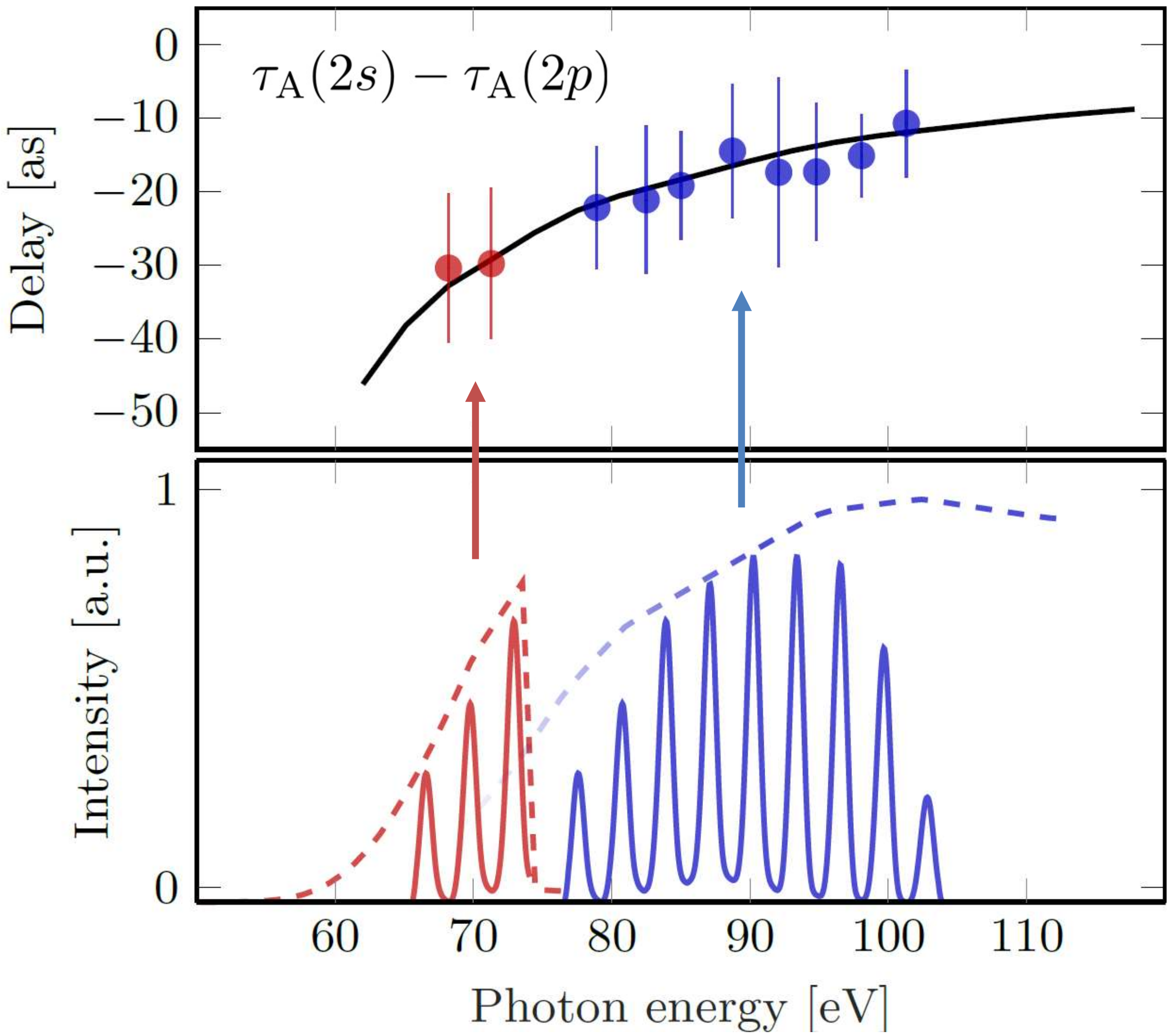



$$\frac{\Delta\phi}{2\omega} = \tau_{\mathrm{XUV}} + \tau_{\mathrm{A}}(2s) \qquad \frac{\Delta\phi}{2\omega} = \tau_{\mathrm{XUV}} + \tau_{\mathrm{A}}(2p)$$

$$\tau_{\mathrm{A}}(2s) - \tau_{\mathrm{A}}(2p)$$

Schultze et al., Science 2010 Isinger et al., Science 2017

# Single ionization of Ne in 2s and 2p shells



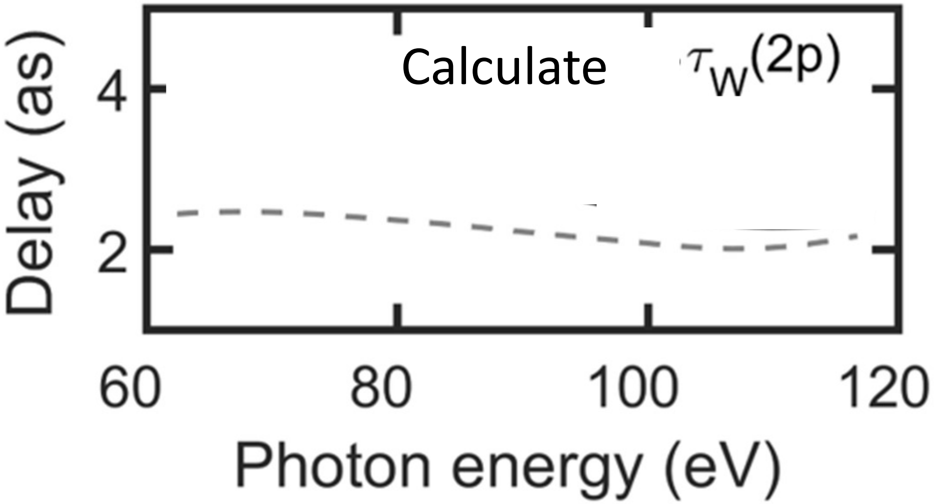

Theory

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

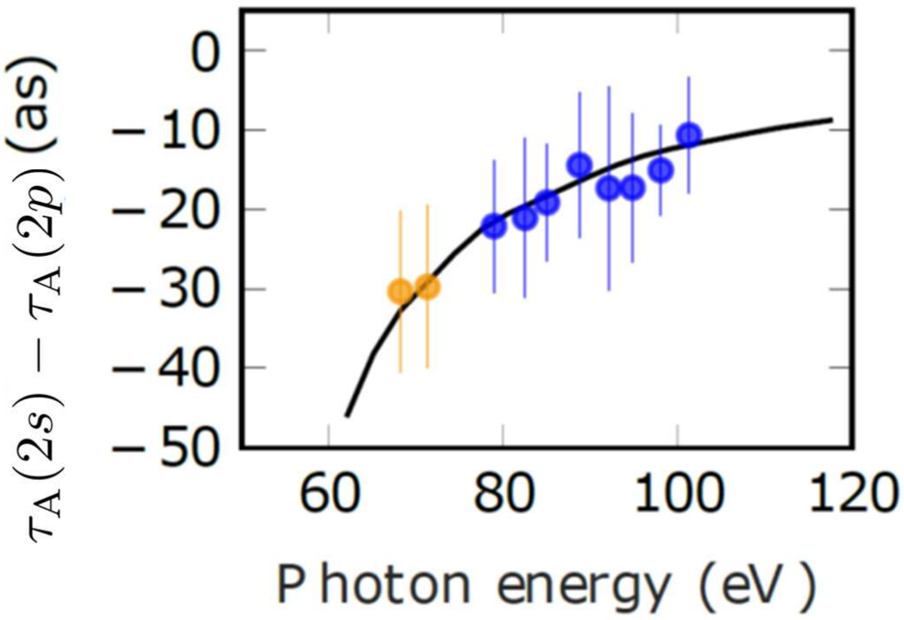

[2] M. Isinger, R. J. Squibb, D. Busto, S. Zhong, A. Harth, D. Kroon, S. Nandi, C. L. Arnold, M. Miranda, J. M. Dahlström, E. Lindroth, R. Feifel, M. Gisselbrecht and A. L'Huillier, *"Photoionization in the time and frequency domain"* Science **358**, 893 (2017)

# “Absolute” Photoionization time delays

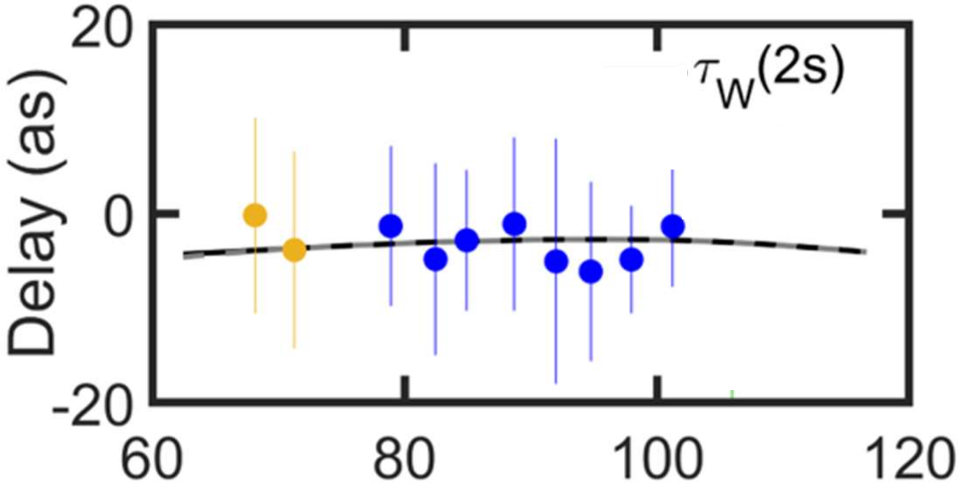
Calculate




$$\tau_A(2s) = \tau_W(2s) + \tau_{cc}(2s)$$
$$\tau_A(2p) = \tau_W(2p) + \tau_{cc}(2p)$$

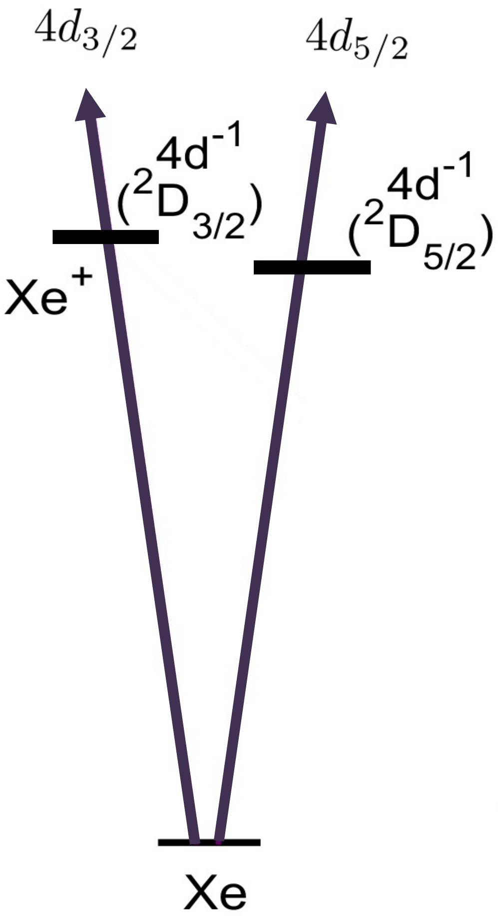



Calculate $\tau_{cc}(2s)$ $\tau_{cc}(2p)$




$$\tau_{cc}(2s) - \tau_{cc}(2p)$$

# Photoionization of Xe in the 4d shell

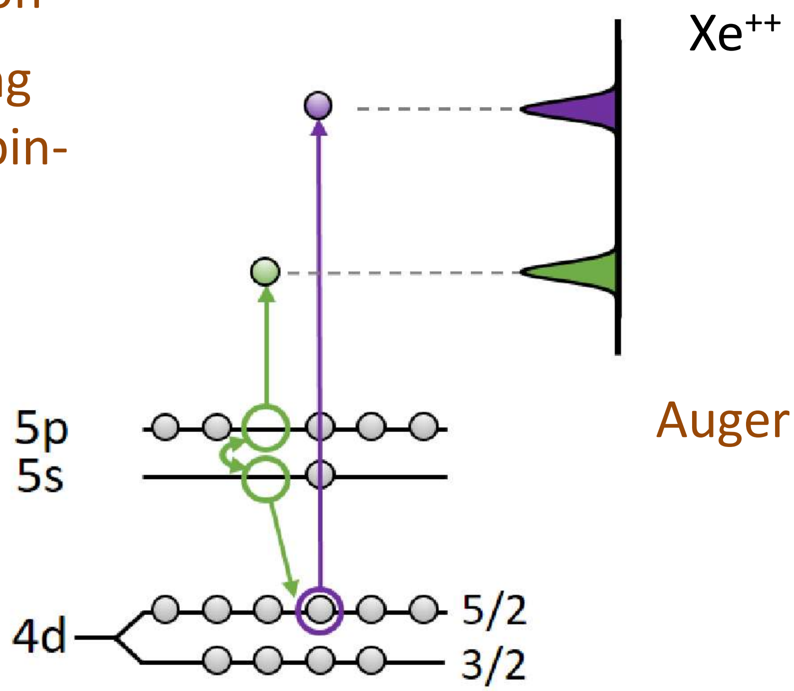



4d- ionization

Giant dipole resonance in Xe 4d

Spin-orbit interaction

Anomalous branching ratio between the spin-orbit channels

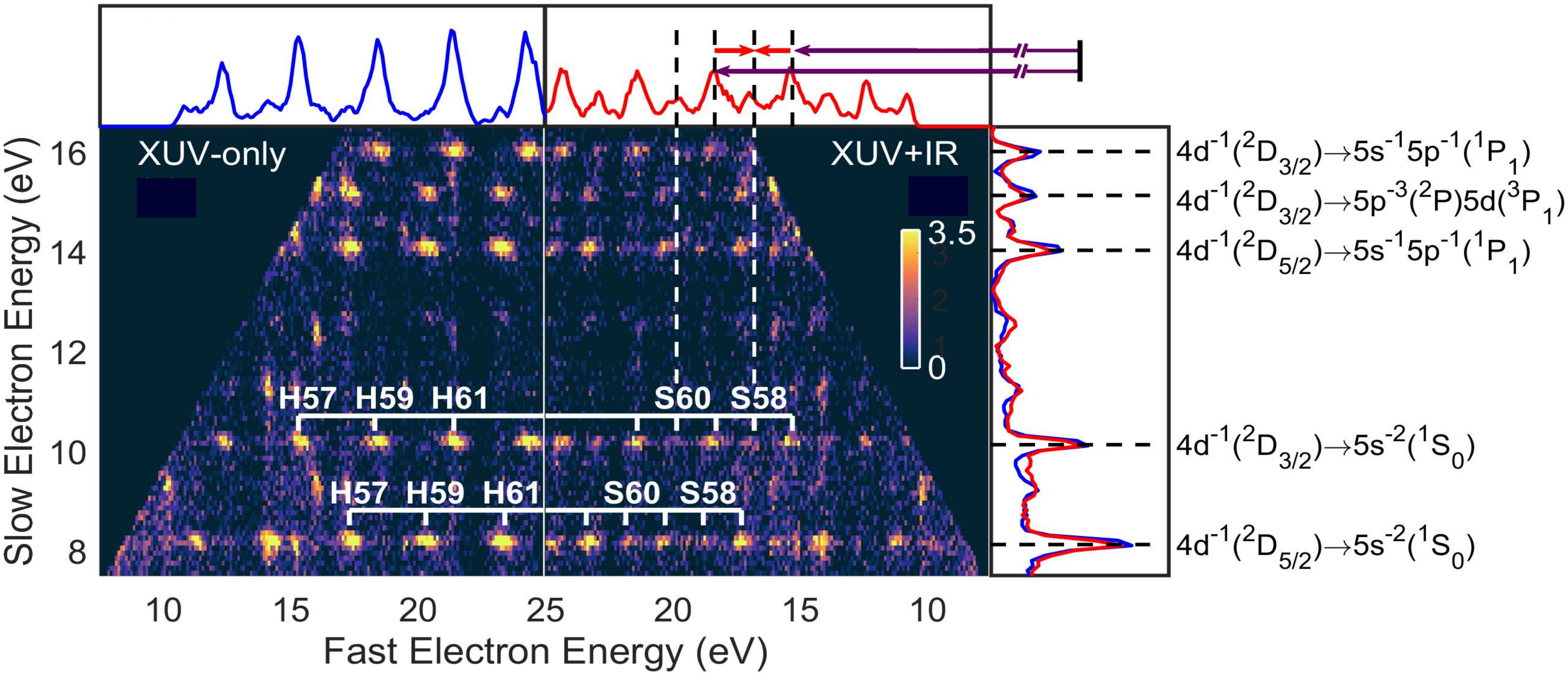



Auger decay

Drescher et al., Nature, 2002

Zhong et al., arXiv:2005.12008 [physics.atom-ph]

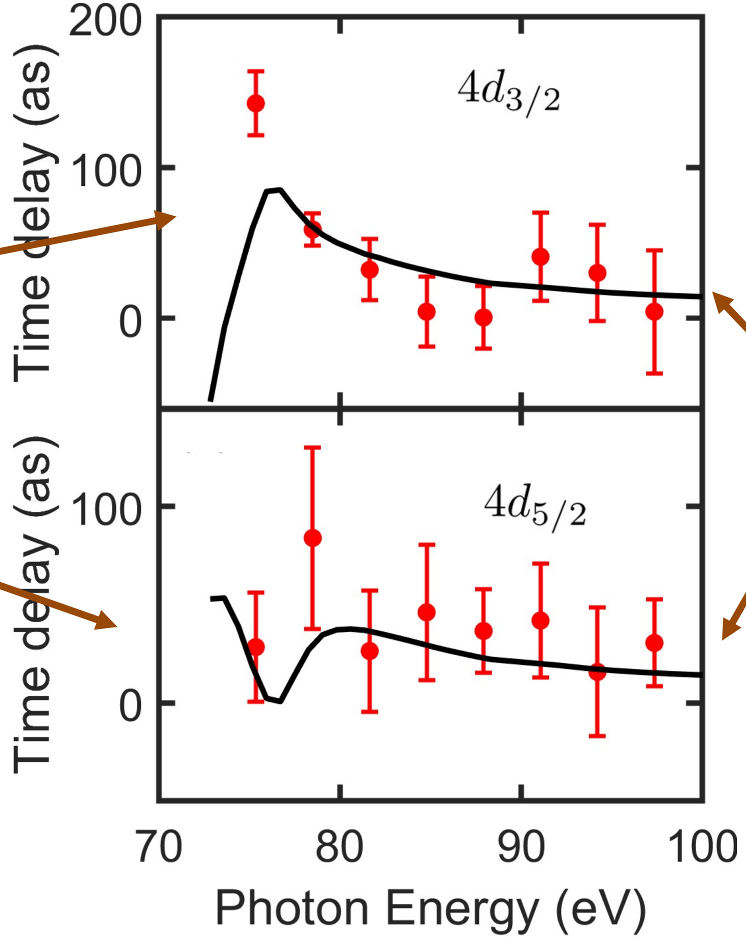
# Attosecond interferometry and coincidence spectroscopy




Auger

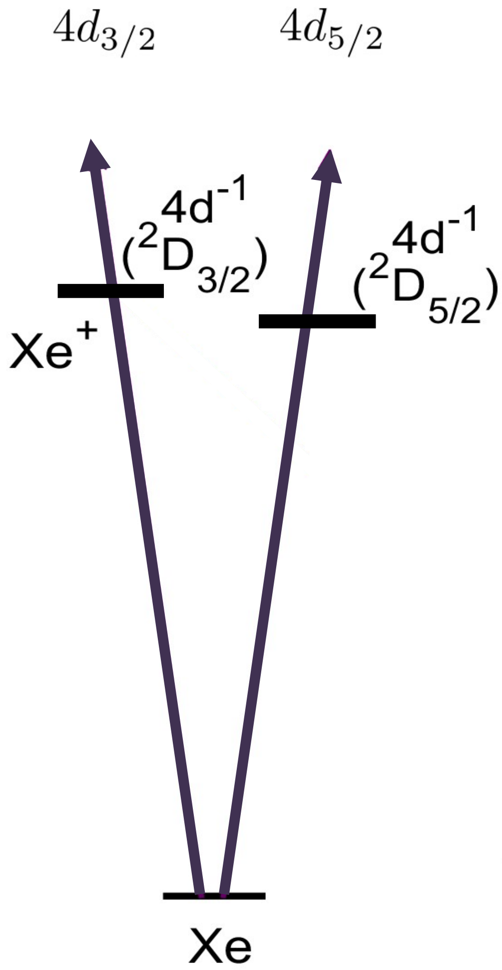
Photoelectron

# Photoionization time delays




Difference at threshold

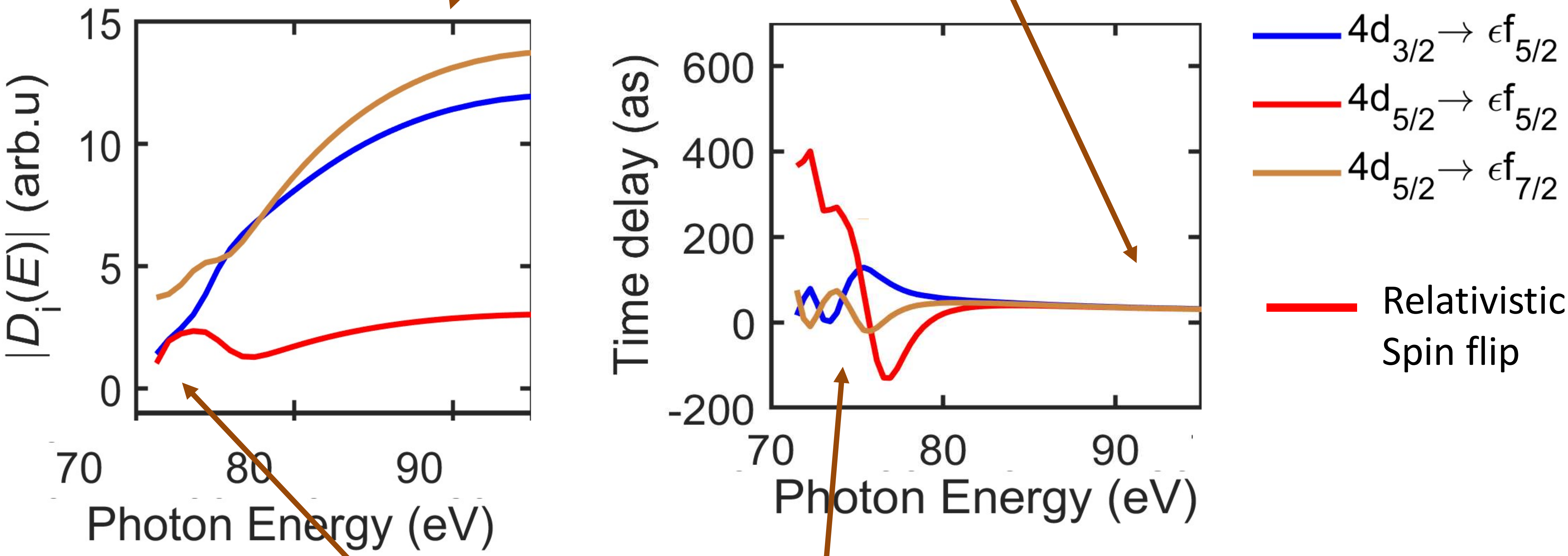
Positive delay




Relativistic Random Phase Approximation (RRPA)

# Channel amplitude and phase (delay)

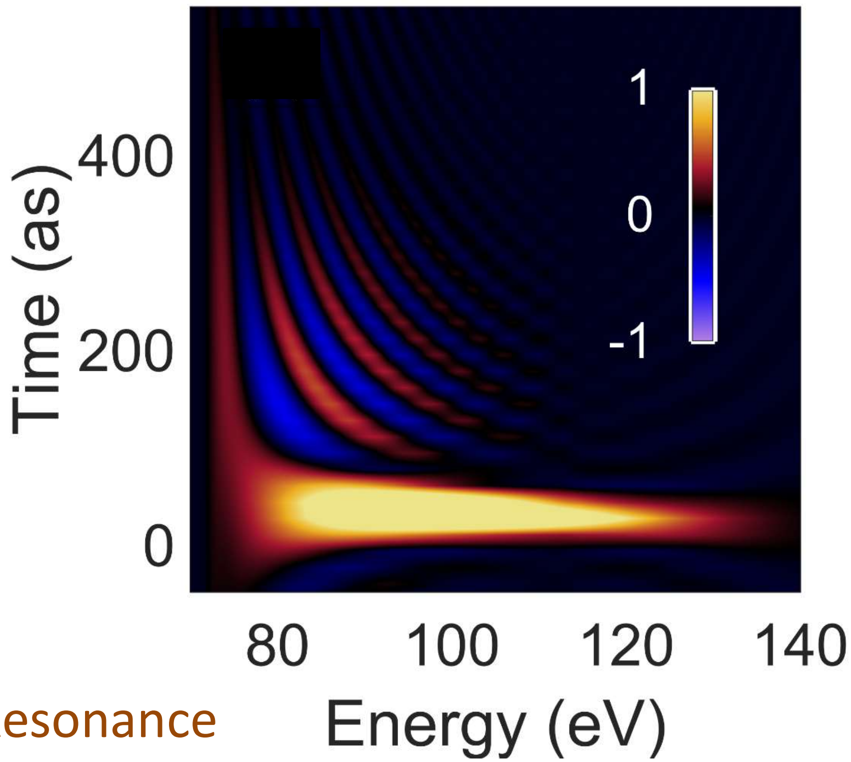
Giant dipole resonance
Electron correlation effects

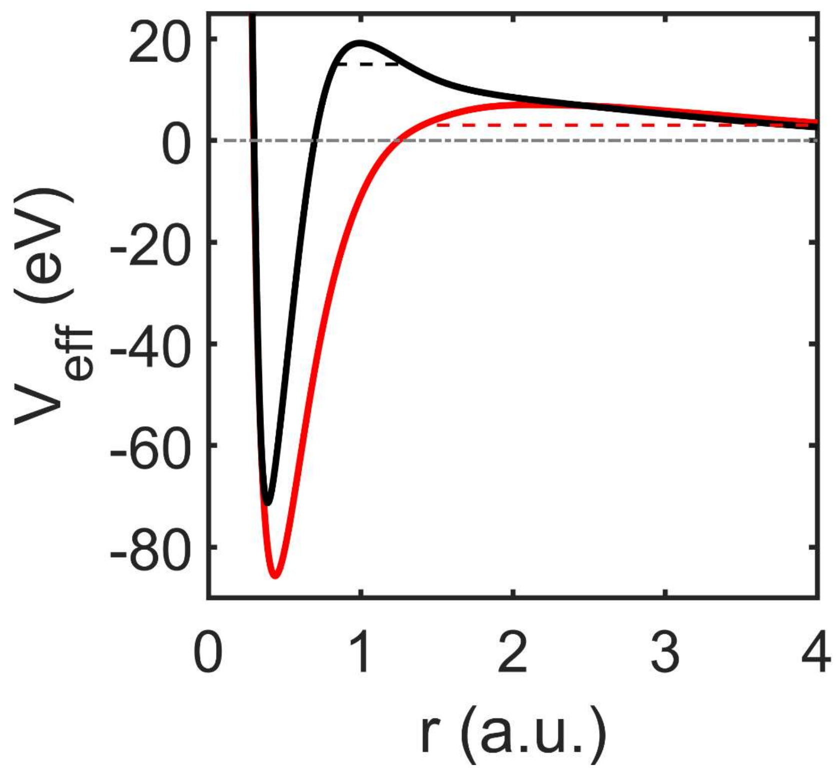



Interferences between
channels

# Ionization mechanisms and time scales

Wigner distribution

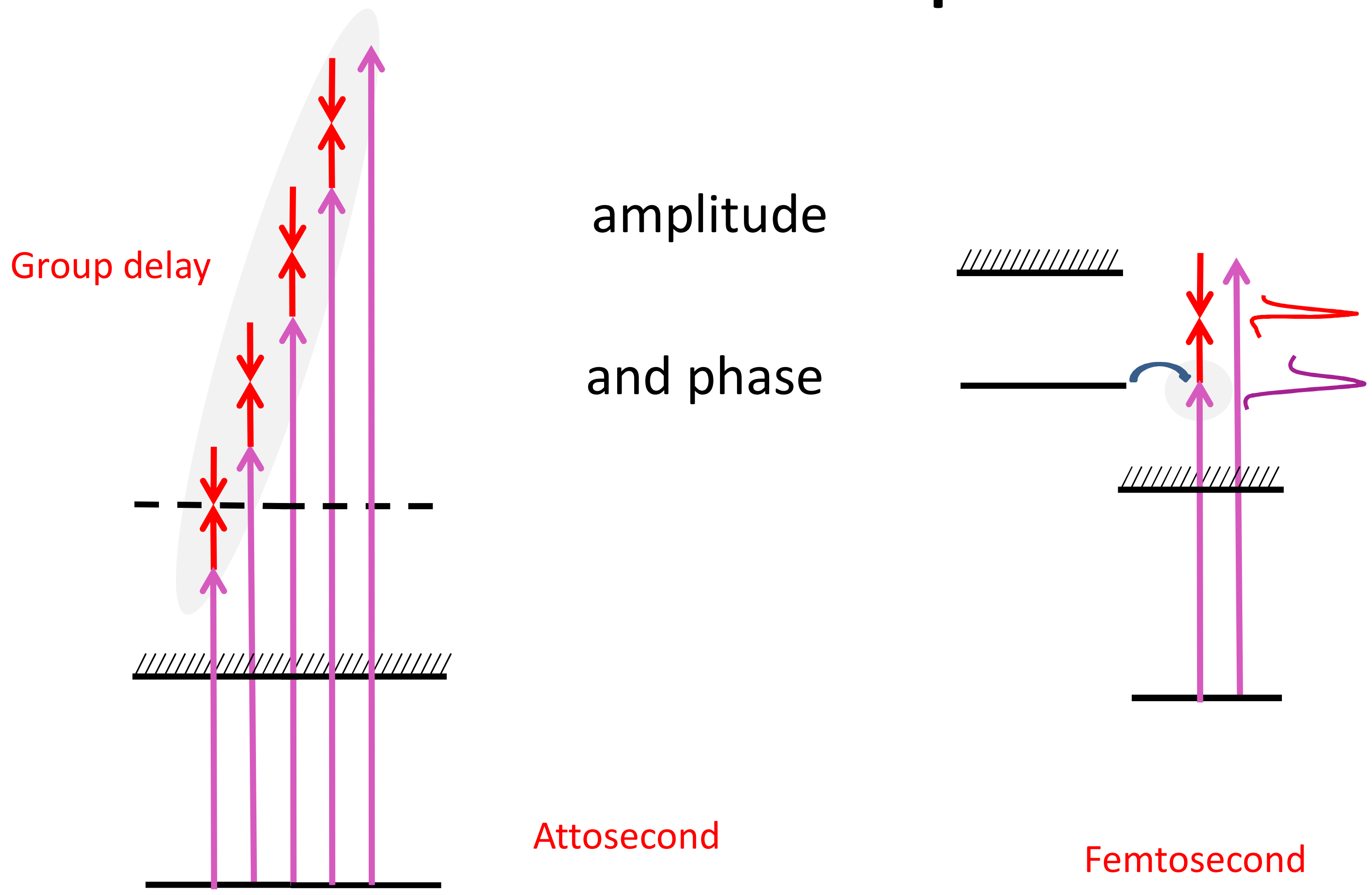


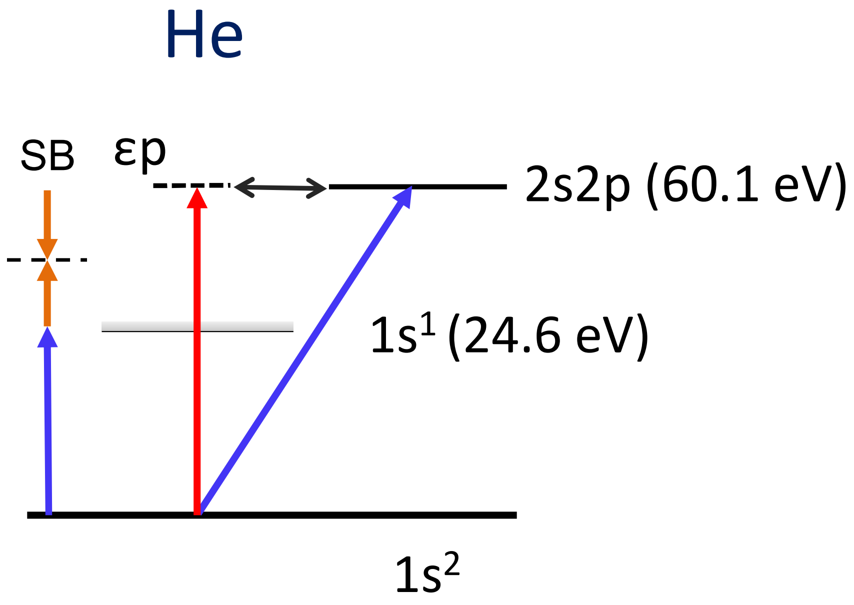

Resonance due to spin-orbit interaction

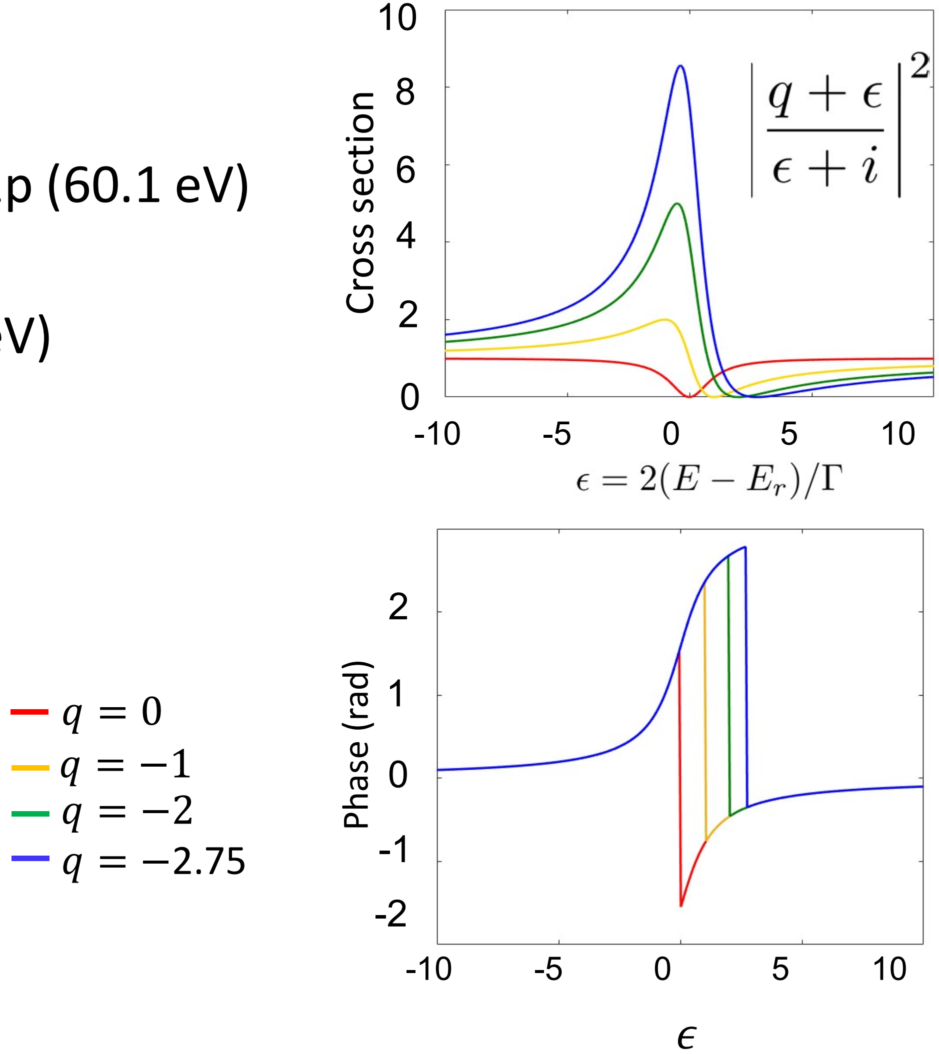
Giant dipole resonance
Electron correlation effects

# Non-resonnant and resonant photoionization



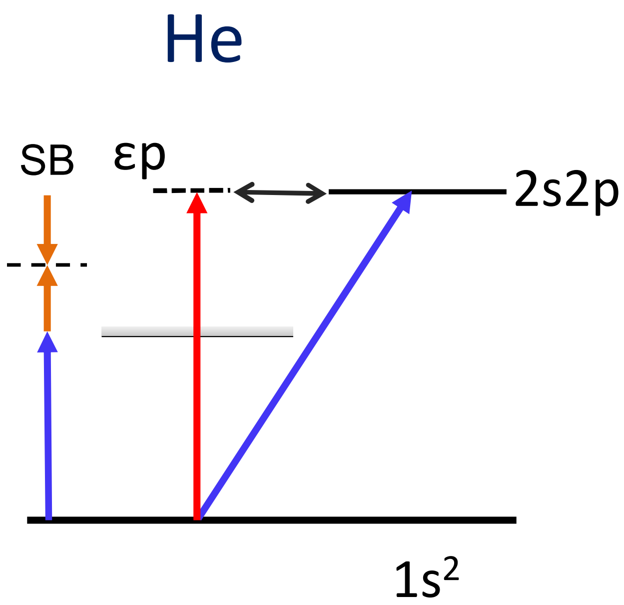
# Fano resonance

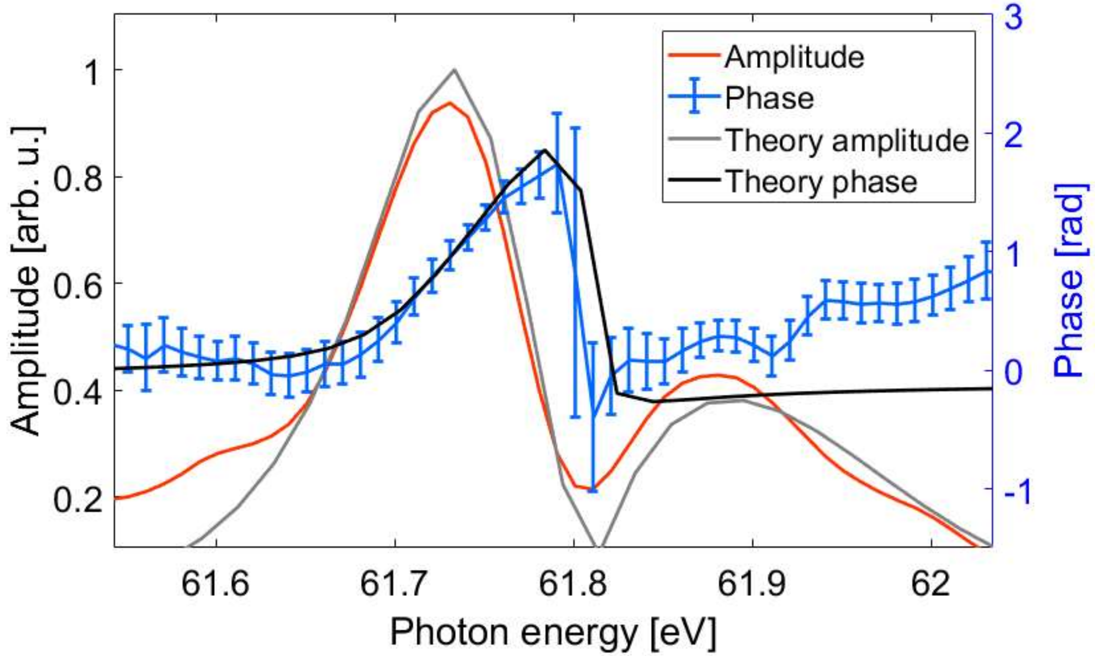



Fano, Phys. Rev. 1961
Ott et al., 2013, 2014
Kotur et al., Nat. Comm., 2016
Gruson et al., Science, 2016

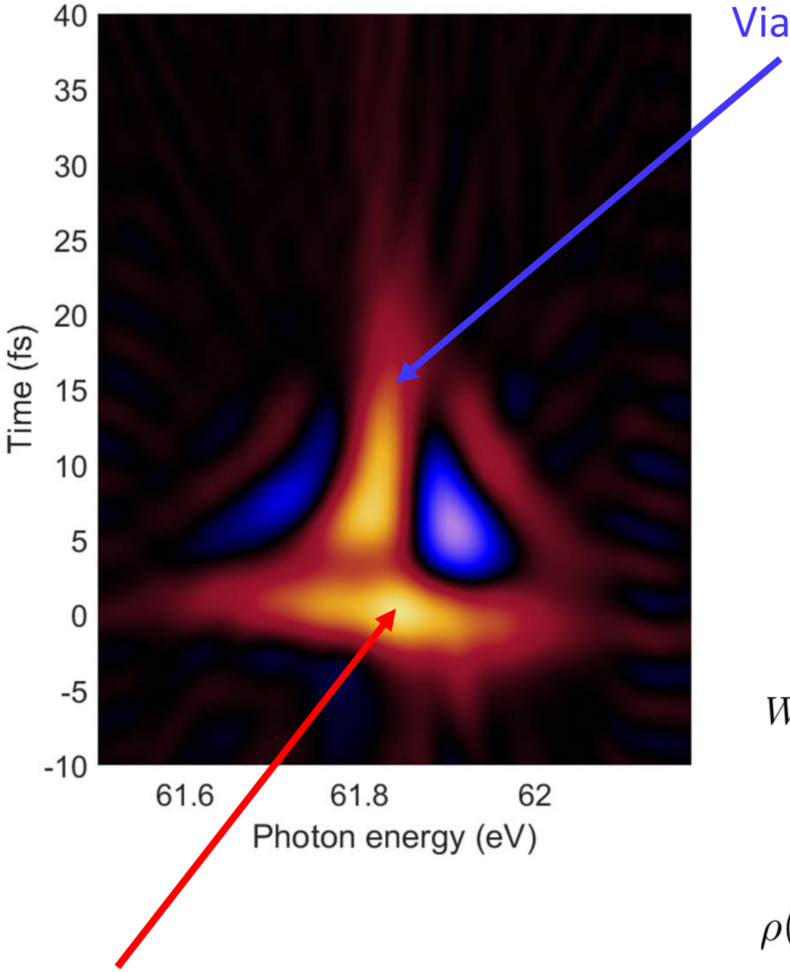
# High resolution RABBIT

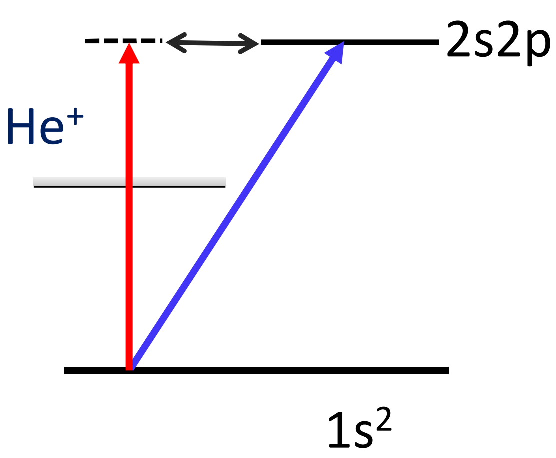



[3] D. Busto et al. *"Time-frequency representation of autoionization dynamics in helium"* J. Phys. B **51**, 044002 (2018)

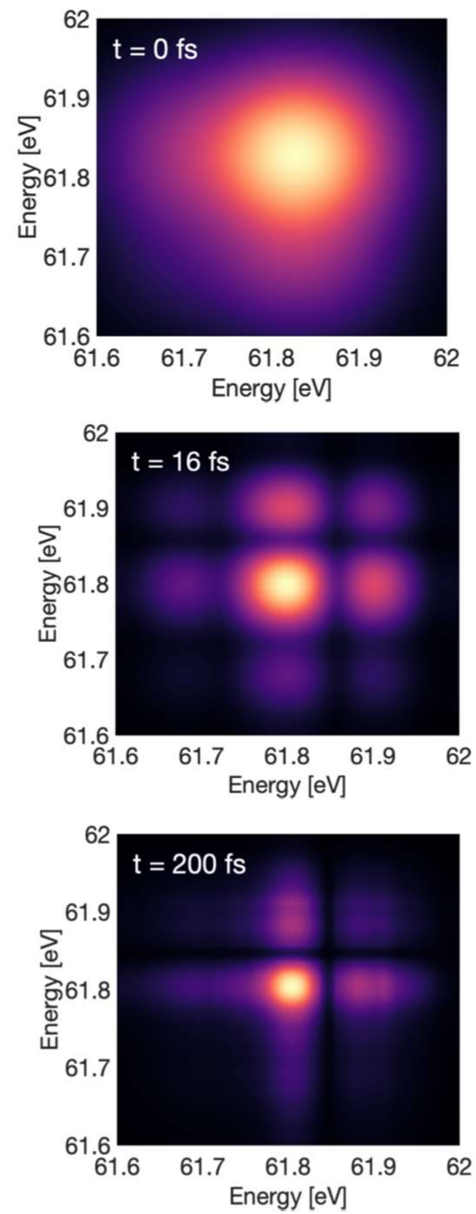
# Wigner distribution and density matrix




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

$$\rho(\varepsilon,\varepsilon') = \int_{-\infty}^{+\infty} W\left(\frac{\varepsilon+\varepsilon'}{2}, t\right) e^{it(\varepsilon-\varepsilon')} dt$$



# Summary

- **Attosecond pulses**
  - A brief historical introduction
  - Measurement of attosecond pulses [1]

- **Atomic Photoionization**
  - Photoionization time delays
  - Photoionization time delays Ne (attosecond metrology) [2]
  - Photoionization time delays Xe (atomic physics)
  - Resonant photoionization He (quantum optics) [3]

# Thank you for your attention

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