Lecture Series Buenos Aires

18-3-2024 until 22-3-2024

Lecture M6 – Attosecond atomic physics - 2

Max-Born-Institut

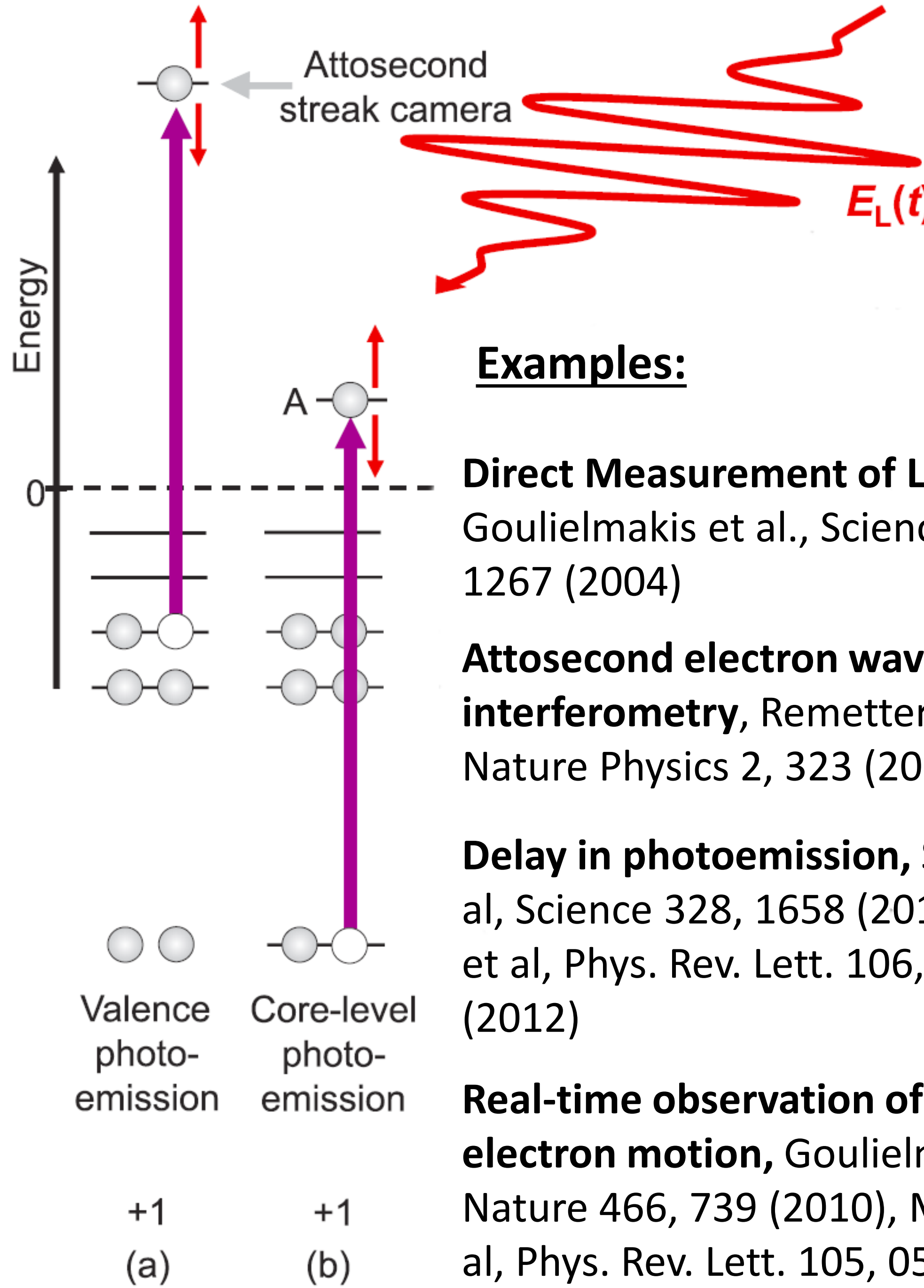
Marc Vrakking

marc.vrakking@mbi-berlin.de

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)

Attosecond atomic physics

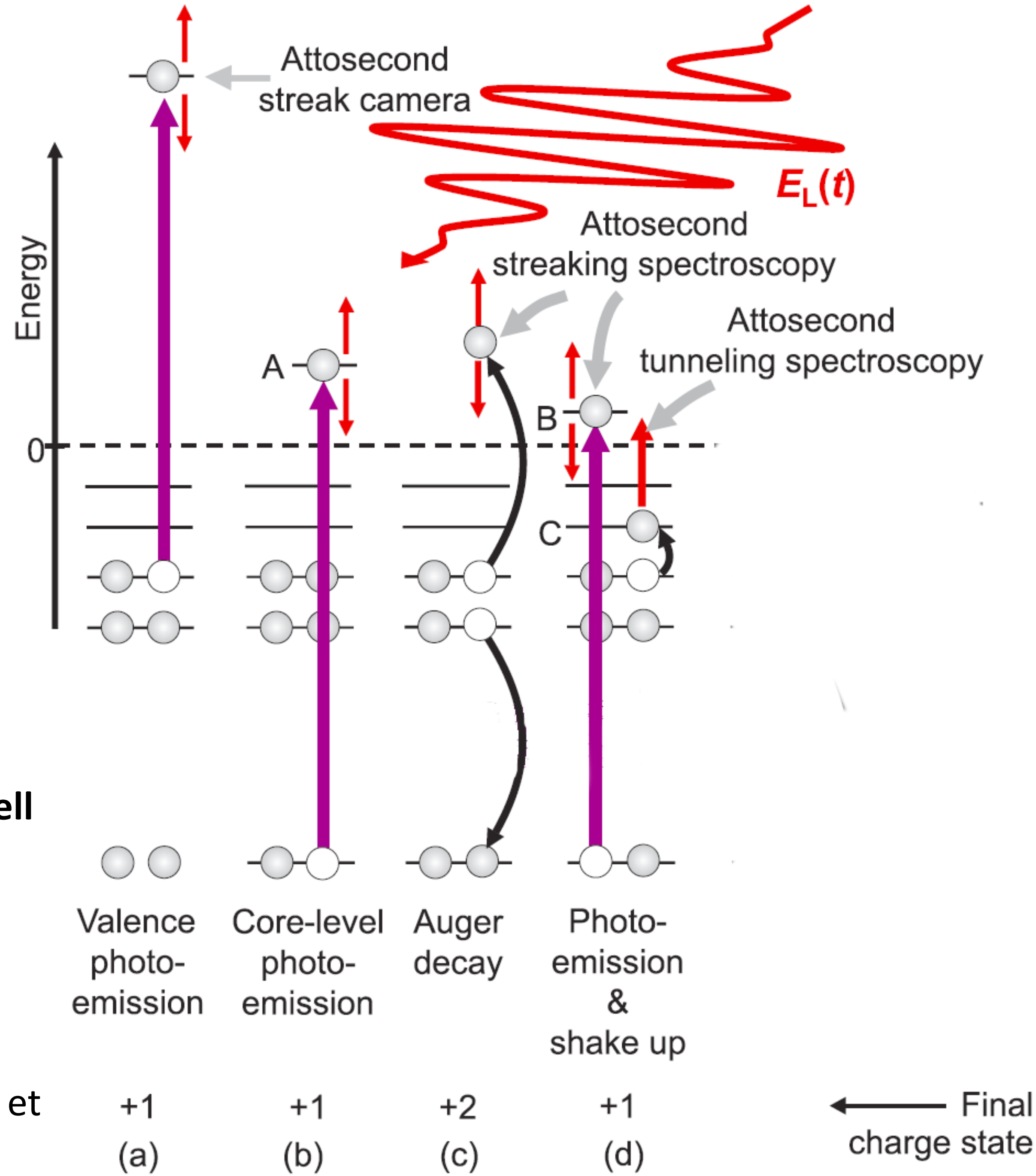
Multi-electron dynamics

- Auger decay
- Shake-up

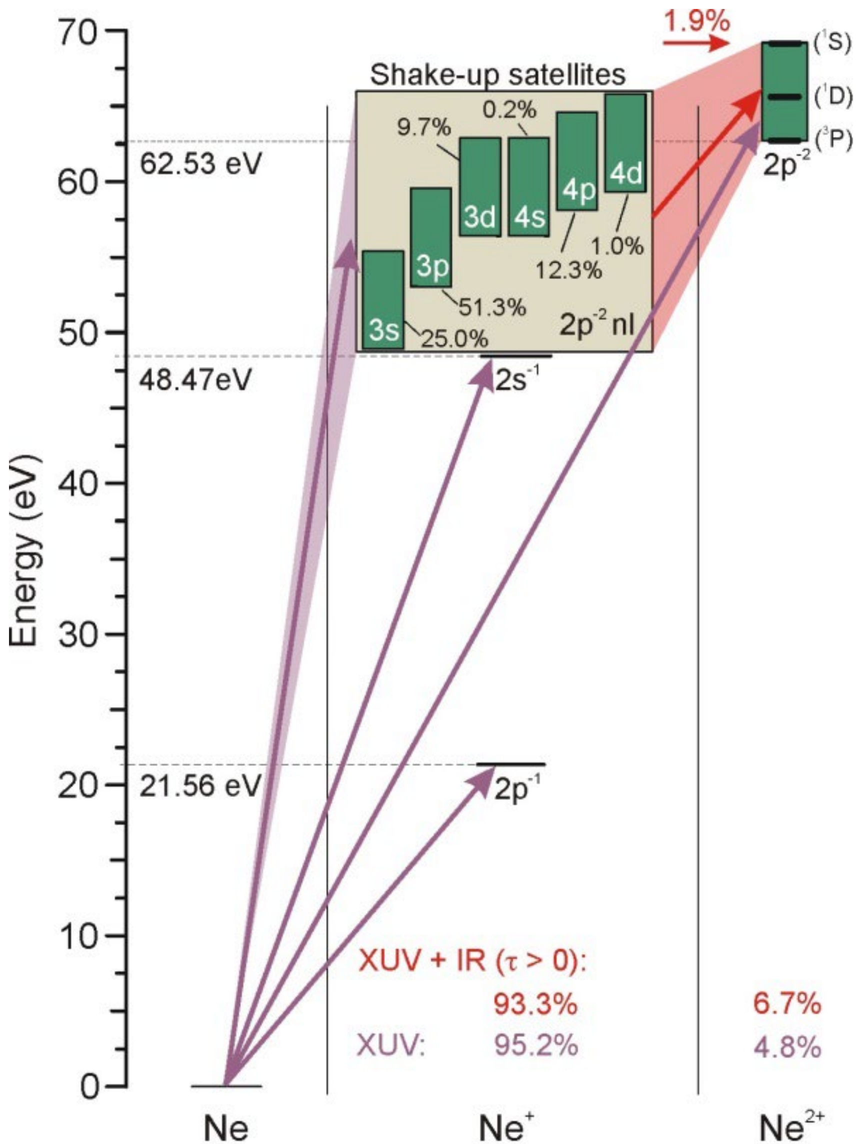
Example:

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

Attosecond real-time observation of electron tunneling in atoms, Uiberacker et al., Nature 446, 627 (2007)



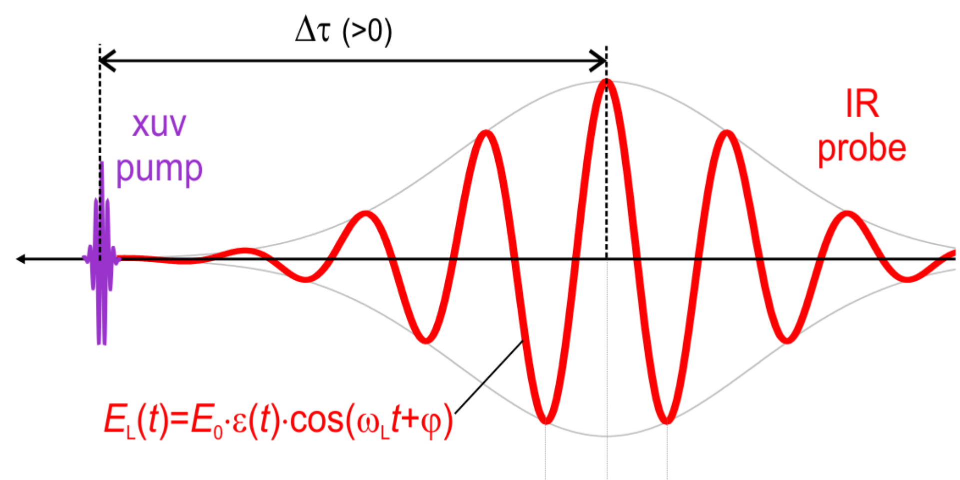
Time-resolving the tunneling process

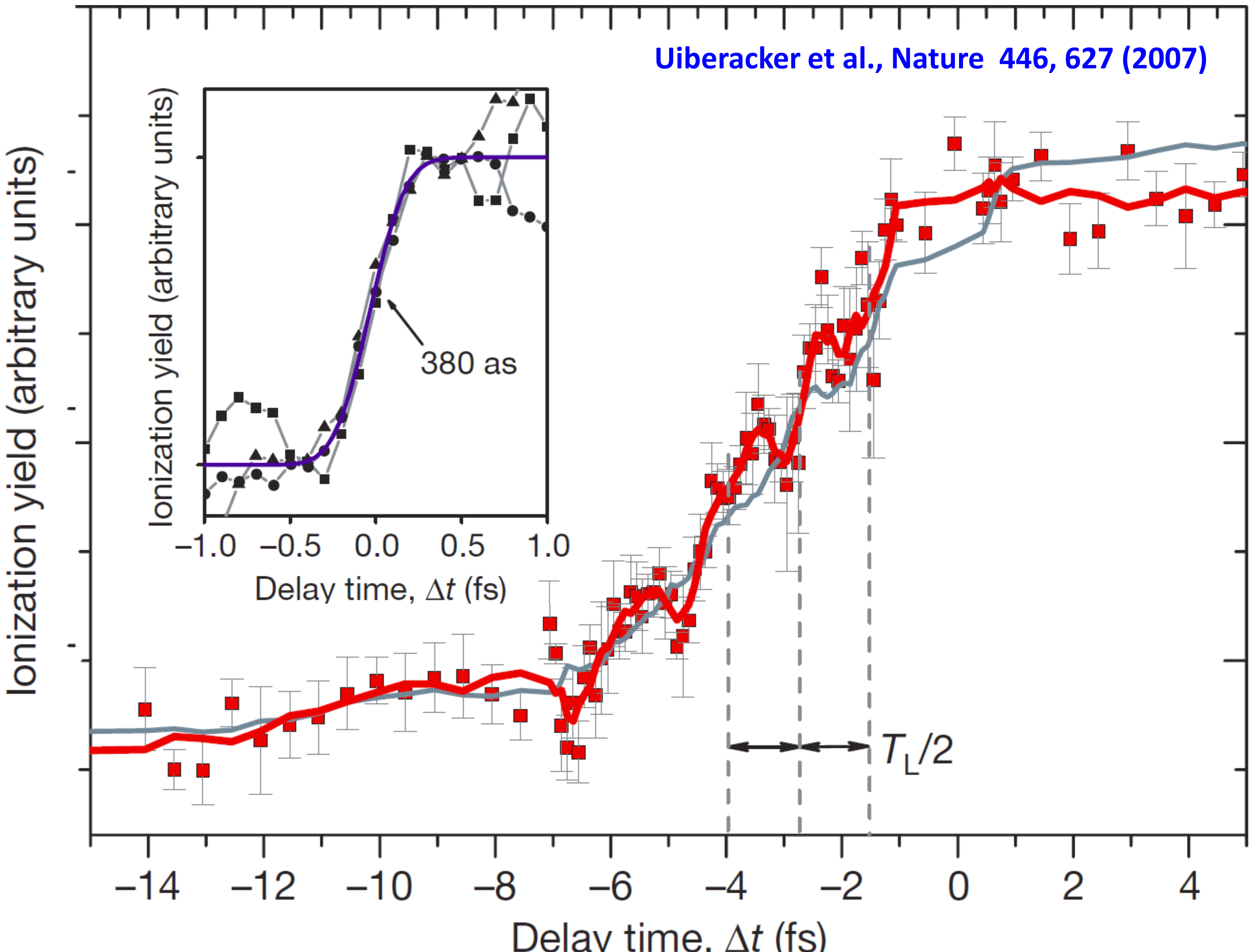


Uiberacker et al., Nature 446, 627 (2007)

Photoionization of Ne at 90 eV leads to both the removal of a 2p valence electron and shake-up of a second 2p electron into a Rydberg state

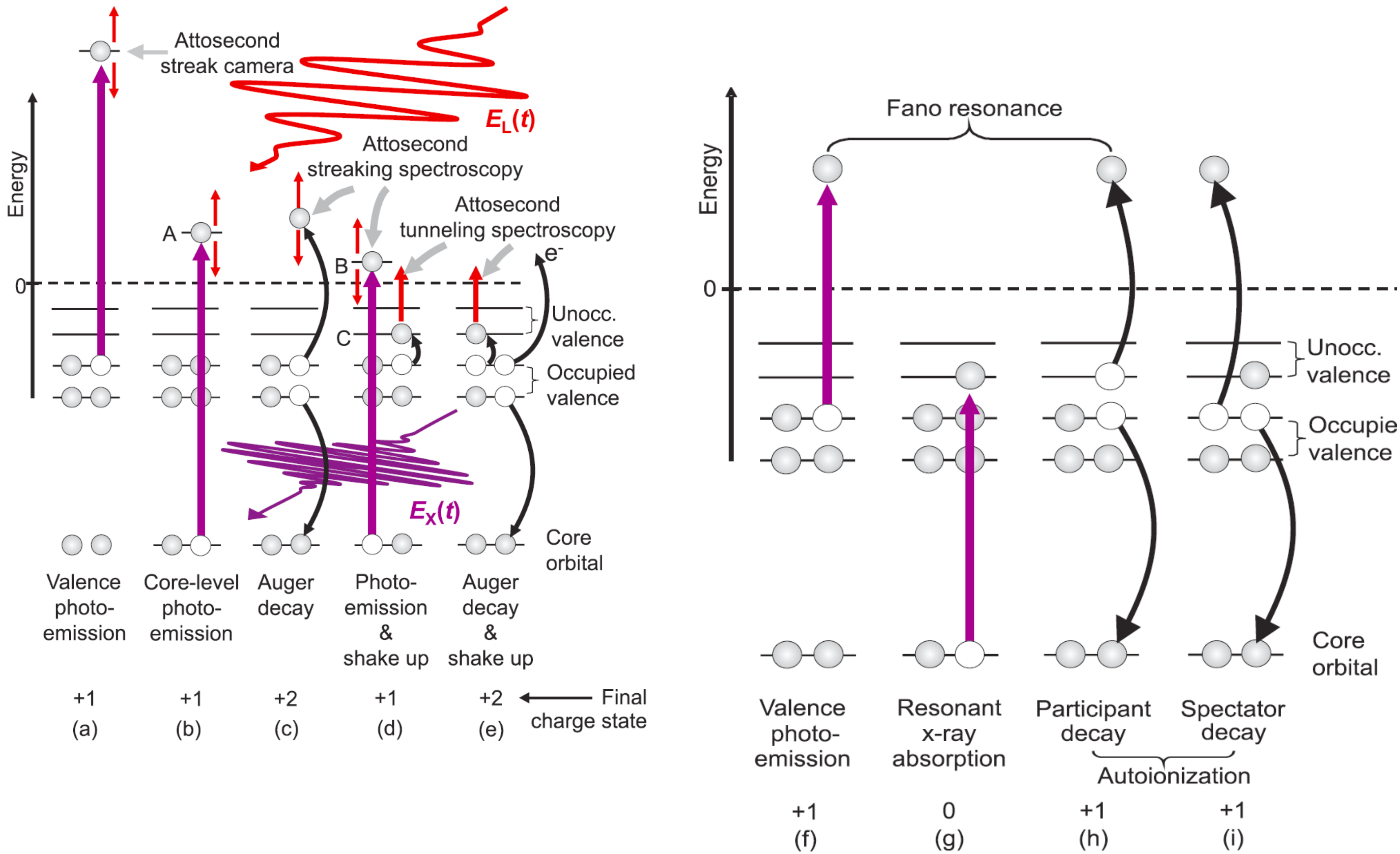
The shake-up electron can be ionized by a low-order NIR multi-photon ionization process





Uiberacker et al., Nature 446, 627 (2007)
Ionization yield (arbitrary units)
380 as
−1.0
−0.5
0.0
0.5
1.0
Delay time, Δt (fs)
T_L/2
−14
−12
−10
−8
−6
−4
−2
0
2
4
Delay time, Δt (fs)

Attosecond atomic physics



Increasingly complex atomic physics problems addressed by attosecond pump-probe spectroscopy

Transient absorption

The energy that is removed from the laser field by an individual atom/molecule is

$$\Delta E = \int_{-\infty}^{\infty} z(t) \frac{\partial \varepsilon(t)}{\partial t} dt$$

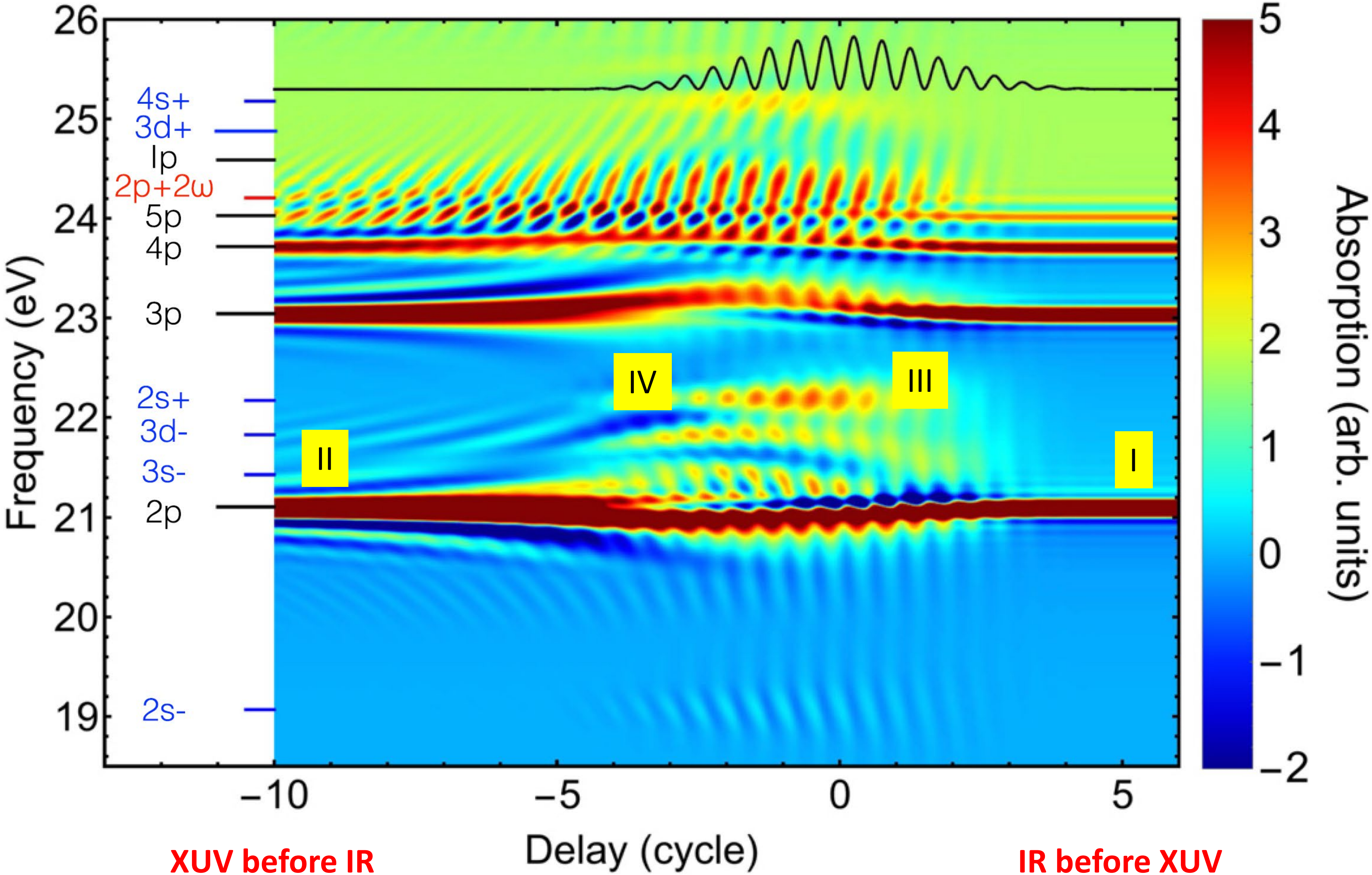
where z(t) is the expectation value of the position of the interacting electron (= - dipole)

The ratio between the energy absorbed per atom per unit frequency and the incoming spectral flux (~optical density in experiment) is given by

$$\sigma(\omega) = g 4\pi\alpha\omega \, Im \left\{ \frac{\tilde{d}(\omega)}{\tilde{\varepsilon}(\omega(} \right\}$$

Wu et al., J. Phys. B, 49, 062003 (2016)

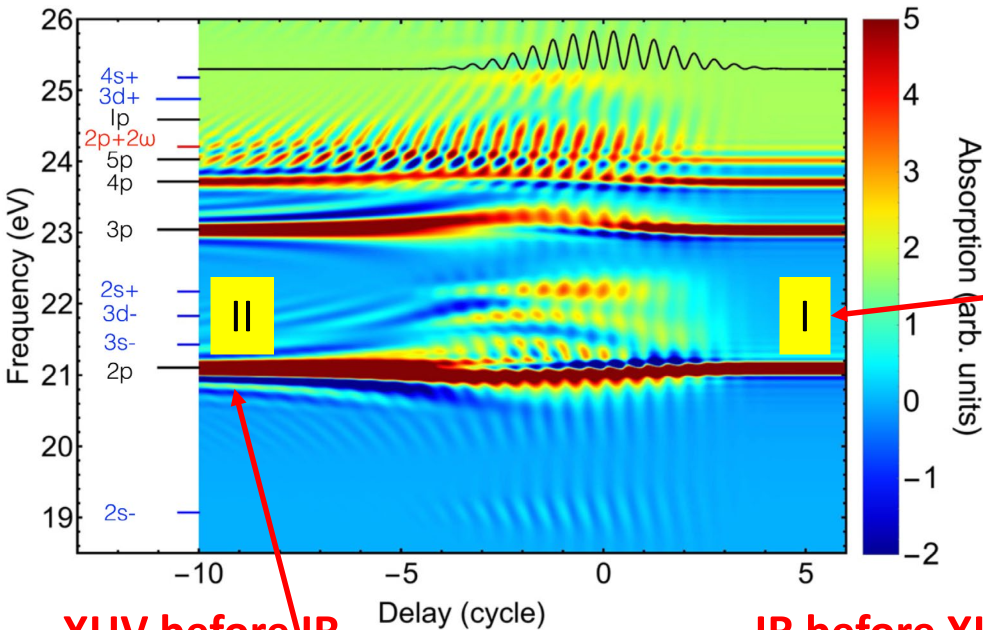
Transient absorption in He



XUV before IR

IR before XUV

Transient absorption in He



XUV before IR

IR before XUV

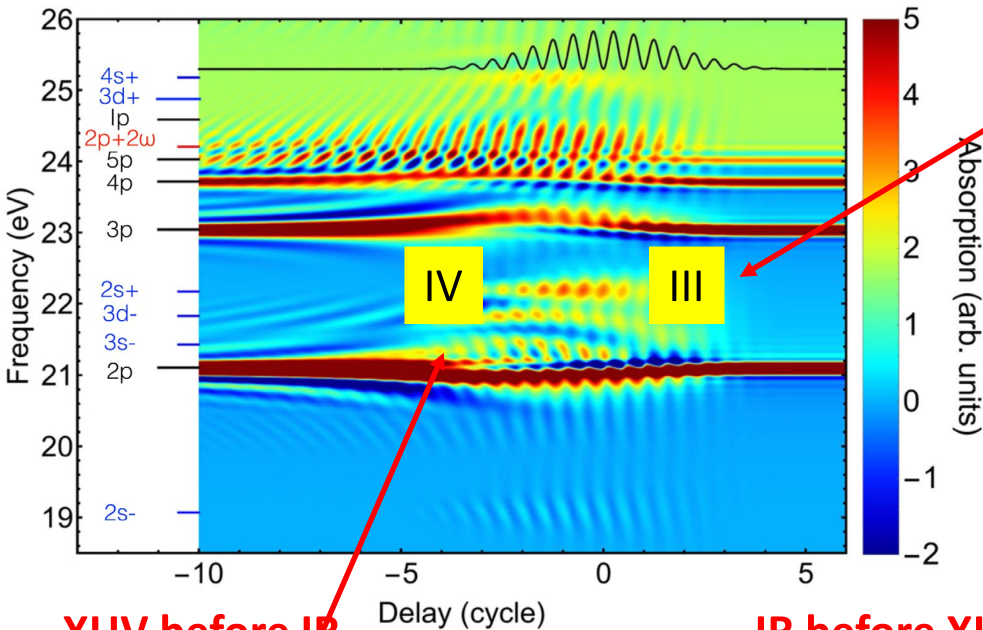
When the IR comes before the XUV, then it does not influence the absorption process

The XUV induces a dipole that re-radiates out of phase with the incoming radiation (free induction decay) → Lorentzian absorption profile

When the IR comes after the XUV, then it couples the excited resonance to other levels and thus interupts and/or perturbs the free induction decay
In the transient absorption spectrum we see hyperbolic fringes → constructive interference when $\delta\omega \times \tau = n2\pi$

Wu et al., J. Phys. B, 49, 062003 (2016)

Transient absorption in He

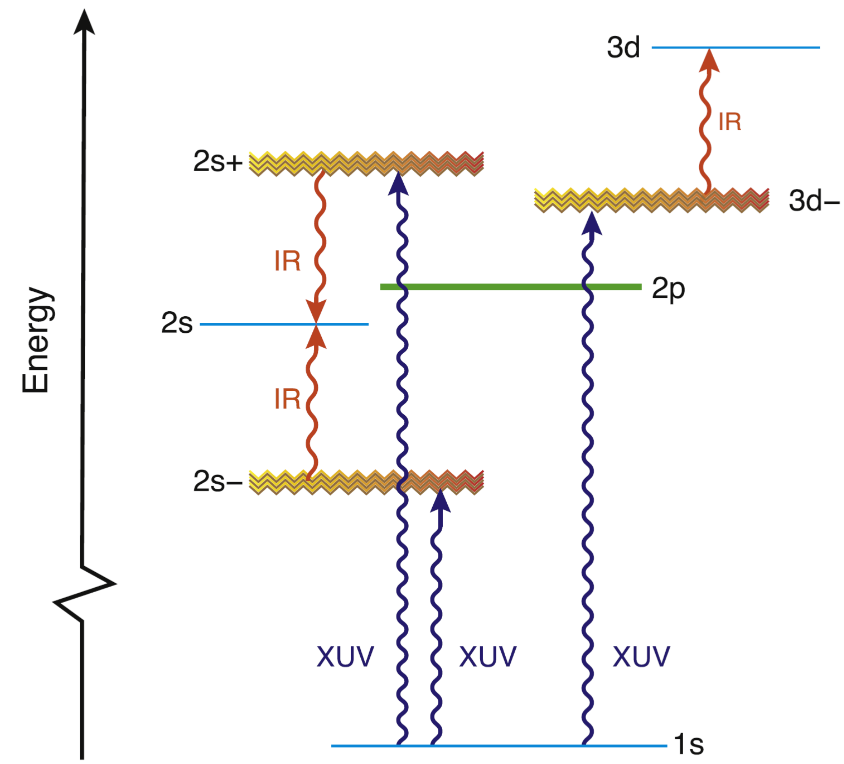


XUV before IR

IR before XUV

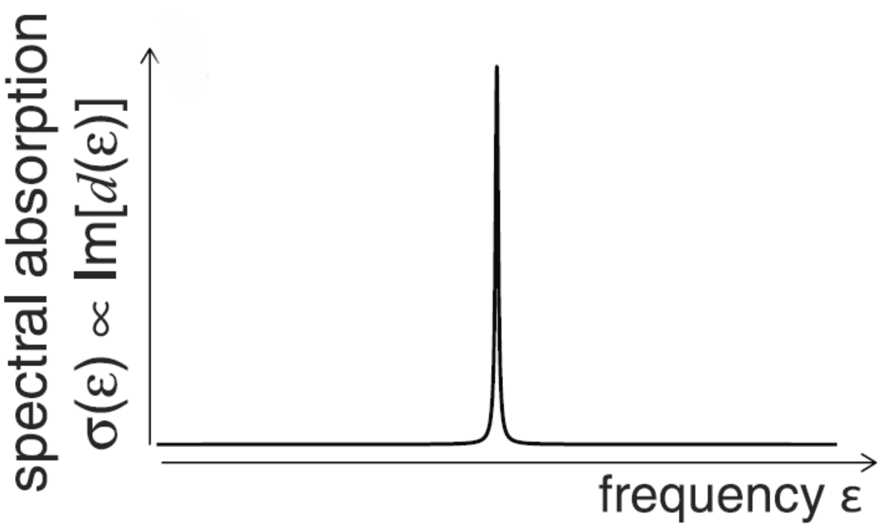
During overlap of the XUV and IR, we see the formation of Light-Induced States (LIS)

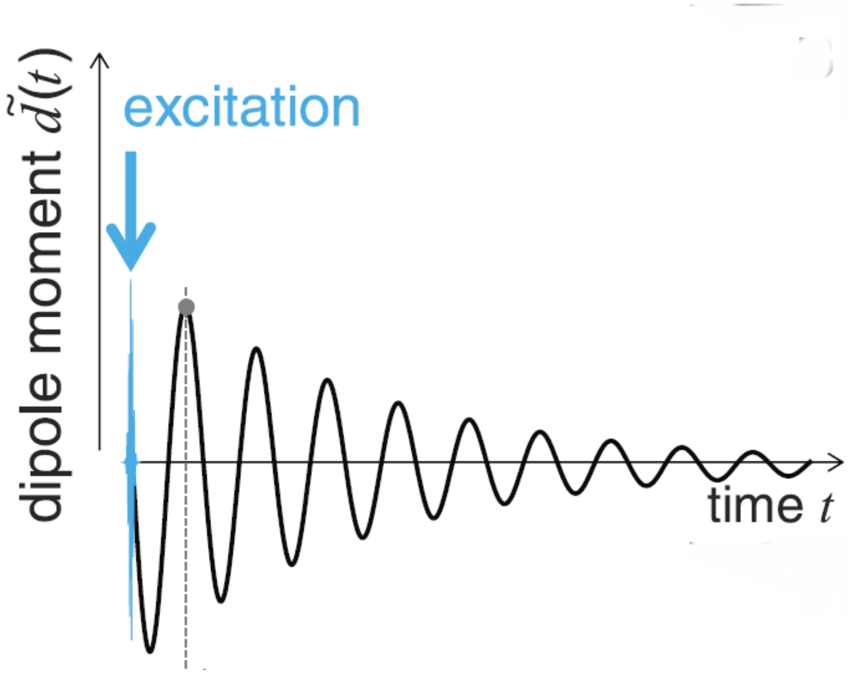
Many of the contributions that we see during XUV/IR time-overlap show RABBITT-like 2ω-oscillations → two-path interferences



Wu et al., J. Phys. B, 49, 062003 (2016)

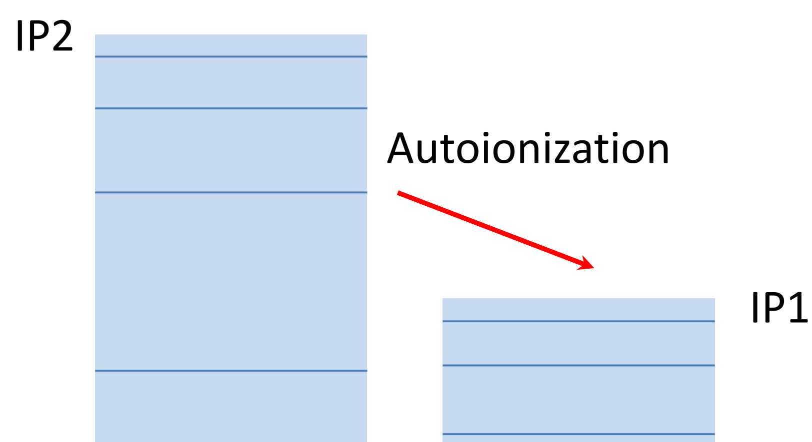
Fano Resonances in Transient Absorption



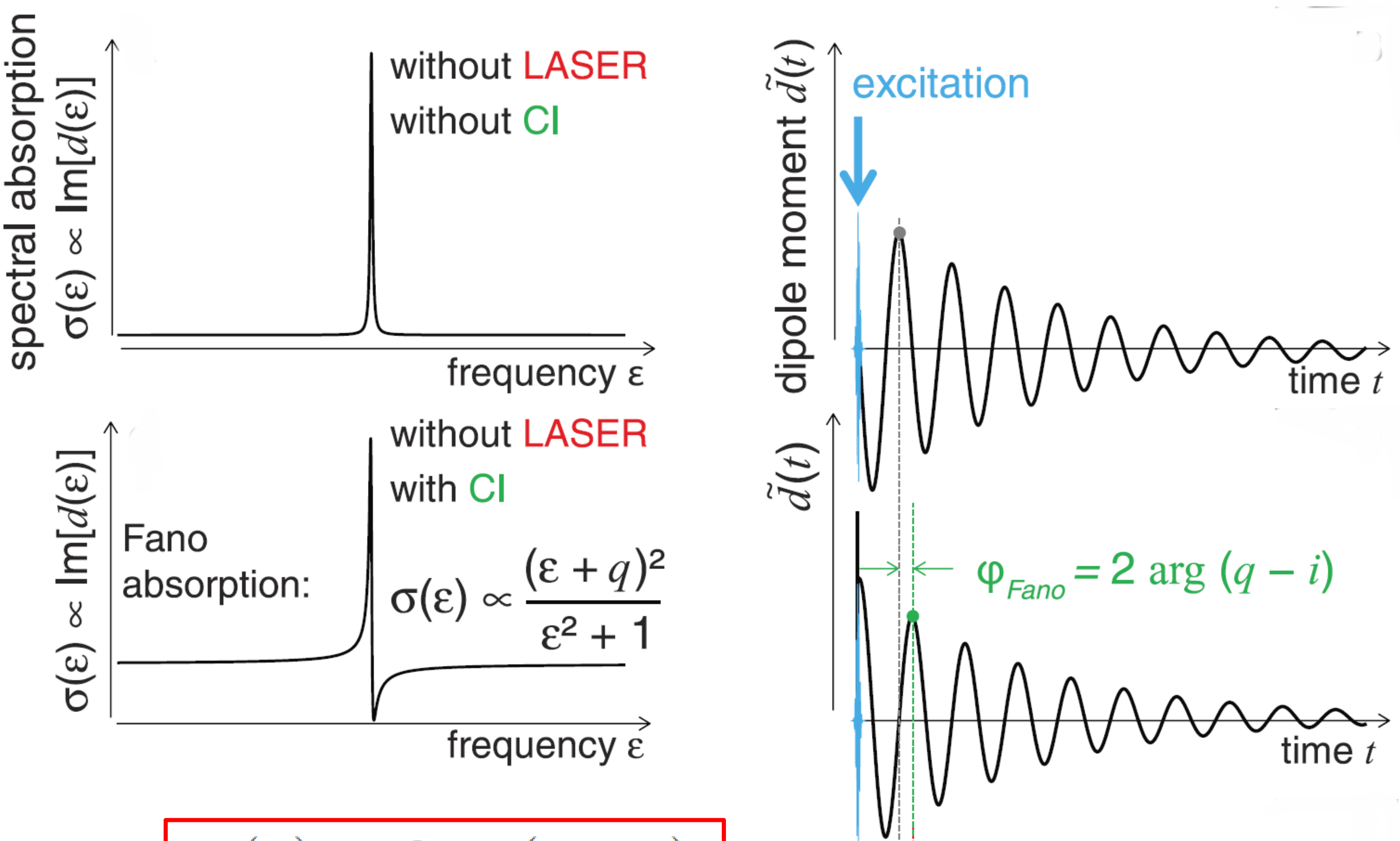


Now consider the case where a resonance is coupled to a continuum, e.g. Auto-ionizing state → Fano-profile

$$\sigma_{\text{Fano}}(E) = \sigma_0 \frac{(q+\varepsilon)^2}{1+\varepsilon^2}, \varepsilon = \frac{E-E_0}{\hbar(\Gamma/2)}$$

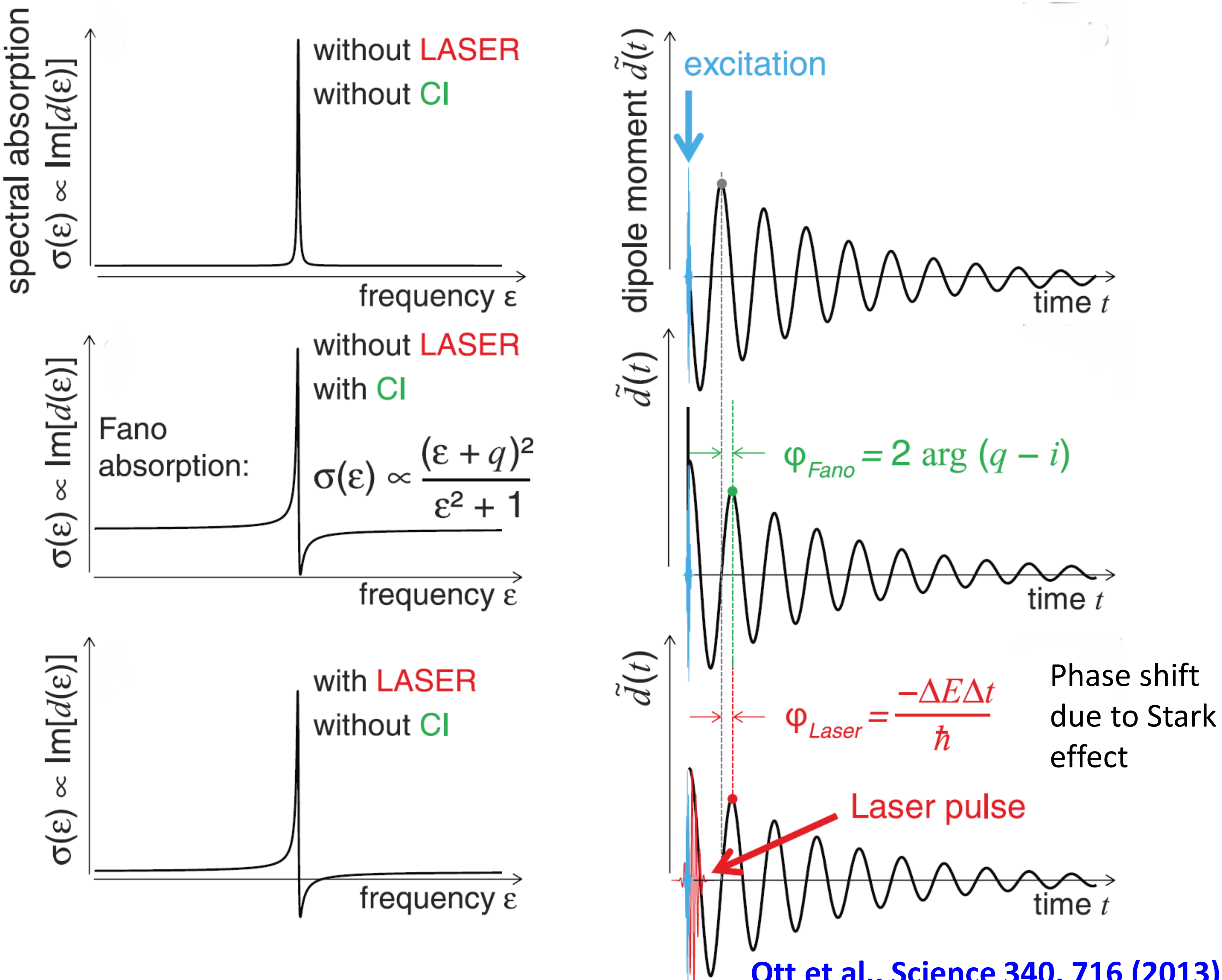


Fano Resonances in Transient Absorption

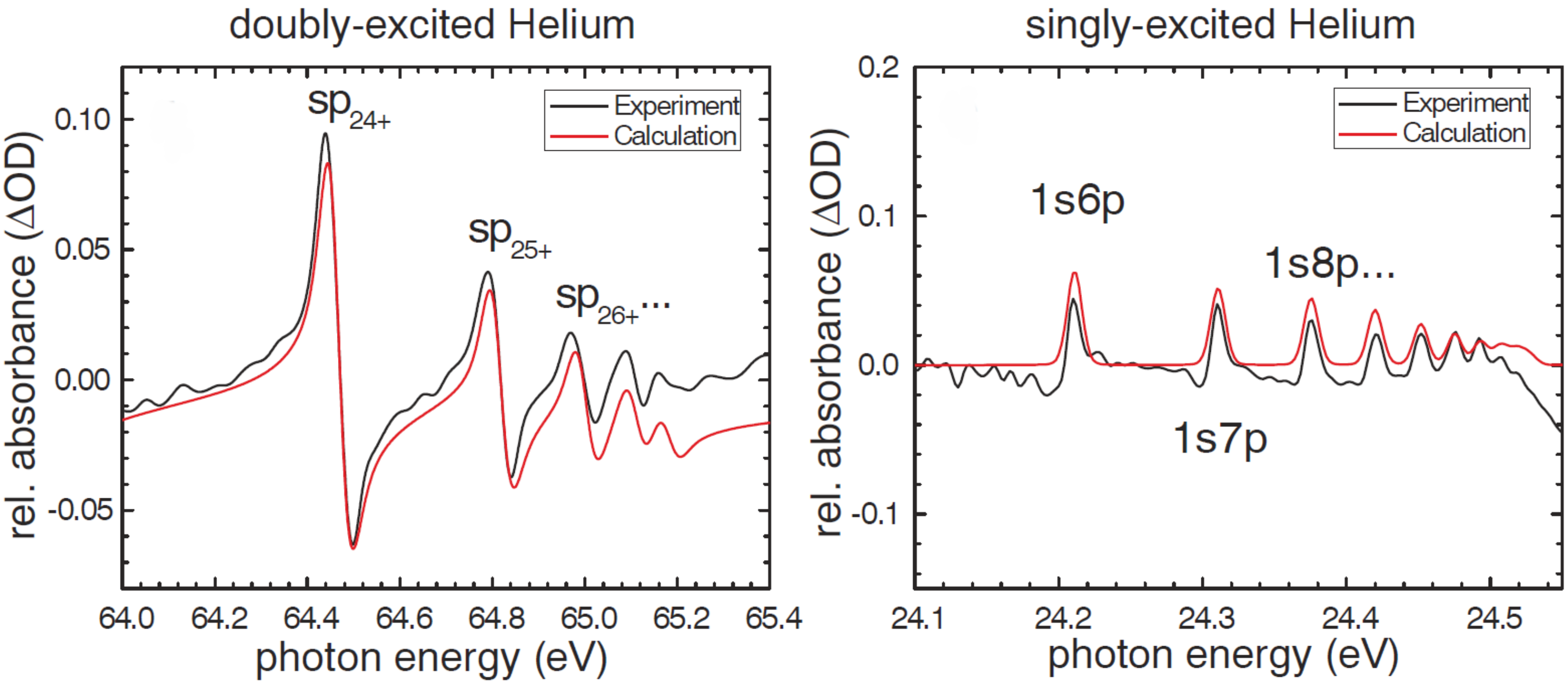


$$\varphi(q) = 2\arg(q - i)$$

Ott et al., Science 340, 716 (2013)

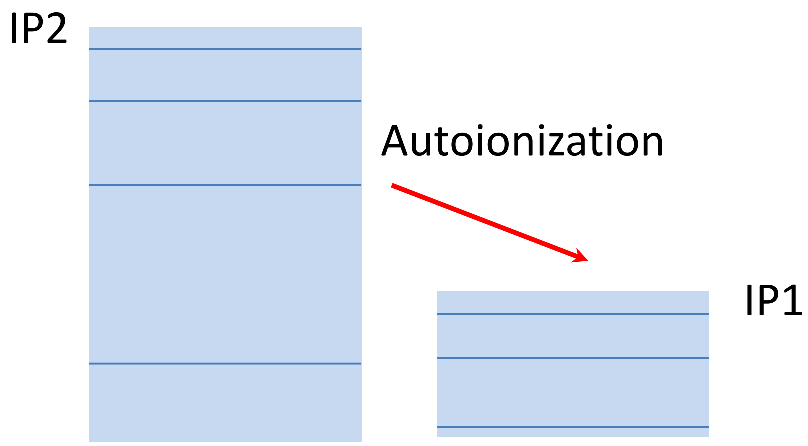


Ott et al., Science 340, 716 (2013)

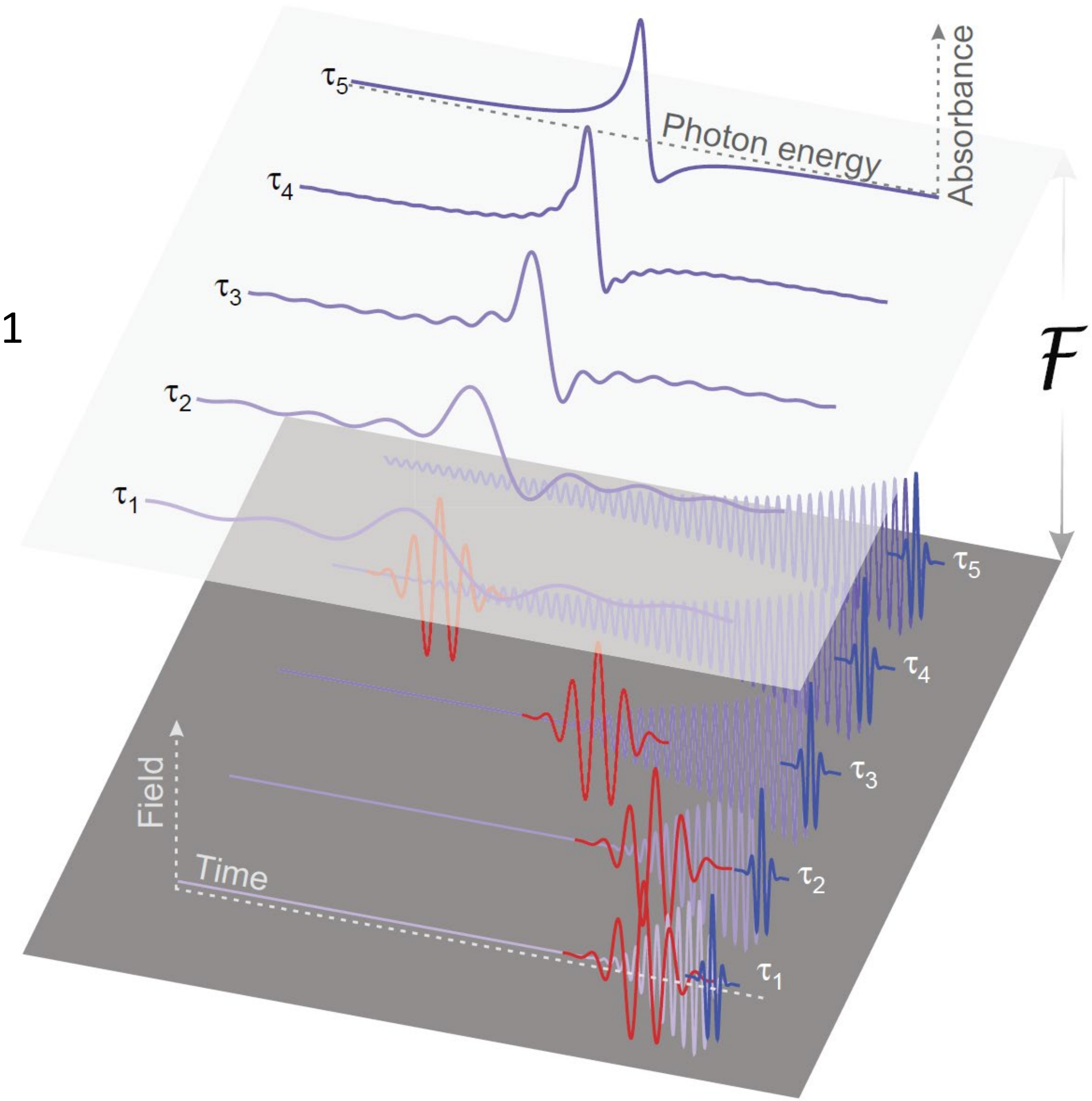

doubly-excited Helium
singly-excited Helium
Experiment
Calculation
sp_{24+}
sp_{25+}
sp_{26+}...
1s6p
1s7p
1s8p...
rel. absorbance (ΔOD)
photon energy (eV)
0.10
0.05
0.00
-0.05
64.0
64.2
64.4
64.6
64.8
65.0
65.2
65.4
0.2
0.1
0.0
-0.1
24.1
24.2
24.3
24.4
24.5

Without IR

The build-up of a Fano Resonance

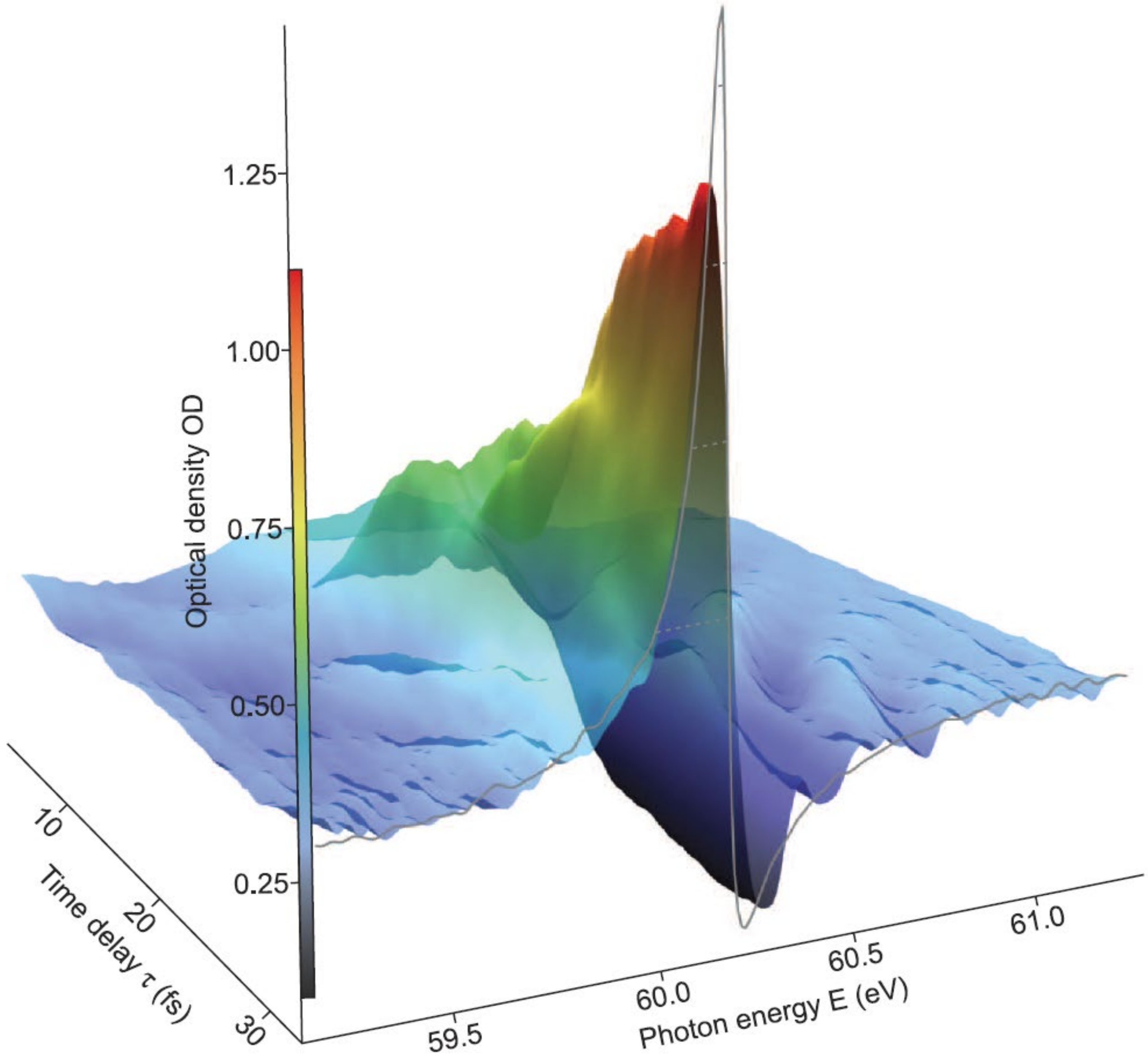


Autoionization can be prevented by exposure to a strong ionizing laser field



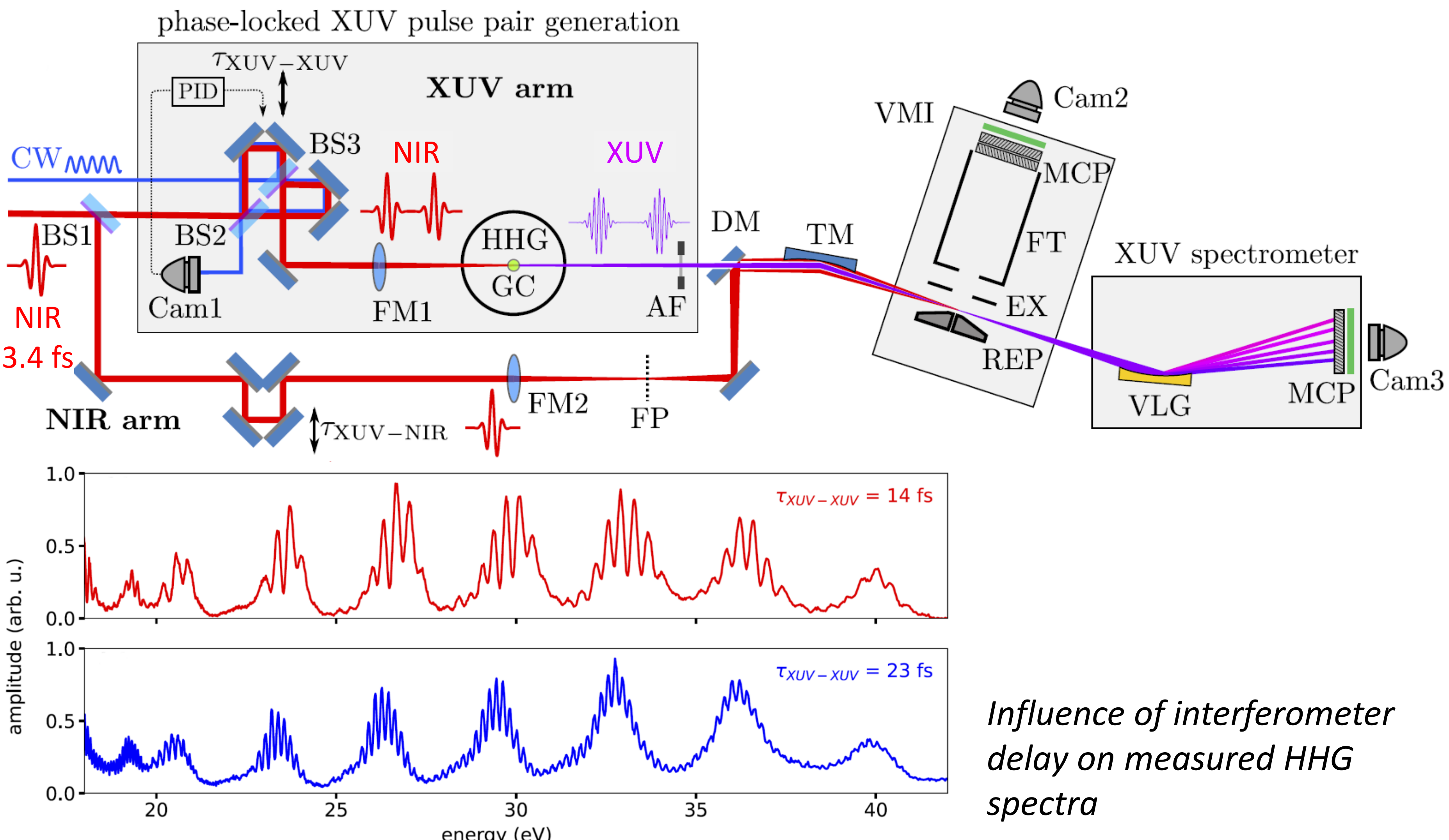
Kaldun et al., Science 354, 738 (2016)

The build-up of a Fano Resonance



Kaldun et al., Science 354, 738 (2016)

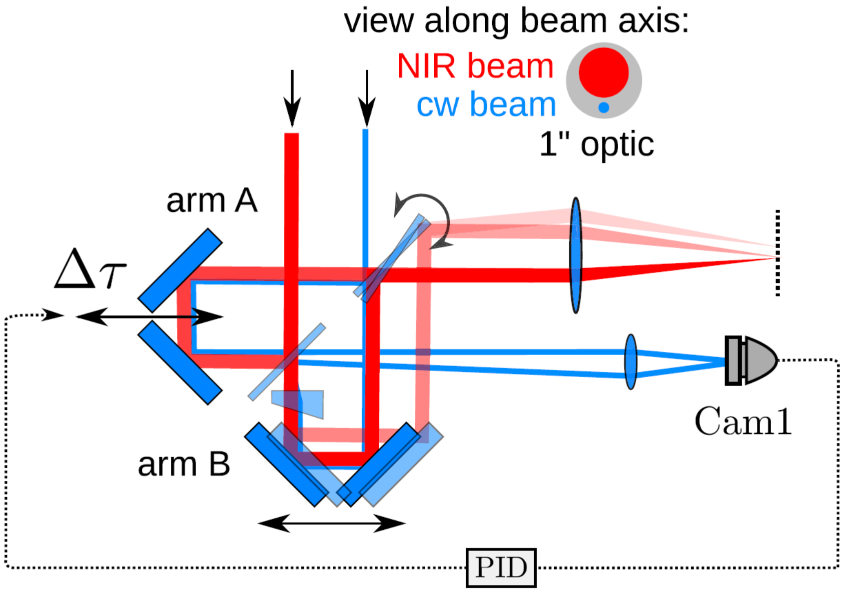
Attosecond FT Spectroscopy



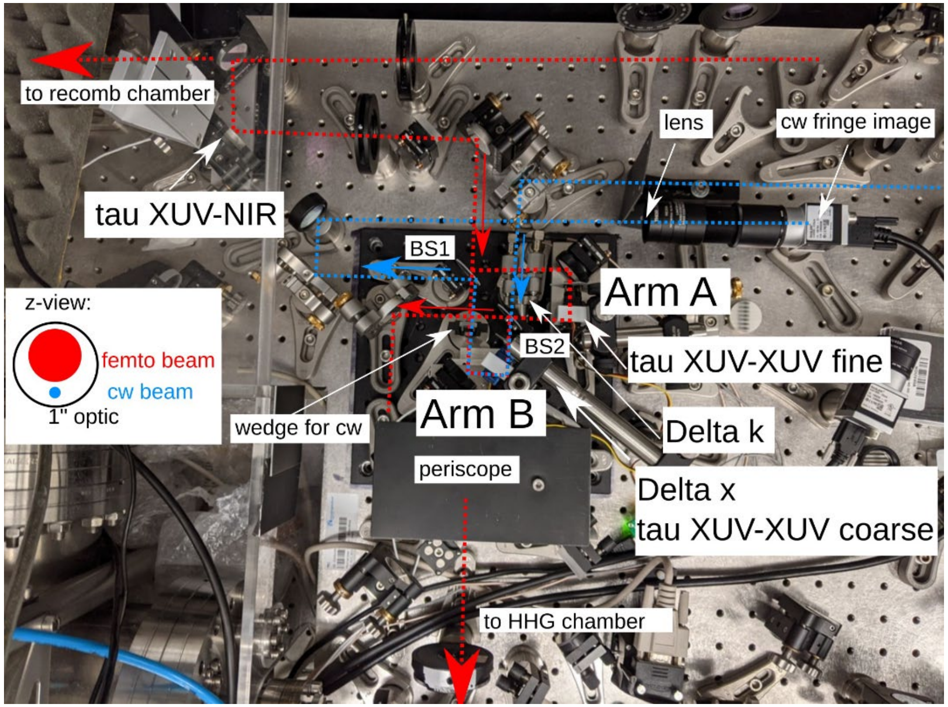
Influence of interferometer delay on measured HHG spectra

L.-M. Koll et al, Opt. Expr. 30, 7082 (2022)

Attosecond FT Spectroscopy



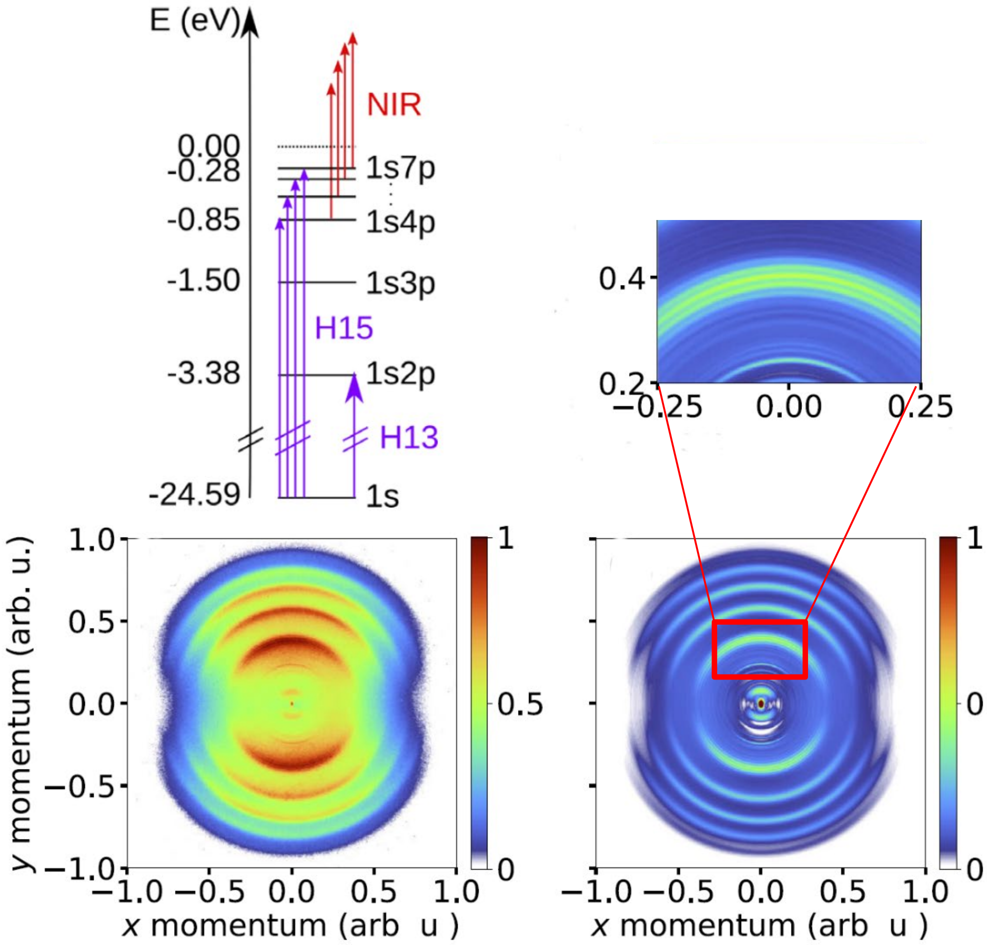
Split the IR beam in the XUV-arm of the Mach Zehnder interferometer, where attosecond pulses are produced by HHG



Actively and passively stabilized Mach Zehnder interferometer

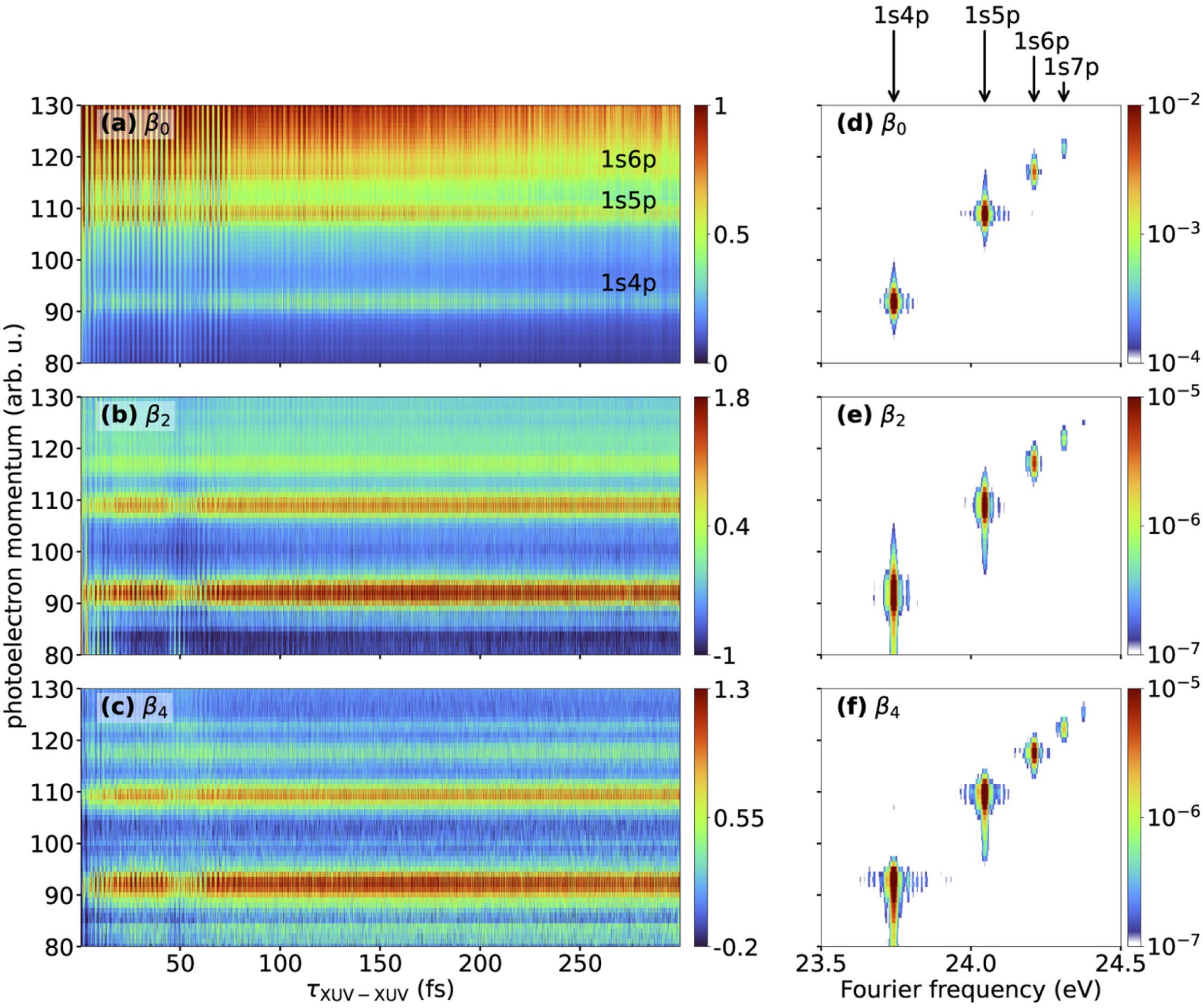
L.-M. Koll et al, Opt. Expr. 30, 7082 (2022)

Test: He FT Spectroscopy

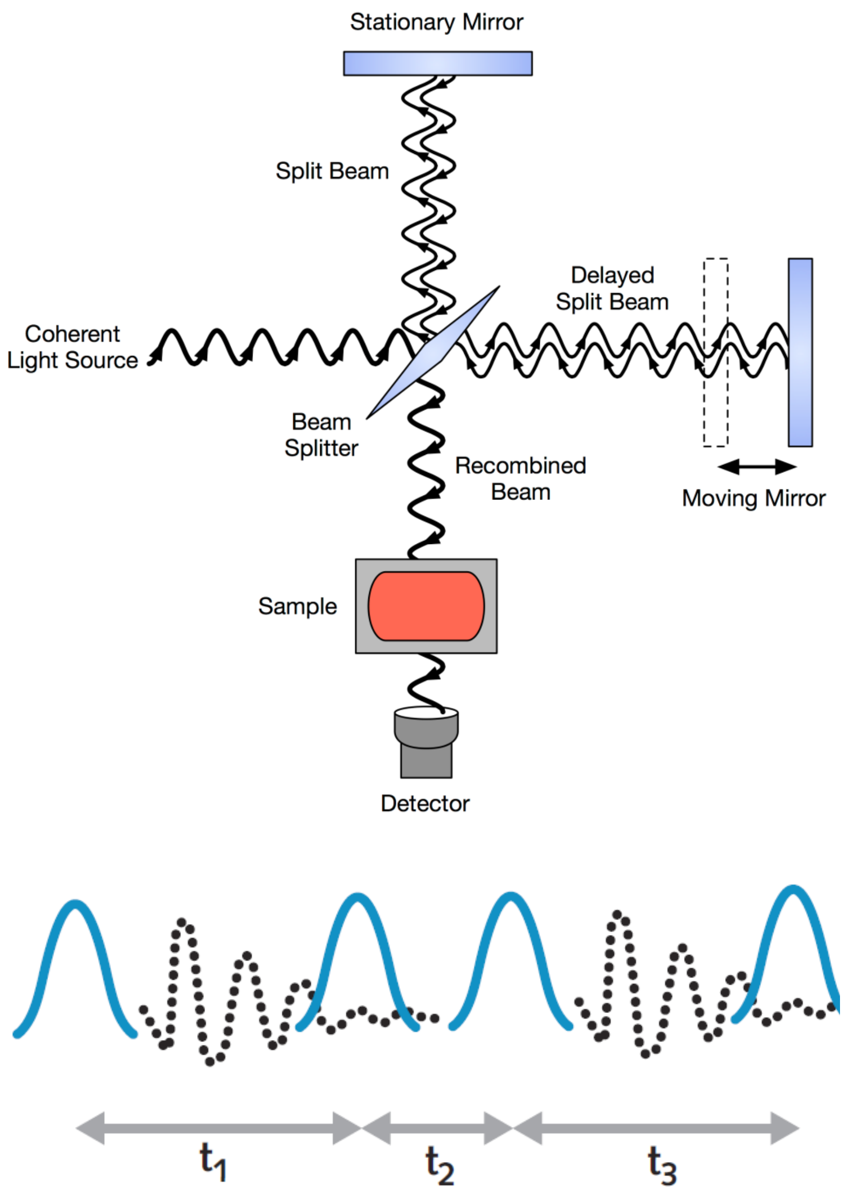


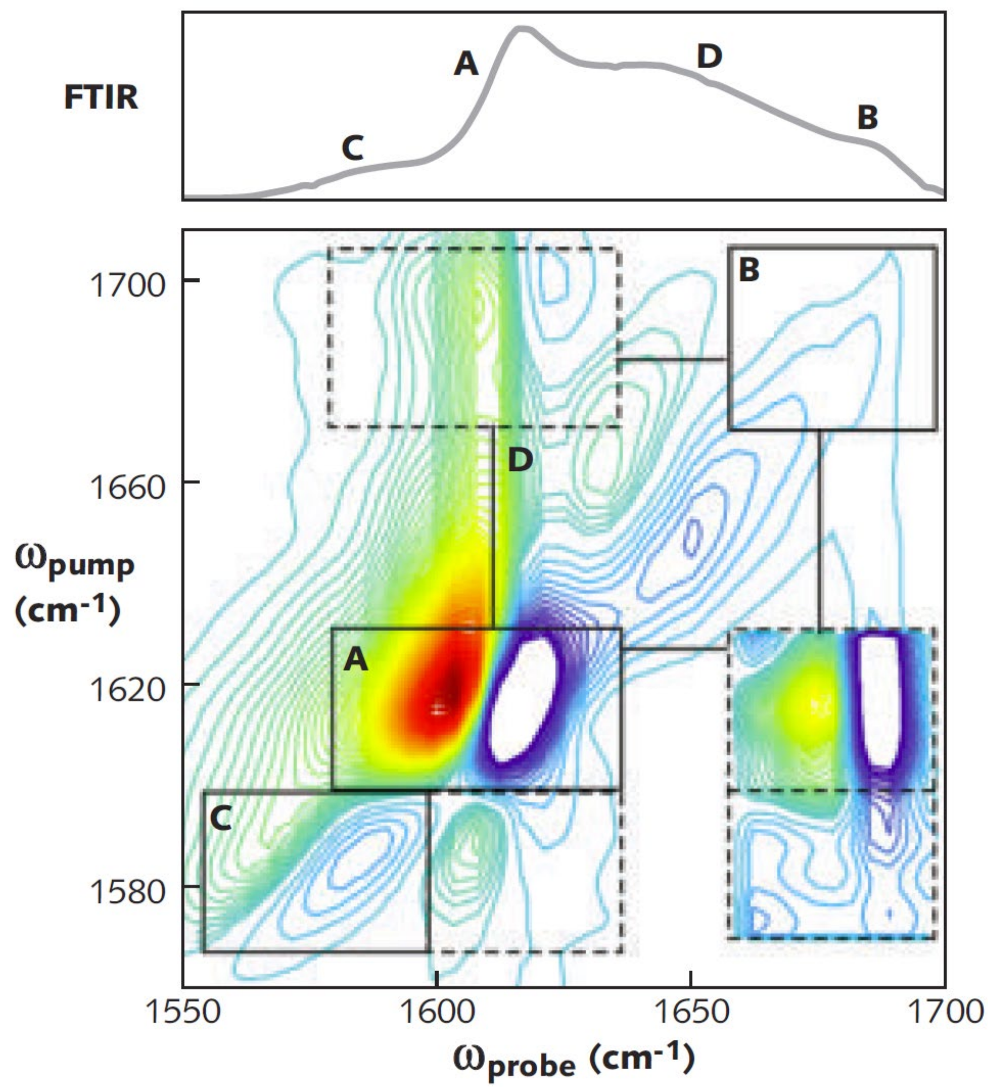
- Photoexcitation of He by XUV pulse pair
- Time-delayed ionization by narrowband IR pulse
- Velocity map imaging detection of resulting photoelectrons

L.-M. Koll et al, Opt. Expr. 30, 7082 (2022)


1s4p
1s5p
1s6p
1s7p
(a) β_0
(b) β_2
(c) β_4
(d) β_0
(e) β_2
(f) β_4
1s6p
1s5p
1s4p
photoelectron momentum (arb. u.)
$\tau_{XUV-XUV}$ (fs)
Fourier frequency (eV)
130
120
110
100
90
80
50
100
150
200
250
23.5
24.0
24.5
1
0.5
0
1.8
0.4
-1
1.3
0.55
-0.2
10^{-2}
10^{-3}
10^{-4}
10^{-5}
10^{-6}
10^{-7}

2D FT absorption spectroscopy





PNAS 2007, themed issue Multidimensional Spectroscopy

Useful materials for further reading (strong field ionization):

L. DiMauro and P. Agostini, Adv. At. Mol. And Opt. Physics 35, 79 (1995)

Wu et al., J. Phys. B: At. Mol. Opt. Phys. 49 (2016) 062003 (29pp)

M. Ivanov and F. Krausz, Reviews of Modern Physics 81, 163-234 (2009)