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

Attosecond Pump-Probe Spectroscopy - part 2

Max-Born-Institut

Marc Vrakking

marc.vrakking@mbi-berlin.de

Lecture at FU Berlin (winter semester 2017/18)

20-10-2017	Lecture 1: Introduction + Lasers 1
27-10-2017	Lecture 2: Lasers 2
3-11-2017	Lecture 3: Lasers 3 + **WG Laser Physics**
10-11-2017	Lecture 4: Pump-probe spectroscopy
17-11-2017	Lecture 5: Pump-probe spectroscopy 2 + **WG Pump-probe spectroscopy**
24-11-2017	Lecture 6: Atoms in strong laser fields –1
1-12-2017	Lecture 7: Atoms in strong laser fields – 2 + **WG VMI**
8-12-2017	Lecture 8: Atoms in strong laser fields – 3
15-12-2017	Lecture 9: High harmonic generation + **WG SFA**
12-1-2018	Lecture 10: Attosecond pulse production and characterization
12-1-2018	Lecture 11: Attosecond experiments in atoms
19-1-2018	Lecture 12: Molecules in strong laser fields - 1
26-1-2018	Lecture 13: Molecules in strong laser fields – 2 + **WG Floquet**
2-2-2018	Lecture 14: Attosecond experiments in molecules
9-2-2018	Lecture 15: Attosecond experiments in molecules (continued) + **WG Alignment**
16-2-2018	Lecture 16: Attosecond experiments: surfaces and nanoparticles + **labtour**

See http://staff.mbi-berlin.de/vrakking/lecture/index.html

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)

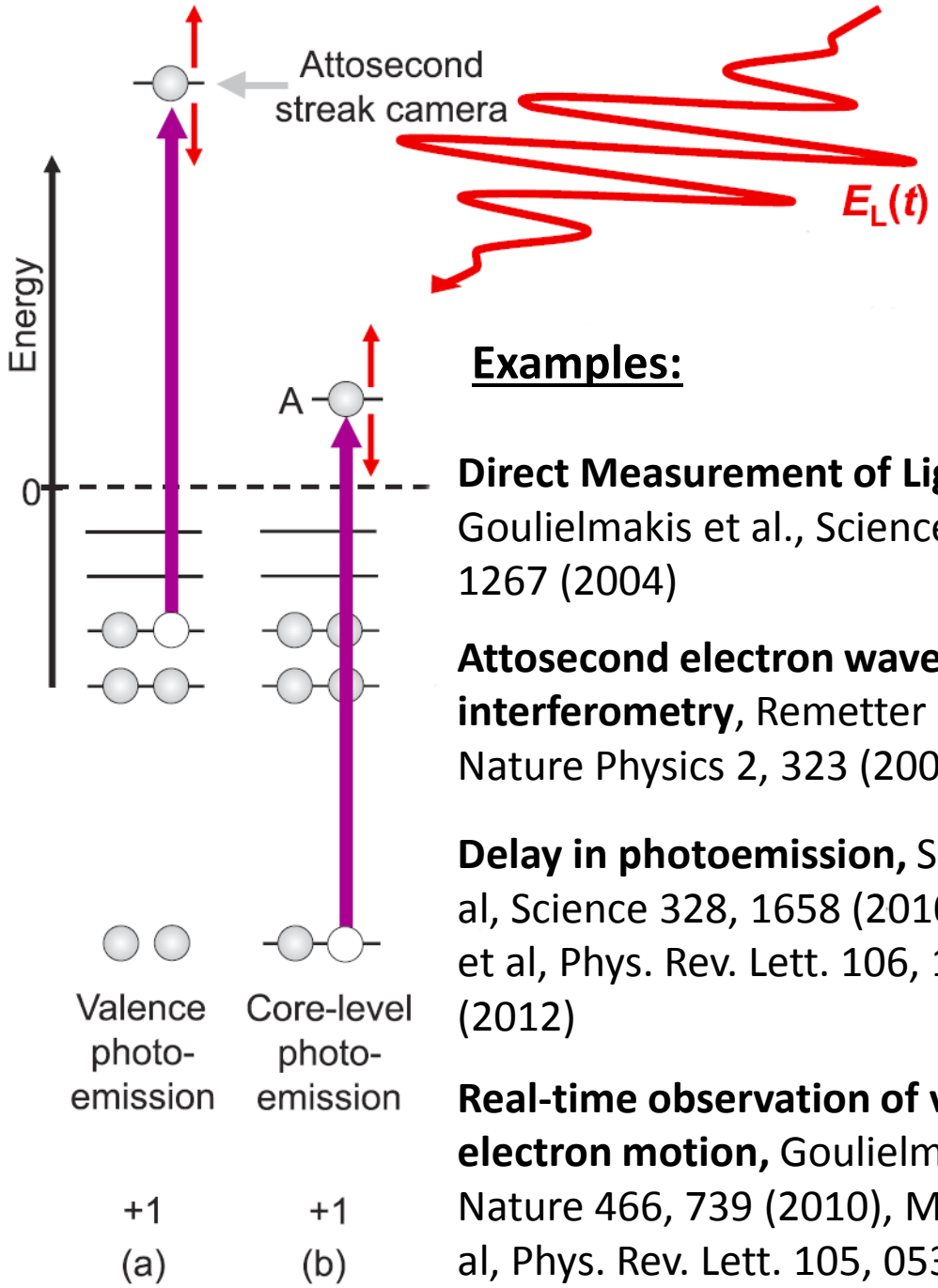
+ several chapters in „Chemistry and Molecular Physics on the Attosecond Timescale: Theoretical Approaches" (ed. By F. Lépine and M.J.J. Vrakking, RSC, summer 2018)

Attosecond pump-probe spectroscopy: atoms

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:

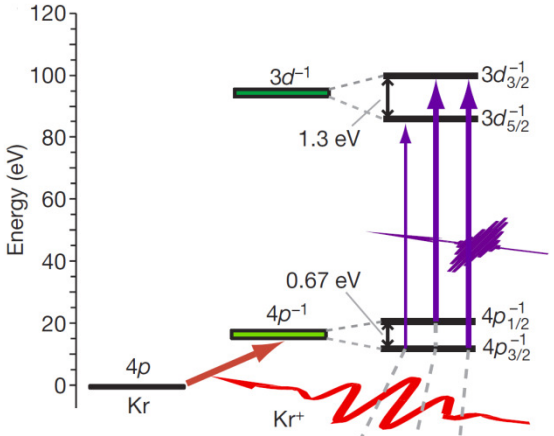
Direct Measurement of Light Waves, Goulielmakis et al., Science 305, 1267 (2004)

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)

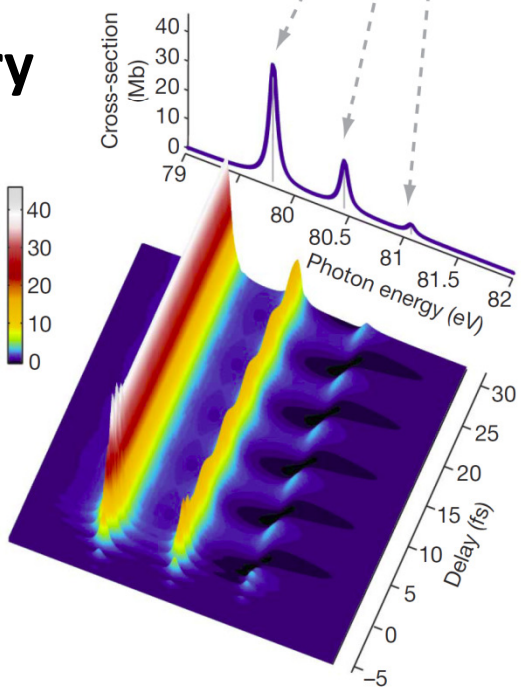
Observation of electronic coherence



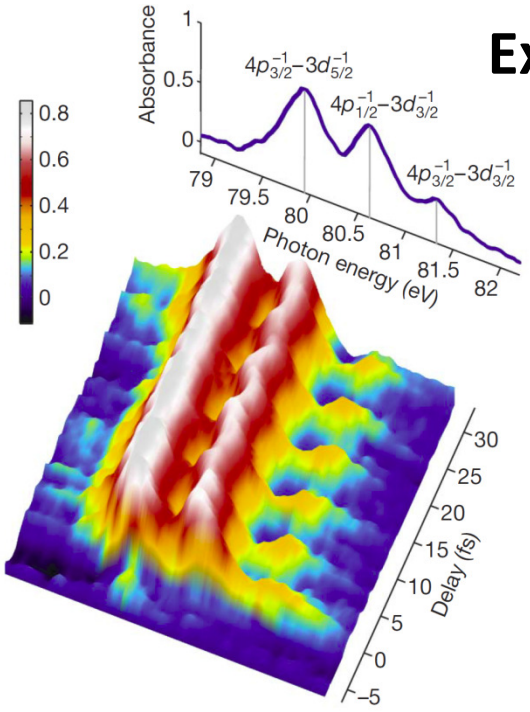
After ionization the Kr^+ ion is in a $4p_{1/2}$ or $4p_{3/2}$ state

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

Theory

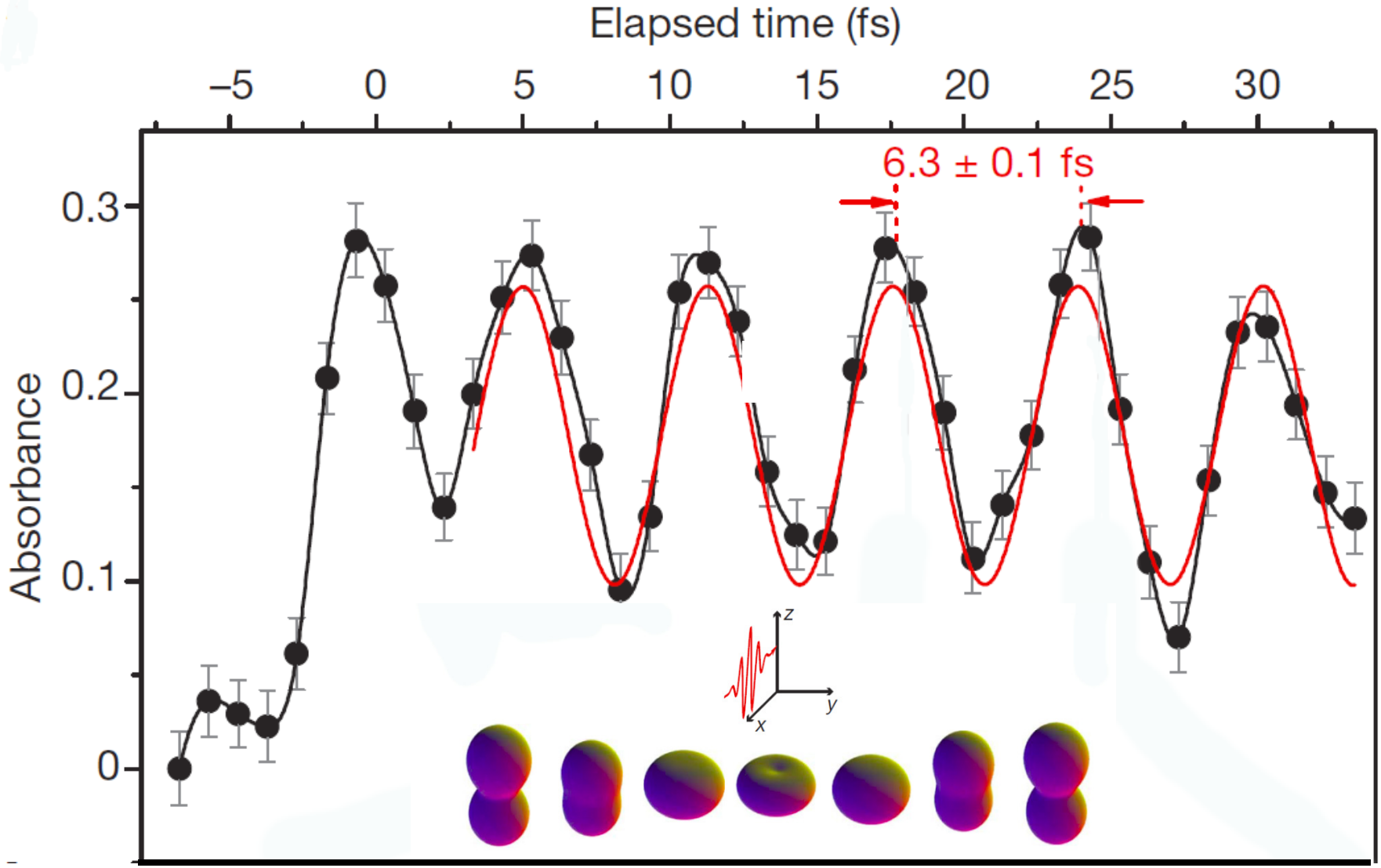


Experiment



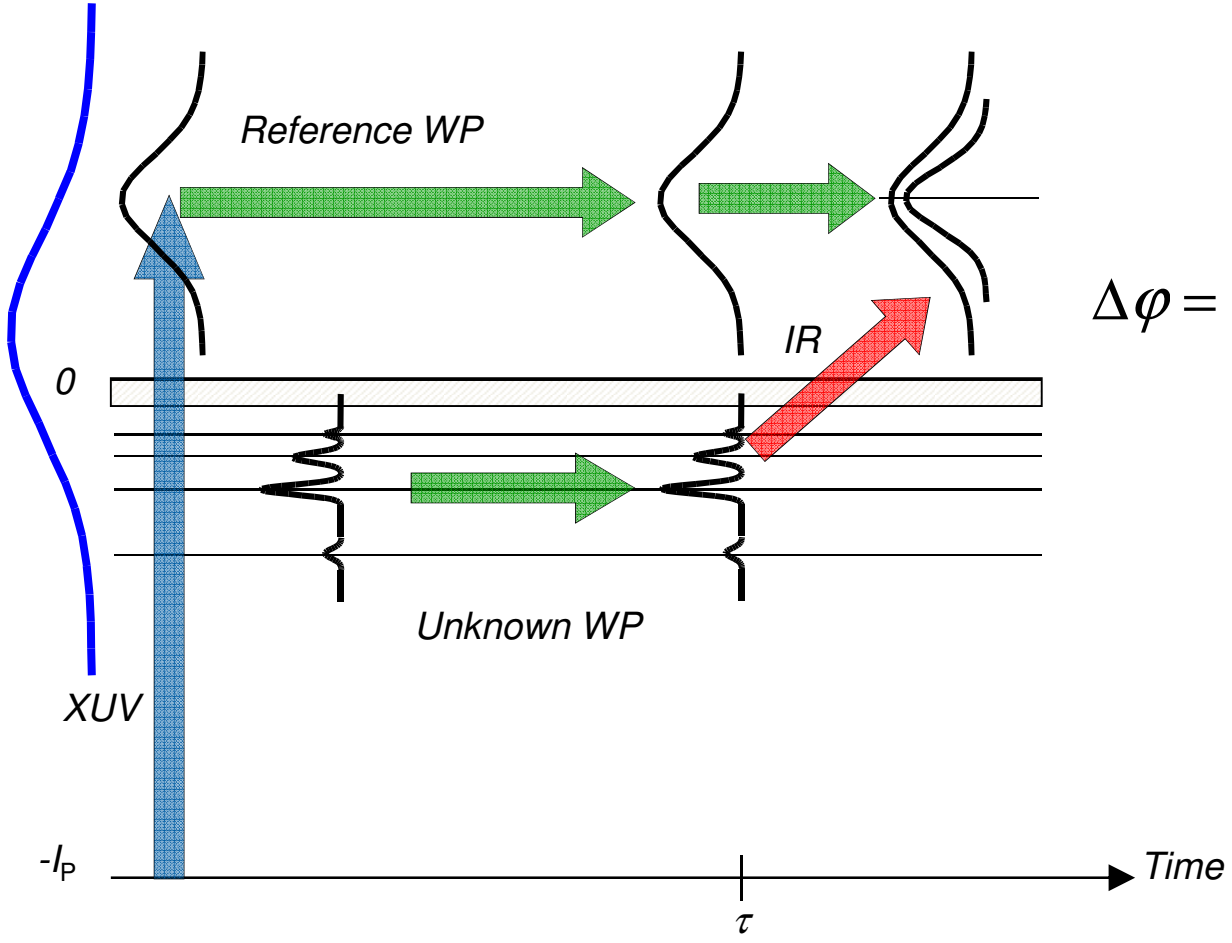
Goulielmakis et al., Nature 466, 739 (2010)

Observation of electronic coherence



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

Holographic observation of electronic coherence

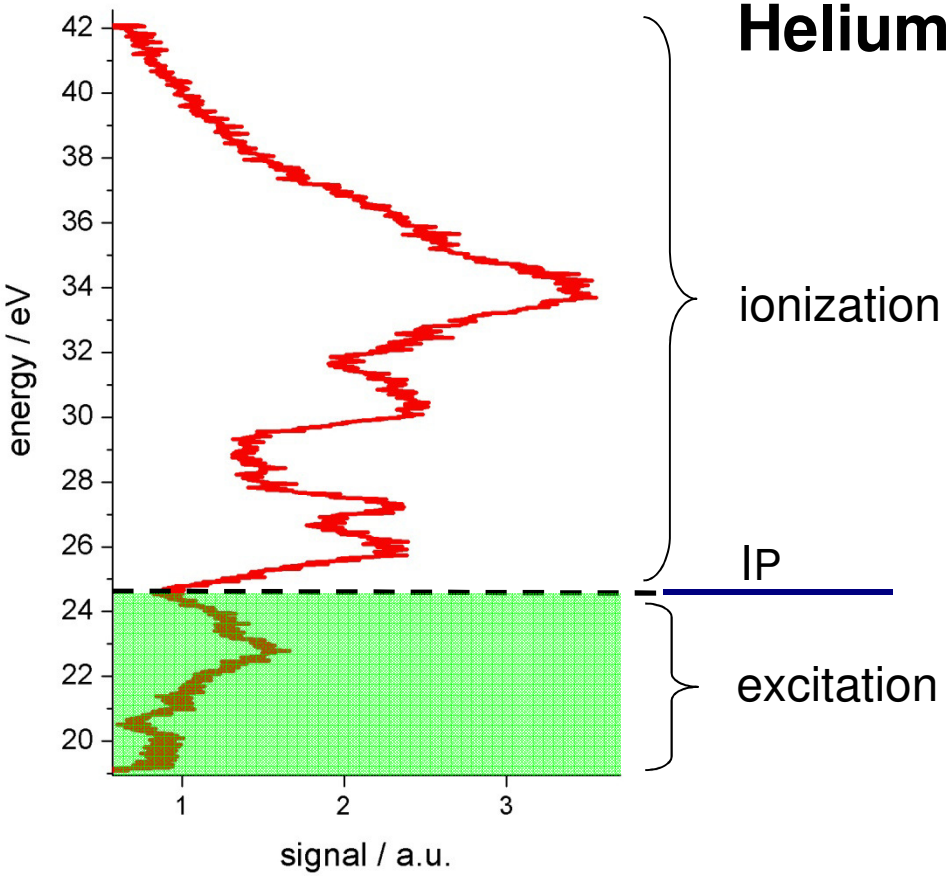


$$\Delta\varphi = (E_{ref} - E_A)\tau \ /\hbar$$

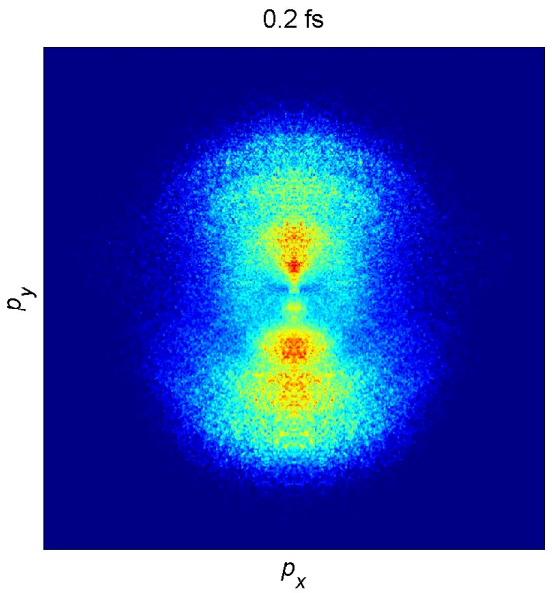
Mauritsson et al, Phys. Rev. Lett. 105, 053001 (2010)

Near-threshold Electron Wavepackets

HHG in Xenon, polarization gated
100 nm Al filter



VMIS image

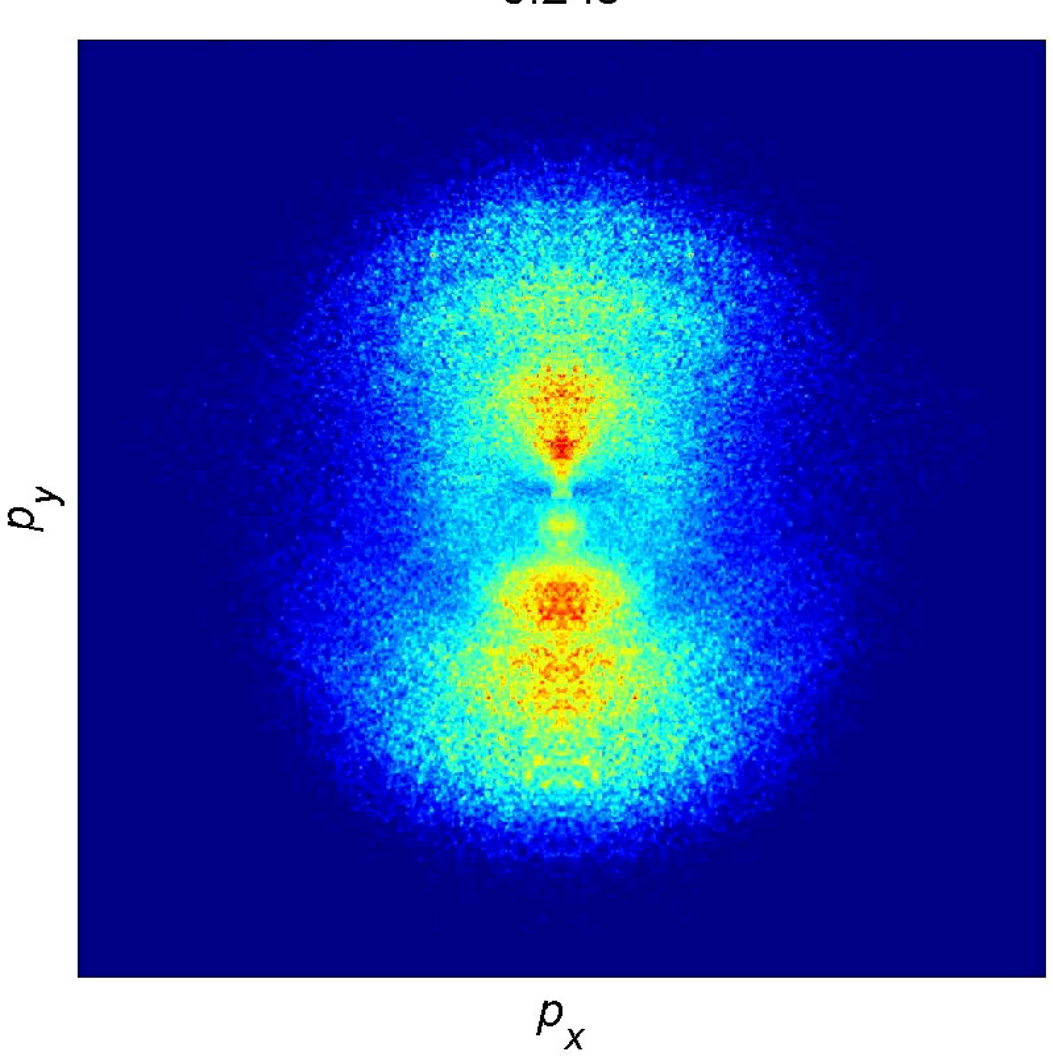


Helium ionization (raw image)

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

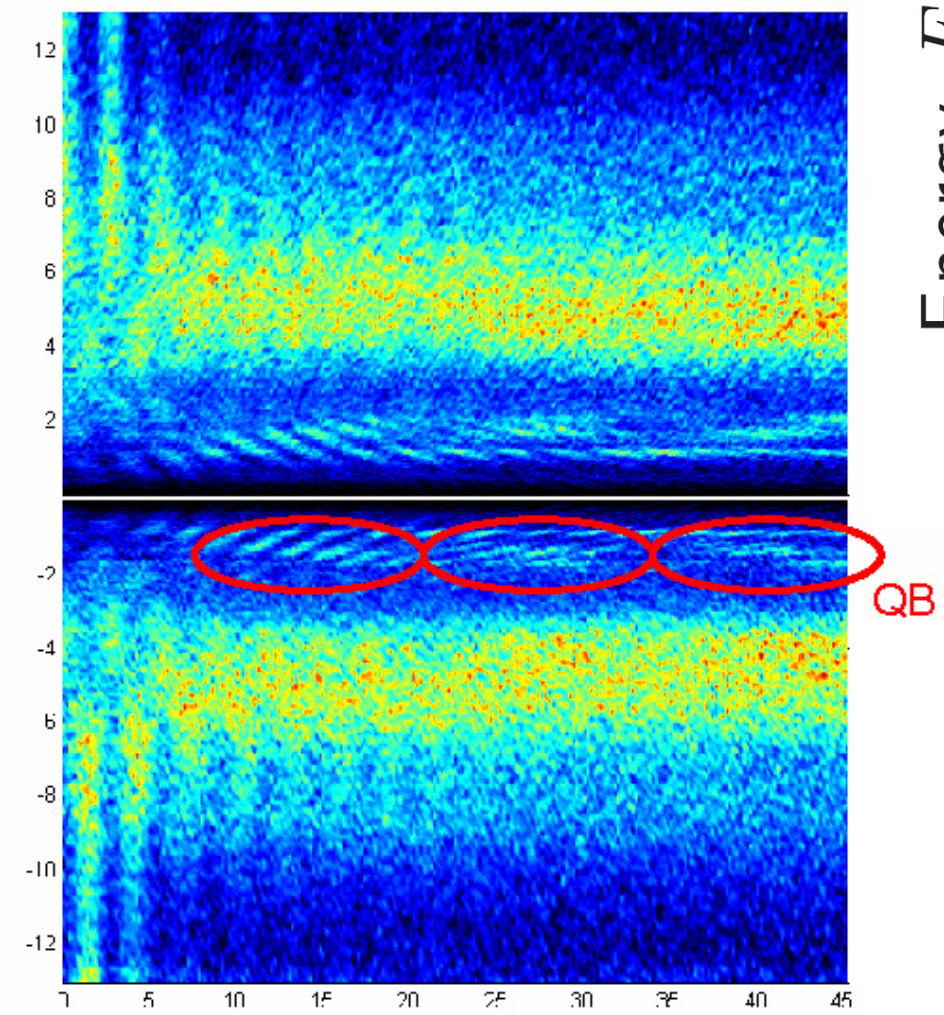
Near-threshold Electron Wavepackets

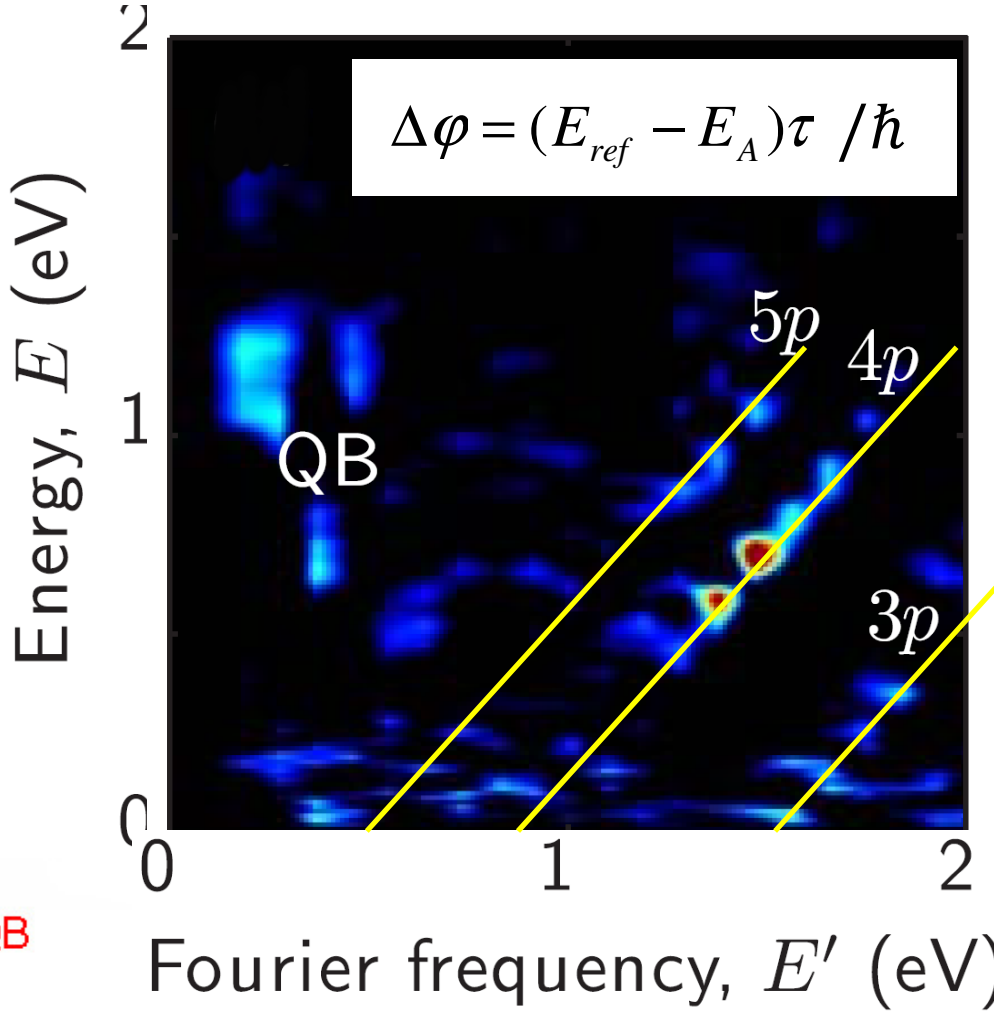
HHG in Xenon, Helium two-color ionization, I ~10^{13} W/cm^2



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

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





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

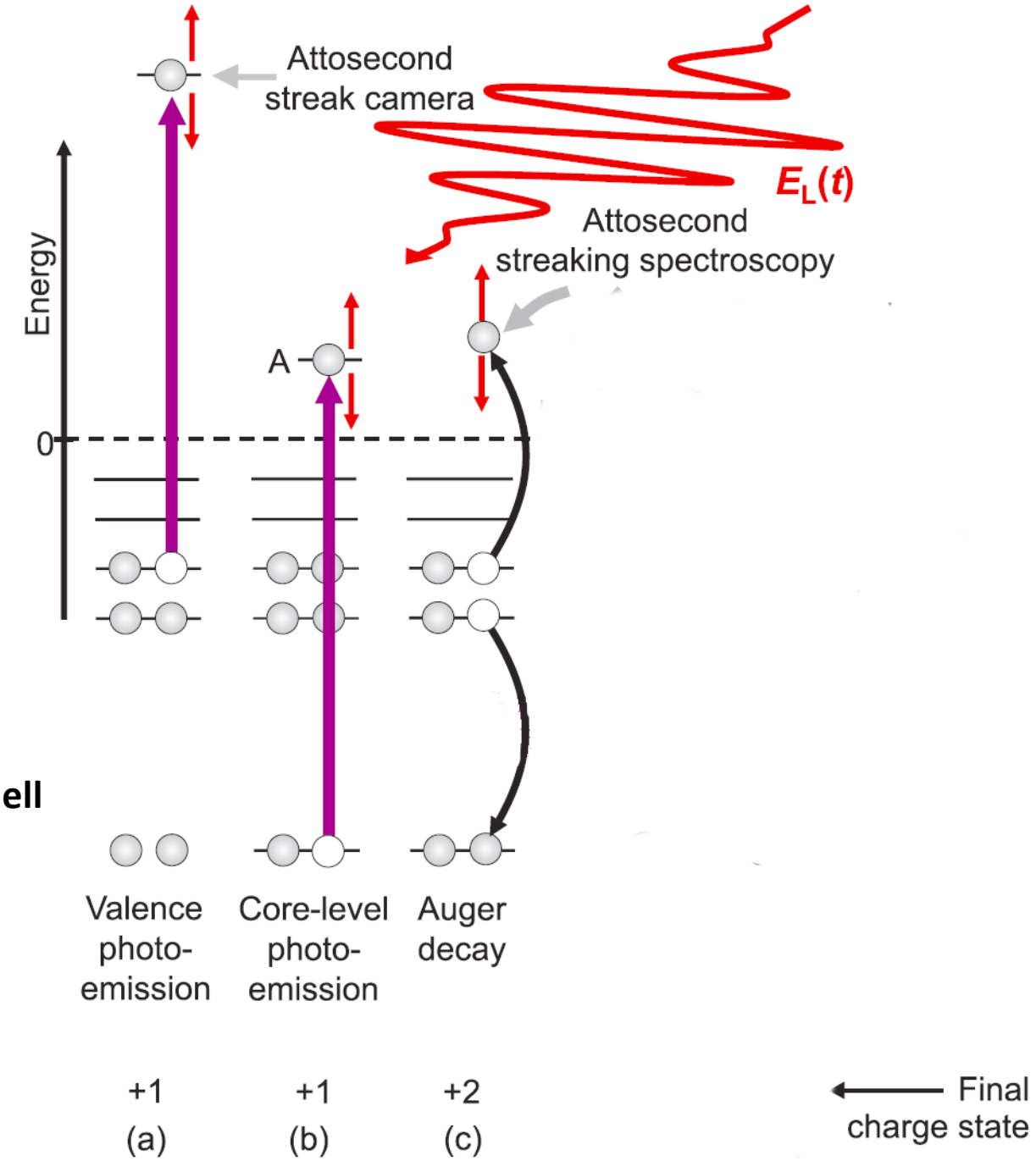
Attosecond atomic physics

Multi-electron dynamics

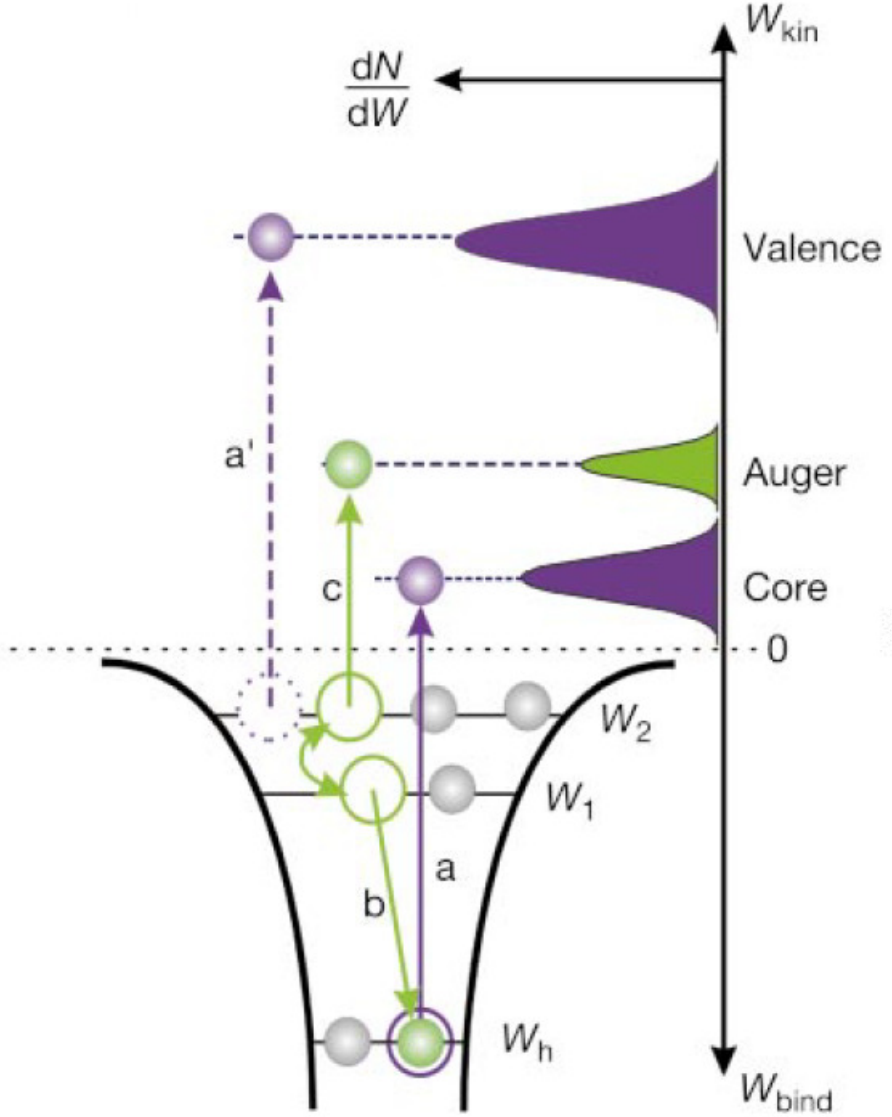
- Auger decay

Example:

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



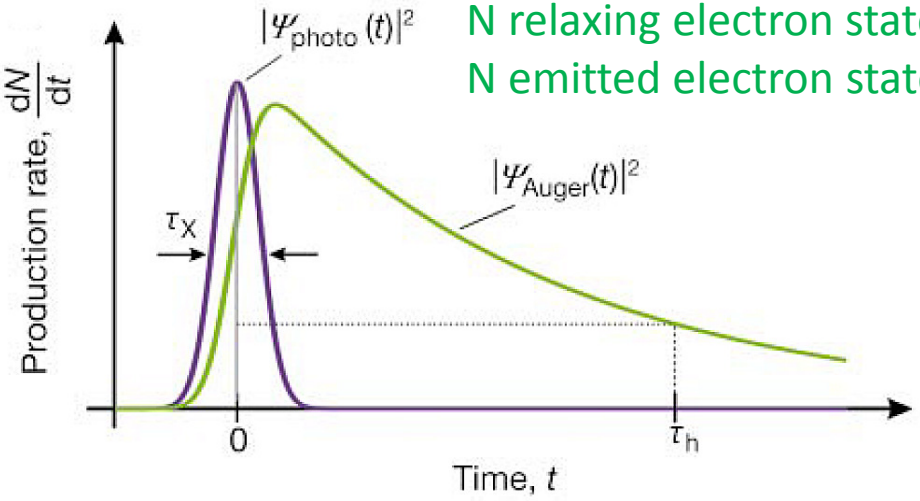
Attosecond measurement of Auger decay



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 liftetime of the core hole

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



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

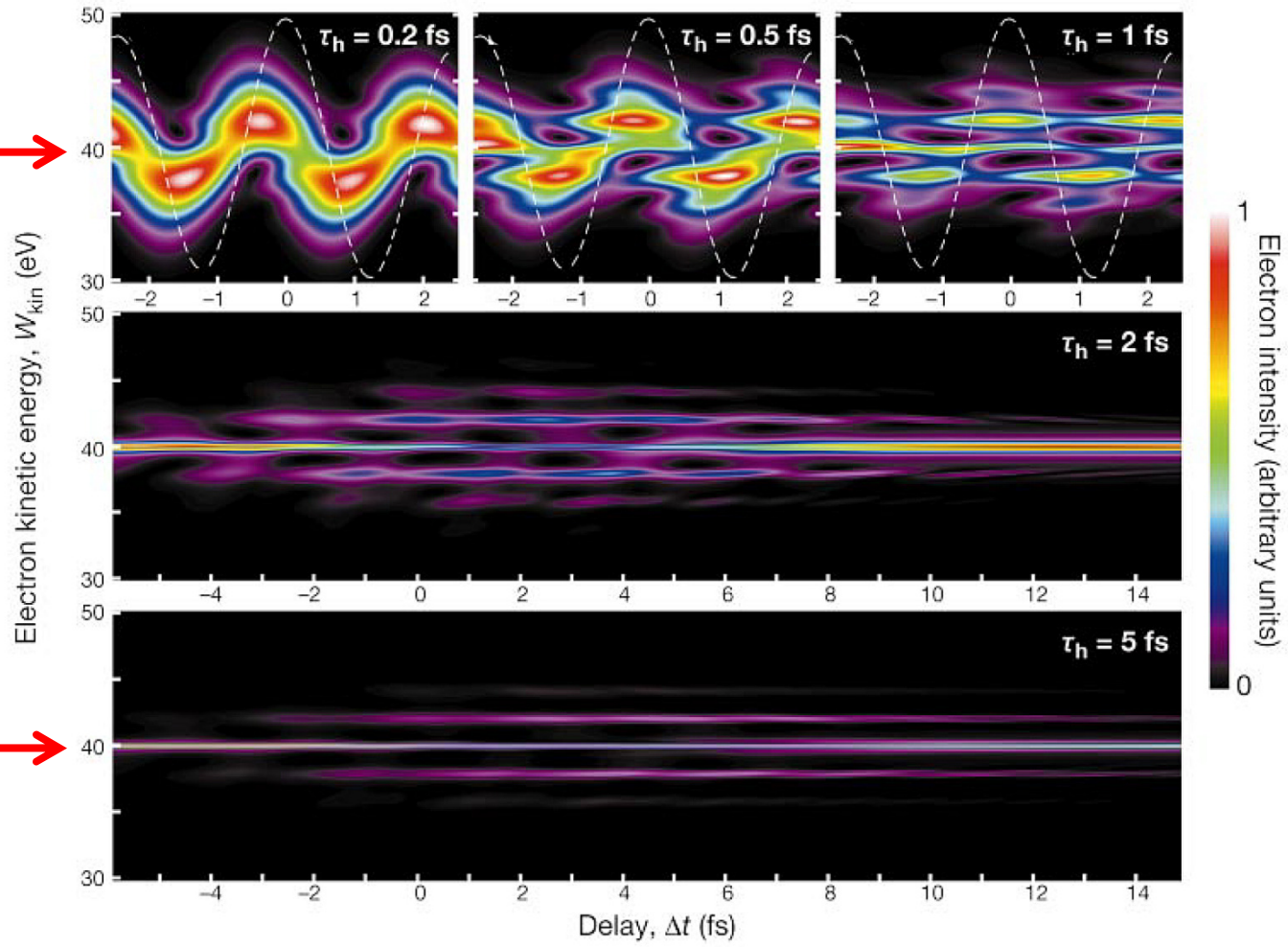
Attosecond measurement of Auger decay

streaking regime

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

sideband regime

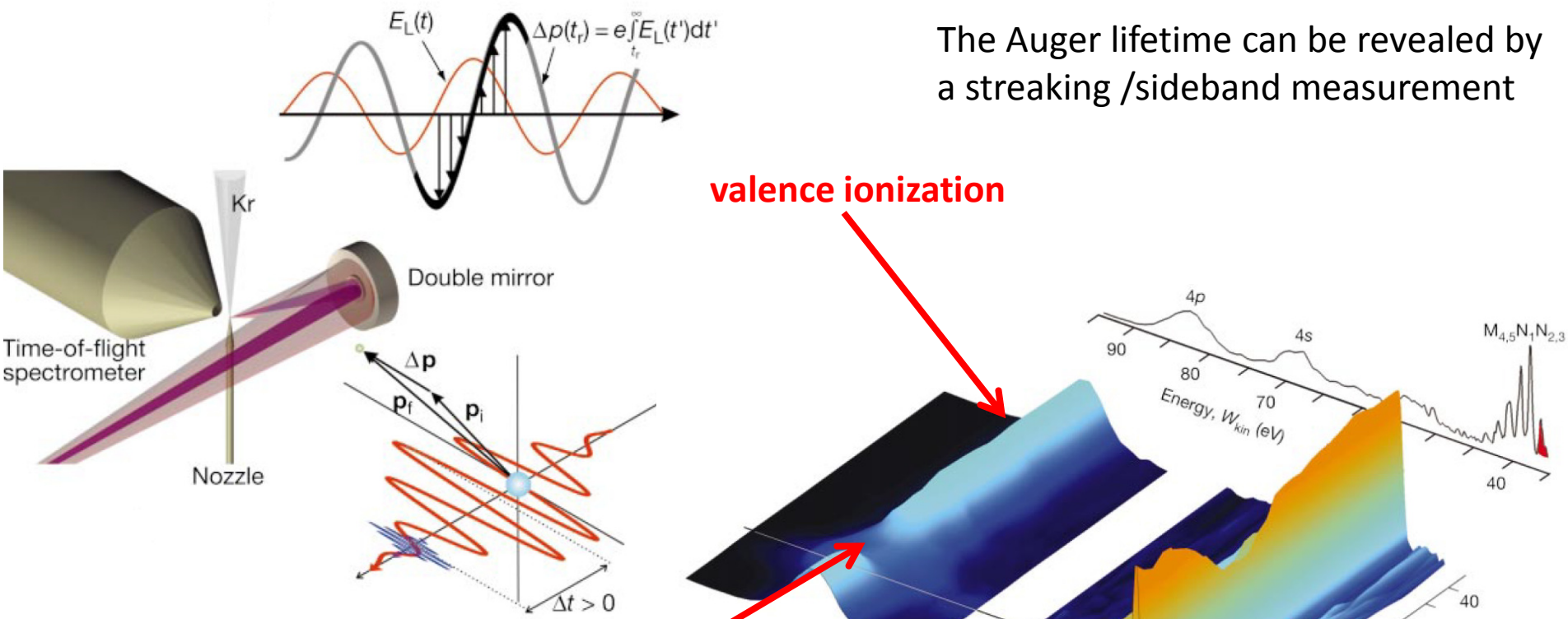
$\tau_{Auger} >> \tau_{laser}$



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

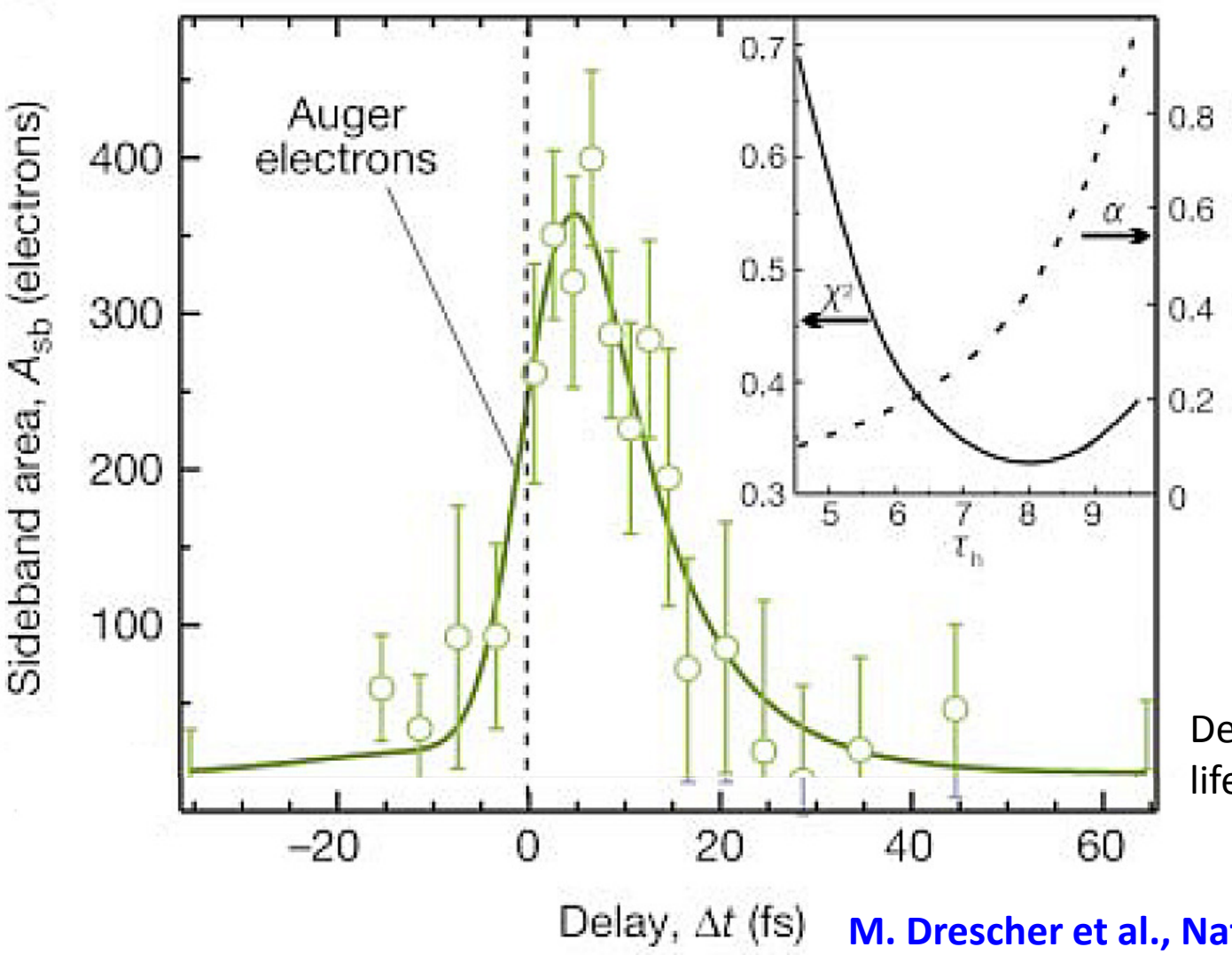
Attosecond measurement of Auger decay

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)

Attosecond atomic physics

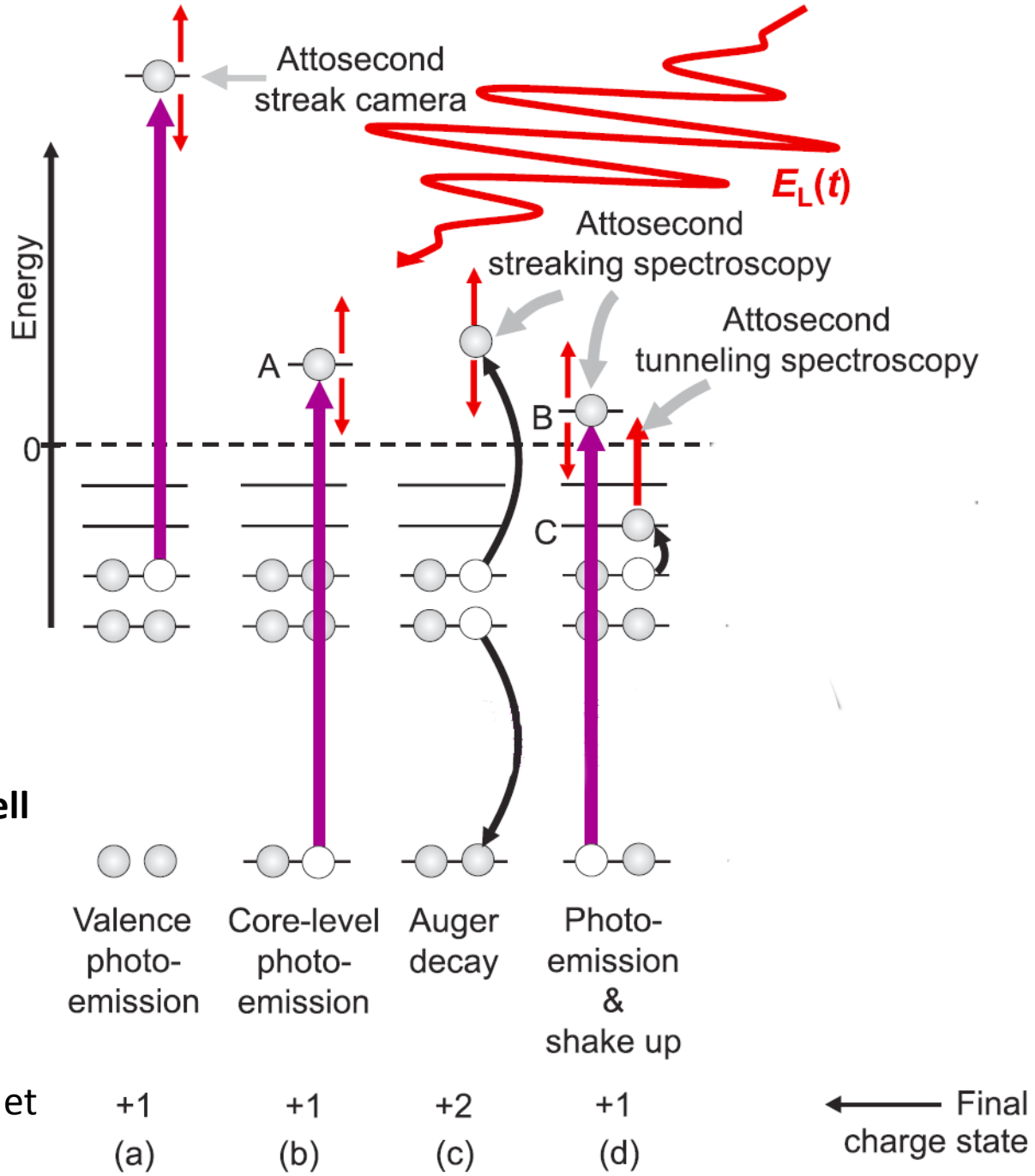
Multi-electron dynamics

- Auger decay
- Shake-up

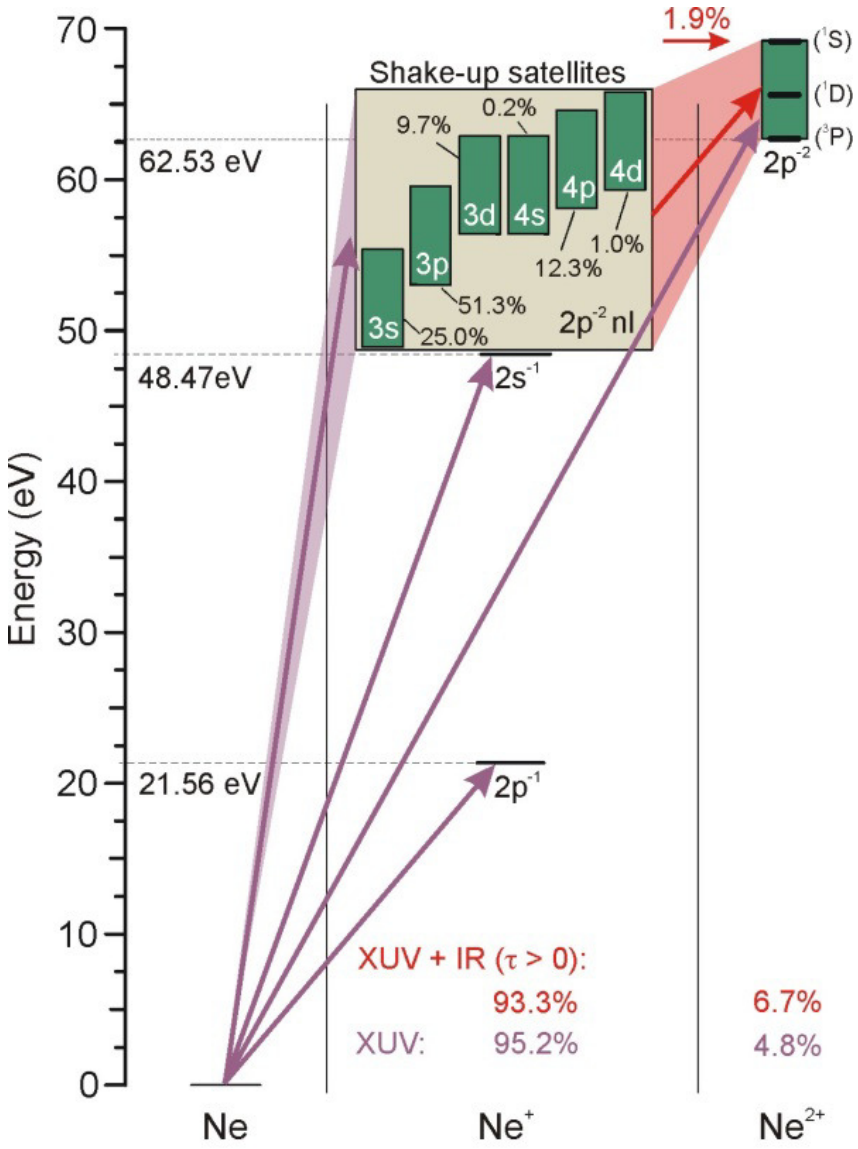
Example:

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)

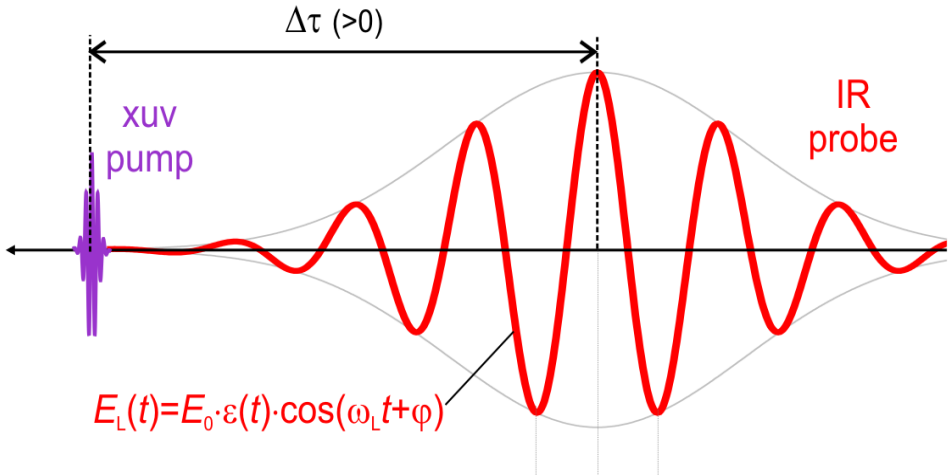


Time-resolving the tunneling process

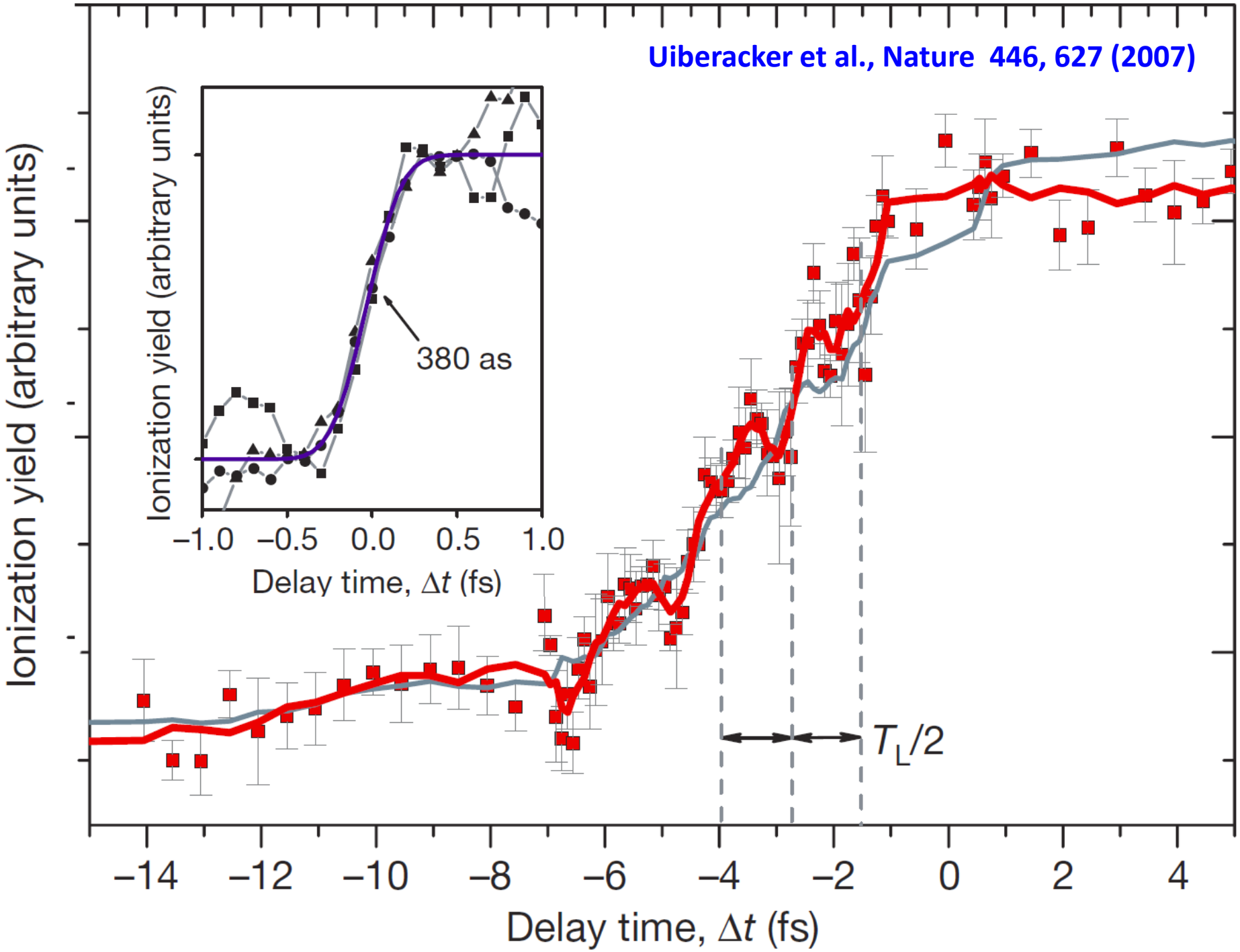


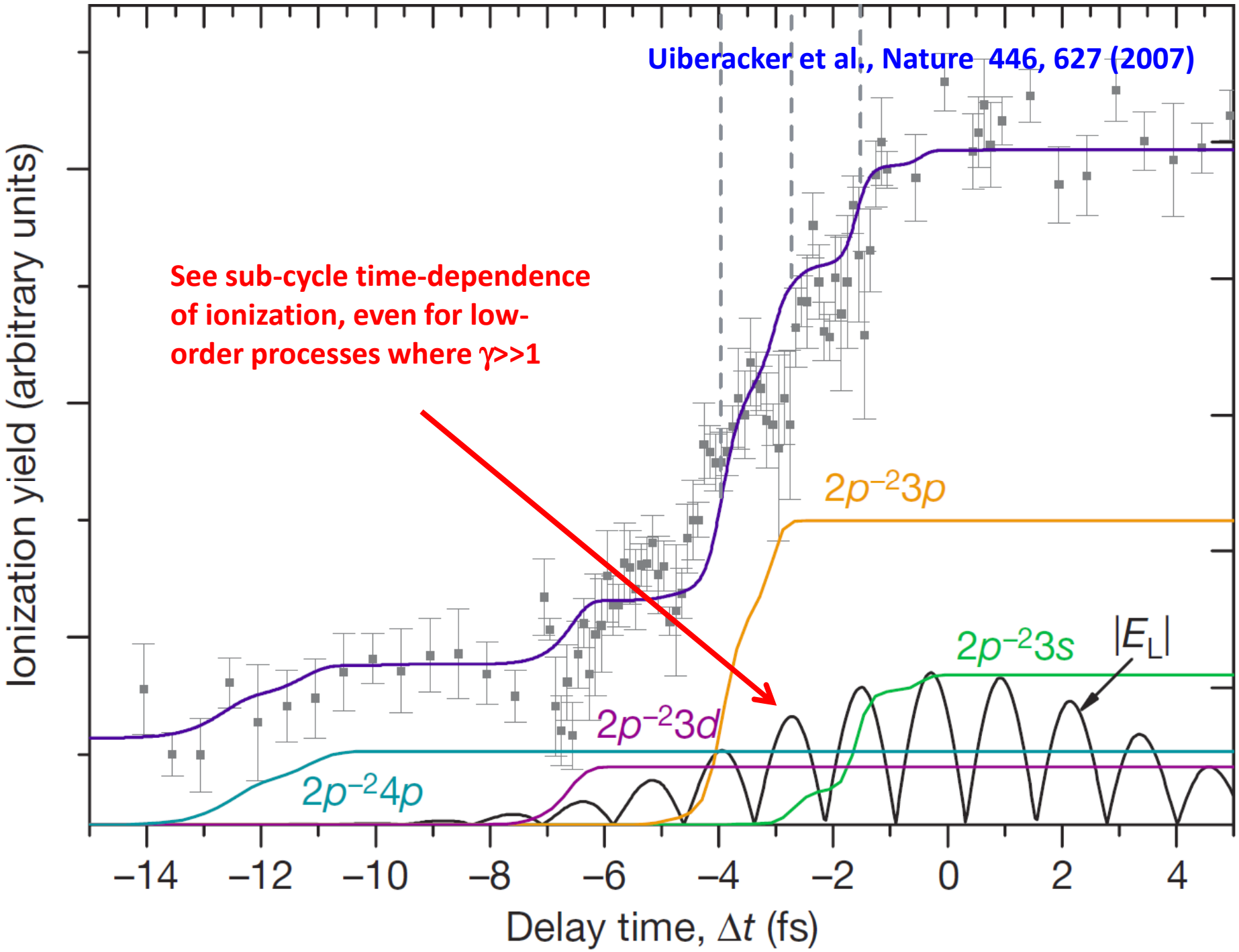
Photoionization of Ne at 90 eV leads to both the removal of a 2p valence electron and shake-up of a second 2p electron into a Rydberg state

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



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


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


Uiberacker et al., Nature 446, 627 (2007)
See sub-cycle time-dependence of ionization, even for low-order processes where γ>>1
Ionization yield (arbitrary units)
Delay time, Δt (fs)
2p⁻²3p
2p⁻²3s
|E_L|
2p⁻²3d
2p⁻²4p
−14
−12
−10
−8
−6
−4
−2
0
2
4

Attosecond pump-probe spectroscopy: molecules

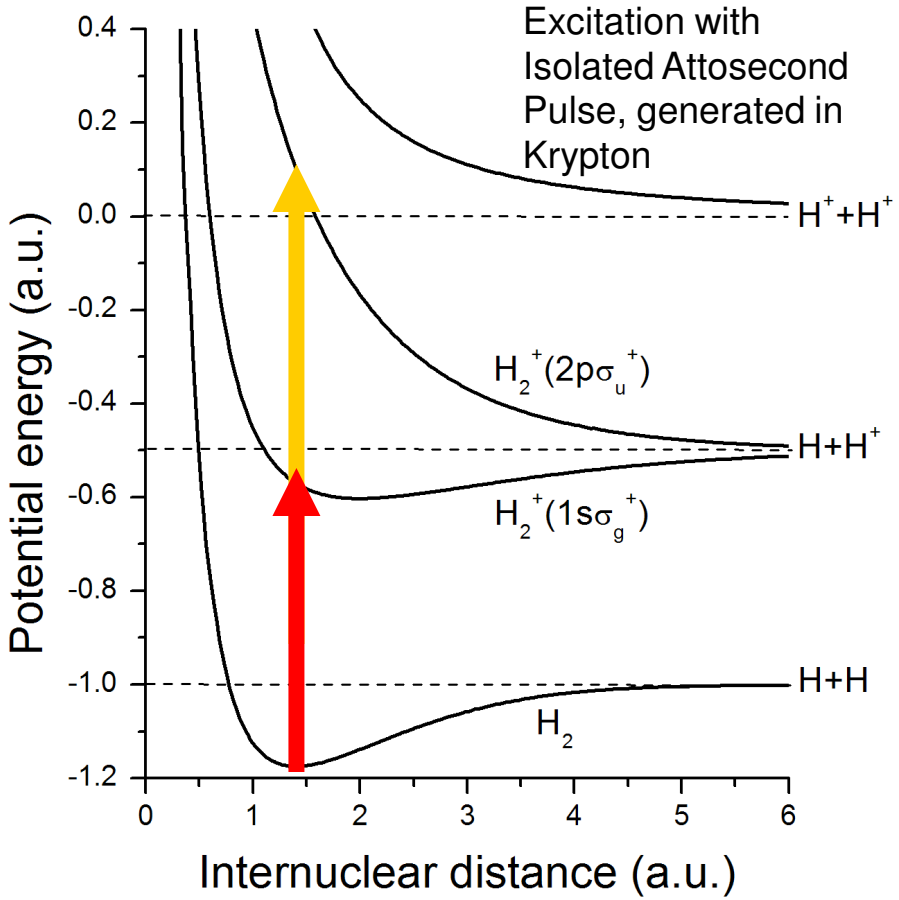
Application of Attosecond Pulses to Molecular Science

Question: why should we care?

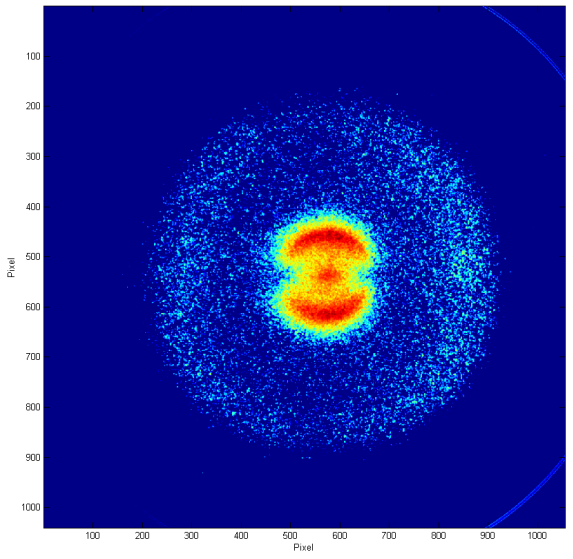
Answer:

- Most light-induced photo-chemical processes start with an electronic interaction; atomic motion sets because of coupling of electronic and nuclear degrees of freedom
- Many light-induced interactions involve multiple electrons – when is coupling of electronic degrees of freedom important?
- By studying molecular dynamics on attosecond to few-femtosecond timescales we may uncover new pathways towards control at the molecular level

Coupled electronic+nuclear dynamics: Pump-probe experiments on H_2 and D_2



Use isolated attosecond pulse generated in Krypton to launch a wavepacket on the $2p\sigma_u^+$ state or the $1s\sigma_g^+$ state and investigate the subsequent IR interaction



Electron localization in H_2^+

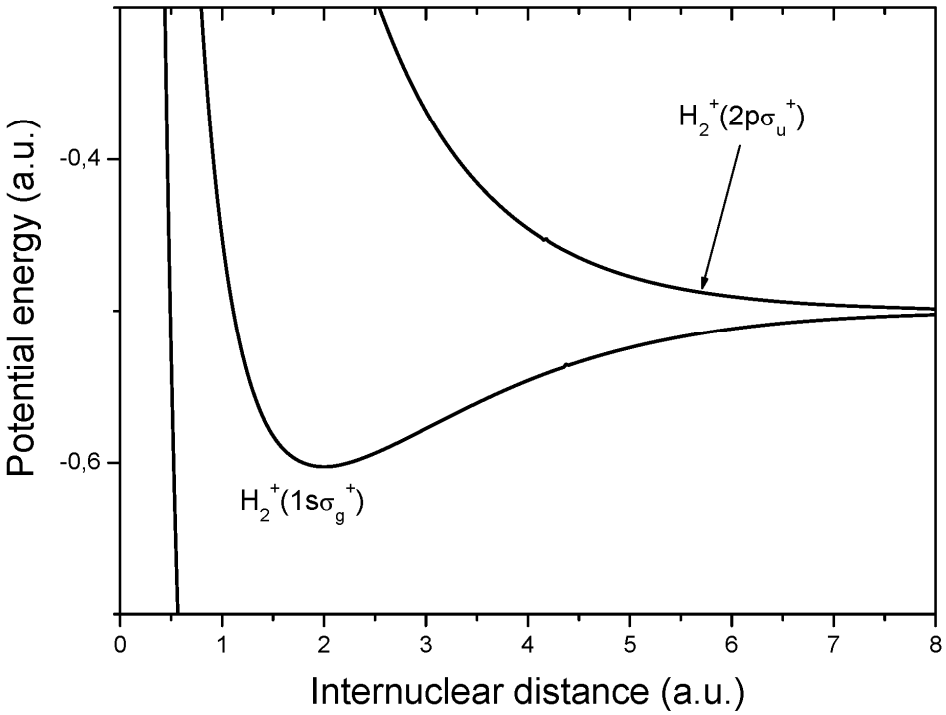
$$\psi_g = (1/\sqrt{2})\{\psi_{left} + \psi_{right}\}$$

$$\psi_u = (1/\sqrt{2})\{\psi_{left} - \psi_{right}\}$$

Rewrite

$$\psi_{left} = (1/\sqrt{2})\{\psi_g + \psi_u\}$$

$$\psi_{right} = (1/\sqrt{2})\{\psi_g - \psi_u\}$$



The observation of electron localization requires that the parity of the ionic wavefunction is broken

Consider the wavefunction of the ion+electron

$$\Psi = c_1[\psi_g \mathcal{E}l_g]_g + c_2[\psi_g \mathcal{E}l_u]_u + c_3[\psi_u \mathcal{E}l_u]_g + c_4[\psi_u \mathcal{E}l_g]_u$$

Rewrite

$$\Psi = \frac{1}{2}\sqrt{2}(c_1 + c_4)\psi_{left}\mathcal{E}l_g + \frac{1}{2}\sqrt{2}(c_2 + c_3)\psi_{left}\mathcal{E}l_u$$
$$+ \frac{1}{2}\sqrt{2}(c_1 - c_4)\psi_{right}\mathcal{E}l_g + \frac{1}{2}\sqrt{2}(c_2 - c_3)\psi_{right}\mathcal{E}l_u$$

Determine the probability the bound electron / fragment ion is found on the right or on the left.

Consider the wavefunction of the ion+electron

$$\Psi = \frac{1}{2}\sqrt{2}(c_1 + c_4)\psi_{left}\mathcal{El}_g + \frac{1}{2}\sqrt{2}(c_2 + c_3)\psi_{left}\mathcal{El}_u$$

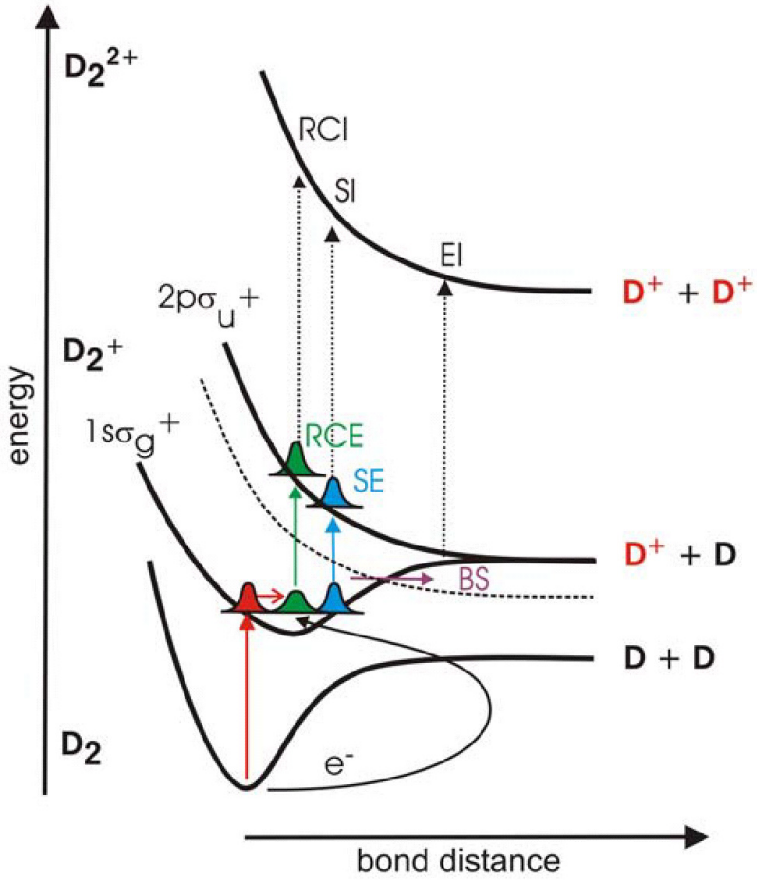
$$+ \frac{1}{2}\sqrt{2}(c_1 - c_4)\psi_{right}\mathcal{El}_g + \frac{1}{2}\sqrt{2}(c_2 - c_3)\psi_{right}\mathcal{El}_u$$

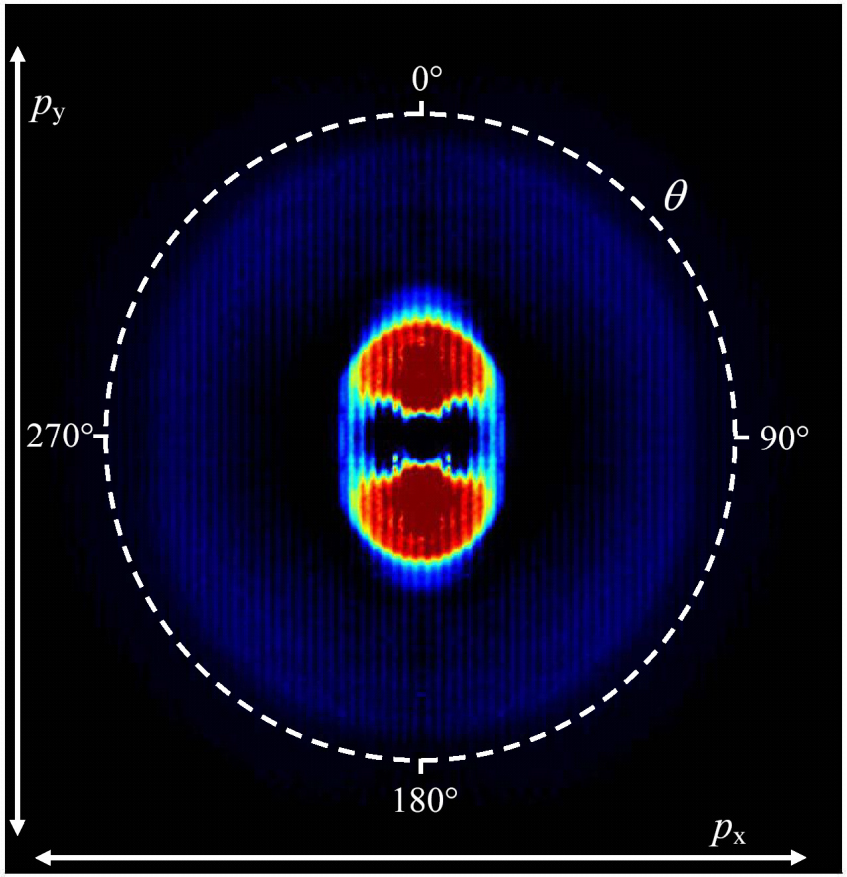
$$I_{left} = \frac{1}{2}|c_1 + c_4|^2 + \frac{1}{2}|c_2 + c_3|^2 \qquad I_{right} = \frac{1}{2}|c_1 - c_4|^2 + \frac{1}{2}|c_2 - c_3|^2$$

Dipole selection rules **forbid** that a difference occurs between I_{left} and I_{right} in single-photon ionization, since c_1 and c_3 are zero

But we can see asymmetries when multi-photon processes involving **both odd and even numbers** of photons are simultaneously active

First: IR-only electron localization control

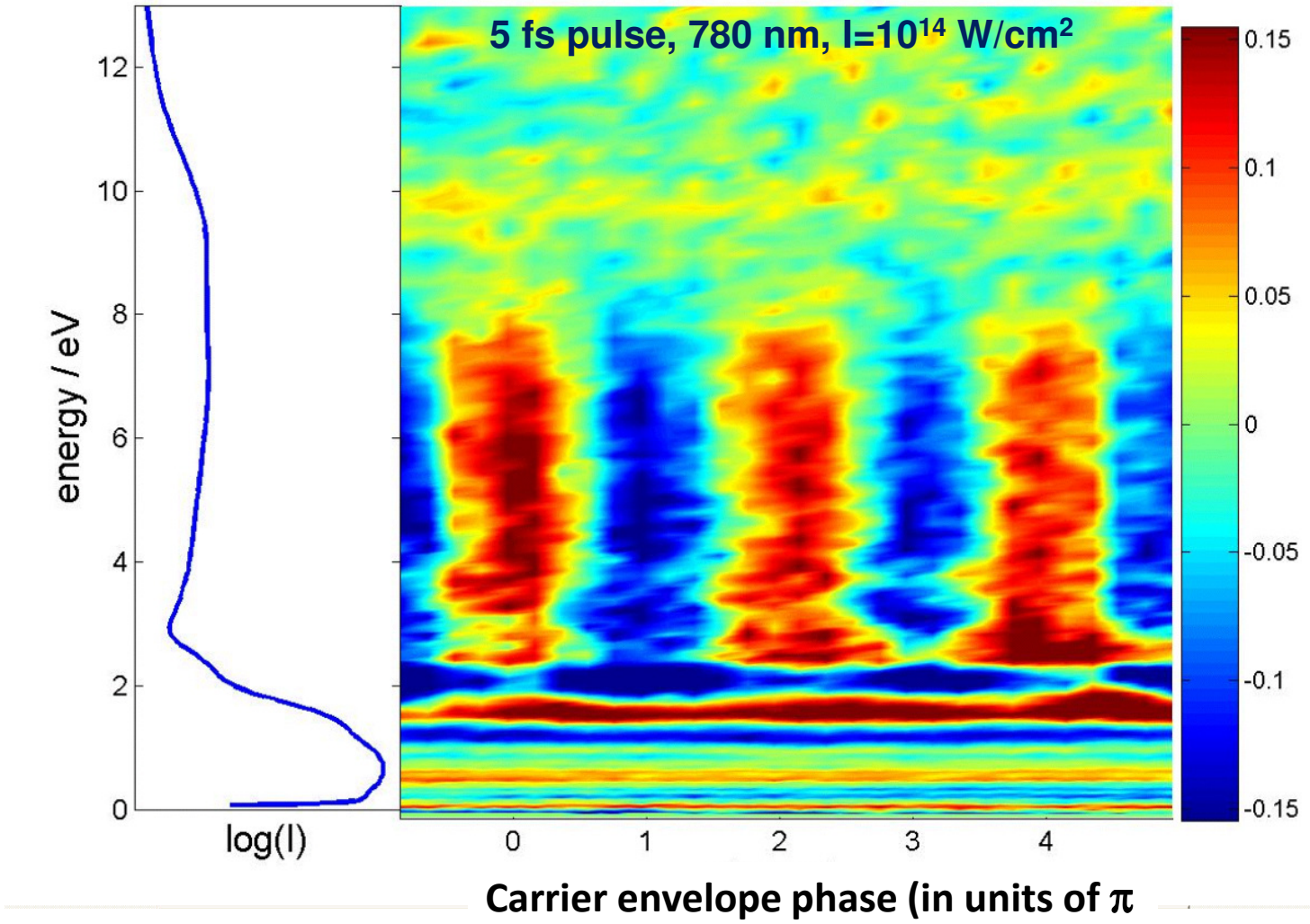




Angle-resolved D^+ ion imaging using CEP-locked few-cycle laser pulses

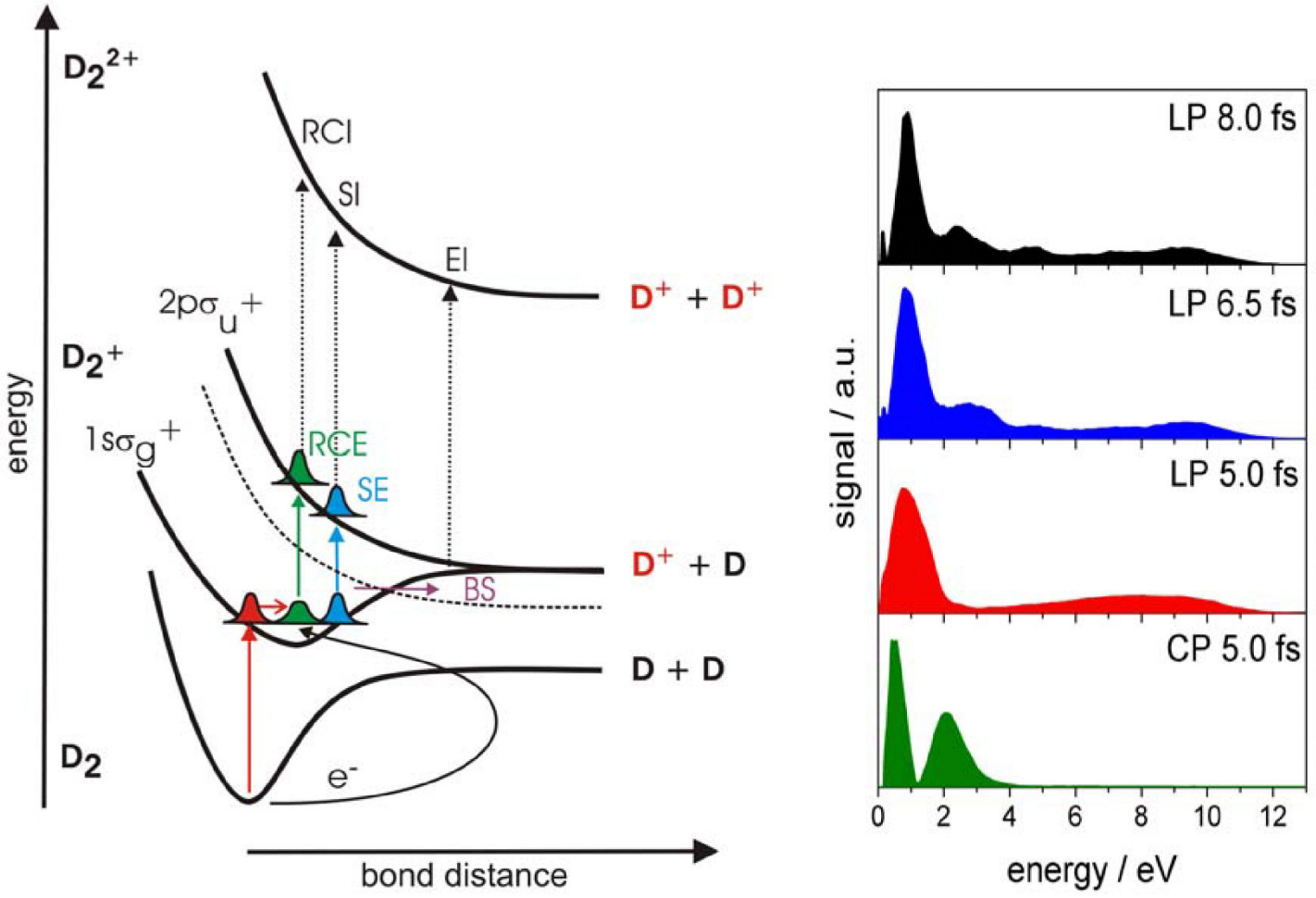
M. Kling et al., Science 312, 246 (2006)

Asymmetry $(D^+_{up}-D^+_{down})/(D^+_{up}+D^+_{down})$

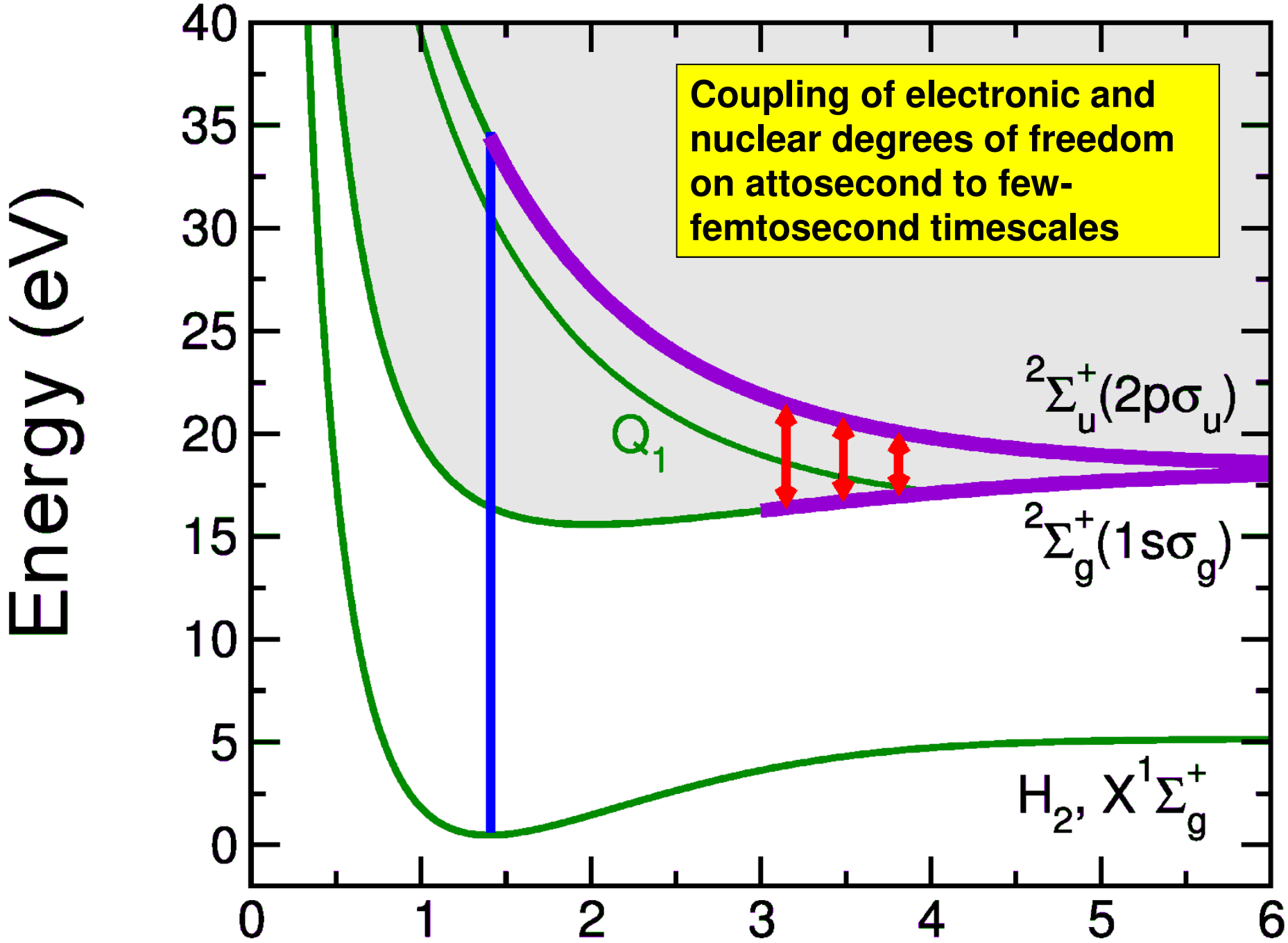


M. Kling et al., Science 312, 246 (2006)

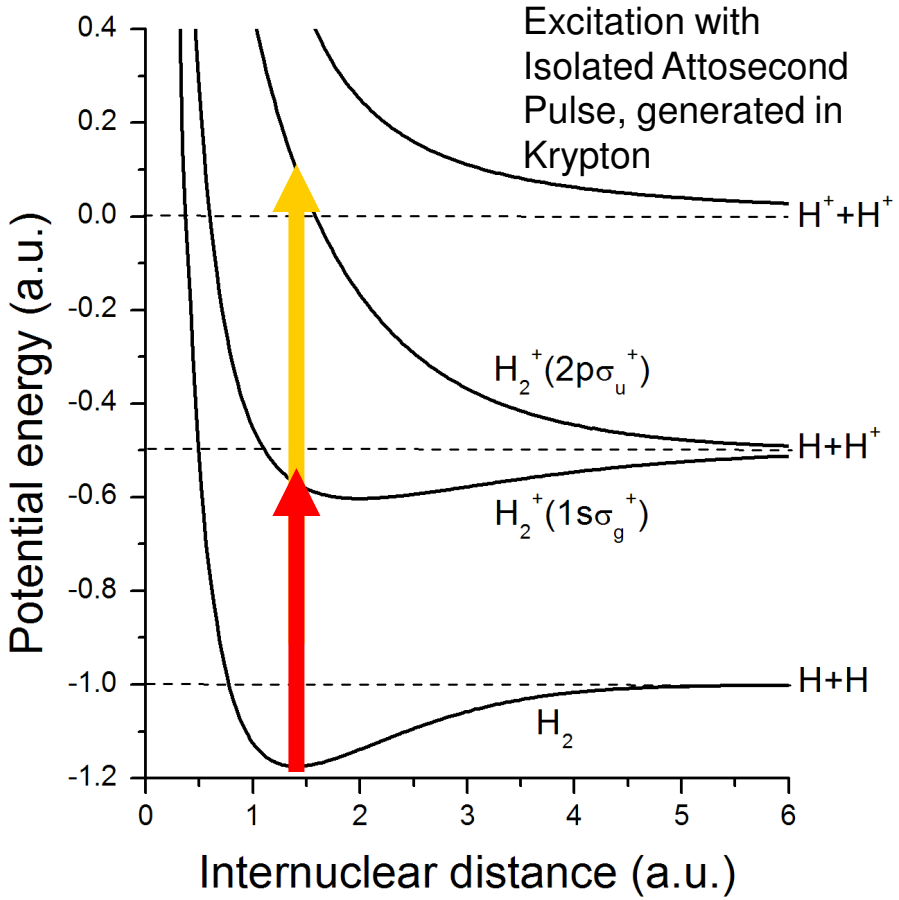
Phase Control Mechanism -1



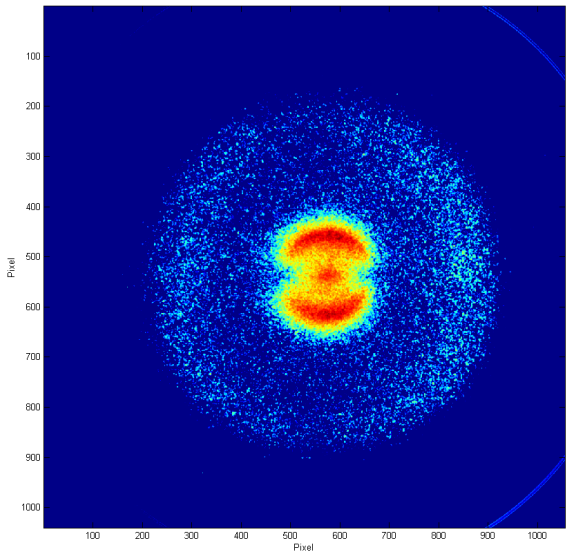
Recollision-induced population of the $2p\sigma_u^+$ state


Energy (eV)
40
35
30
25
20
15
10
5
0
0
1
2
3
4
5
6
Coupling of electronic and nuclear degrees of freedom on attosecond to few-femtosecond timescales
$^2\Sigma_u^+(2p\sigma_u)$
Q_1
$^2\Sigma_g^+(1s\sigma_g)$
H_2, $X\,^1\Sigma_g^+$

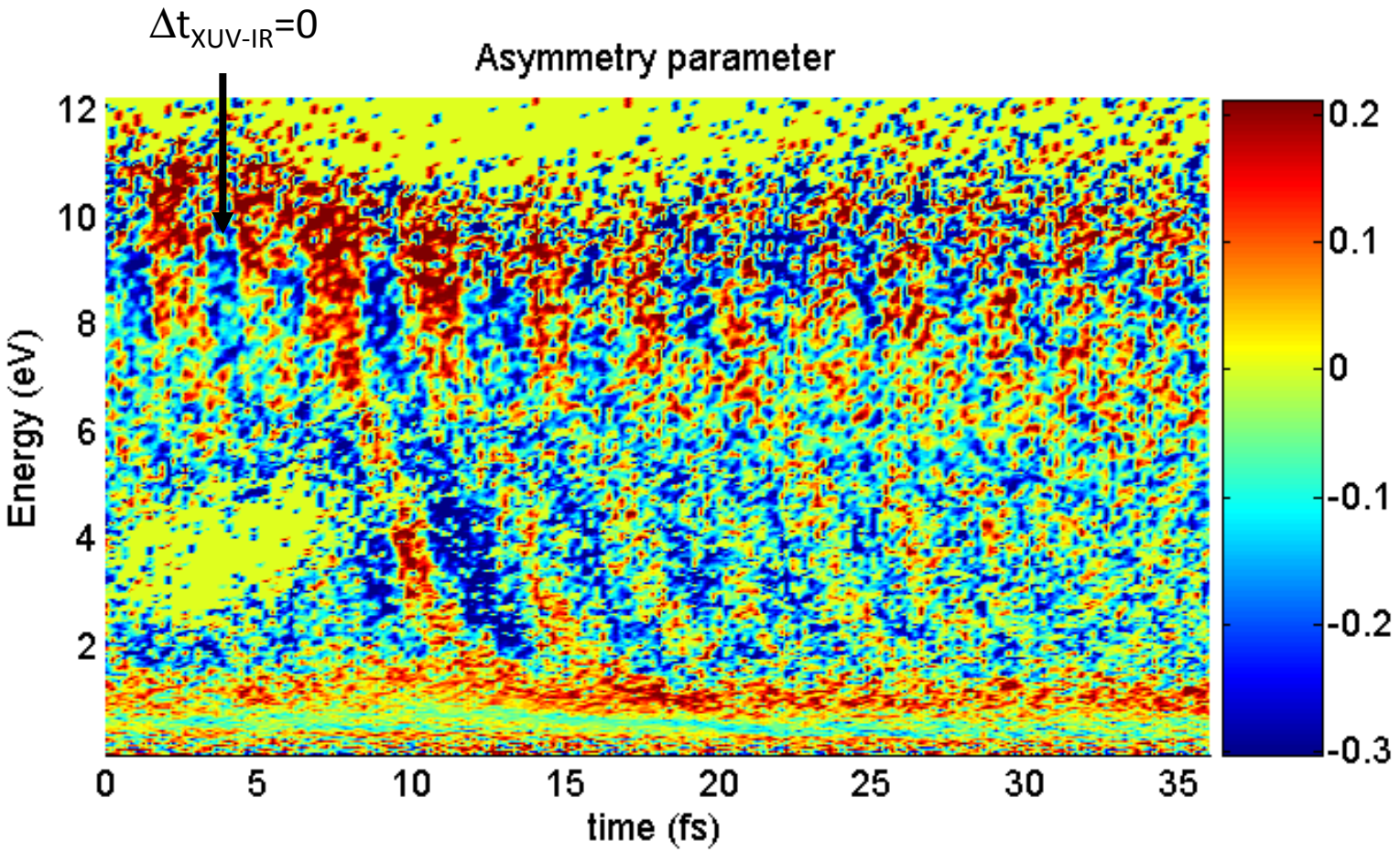
Coupled electronic+nuclear dynamics: Pump-probe experiments on H_2 and D_2



Use isolated attosecond pulse generated in Krypton to launch a wavepacket on the $2p\sigma_u^+$ state or the $1s\sigma_g^+$ state and investigate the subsequent IR interaction



Electron localization in XUV-IR dissociative ionization of H_2 and D_2



Sansone et al., Nature 465, 763 (2010)

Consider the wavefunction of the ion+electron

$$\Psi = \frac{1}{2}\sqrt{2}(c_1 + c_4)\psi_{left}\mathcal{El}_g + \frac{1}{2}\sqrt{2}(c_2 + c_3)\psi_{left}\mathcal{El}_u$$

$$+ \frac{1}{2}\sqrt{2}(c_1 - c_4)\psi_{right}\mathcal{El}_g + \frac{1}{2}\sqrt{2}(c_2 - c_3)\psi_{right}\mathcal{El}_u$$

$$I_{left} = \frac{1}{2}|c_1 + c_4|^2 + \frac{1}{2}|c_2 + c_3|^2 \qquad I_{right} = \frac{1}{2}|c_1 - c_4|^2 + \frac{1}{2}|c_2 - c_3|^2$$

Dipole selection rules **forbid** that a difference occurs between I_{left} and I_{right} in single-photon ionization, since c_1 and c_3 are zero

Consider the wavefunction of the ion+electron

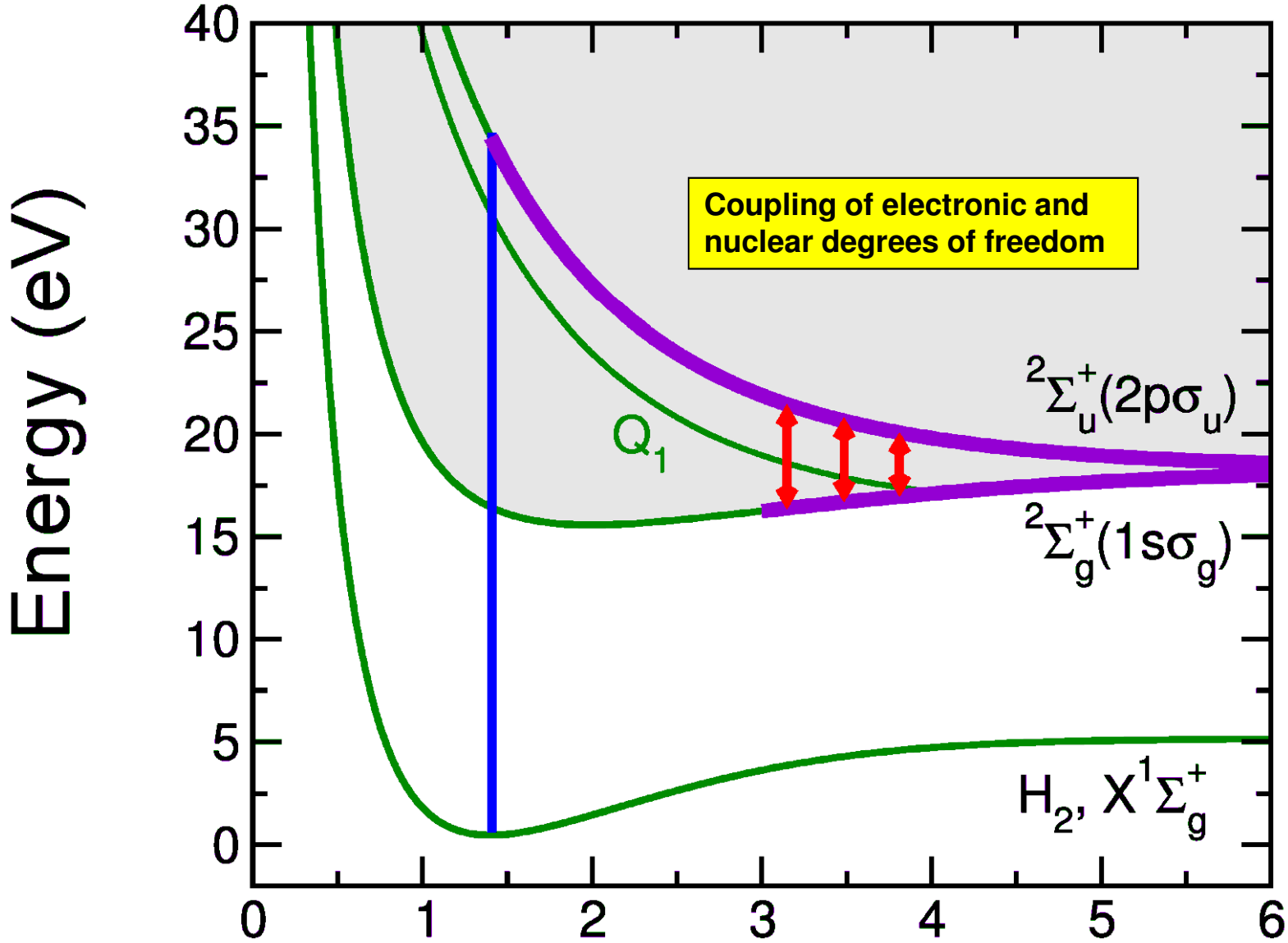
$$\Psi = c_1[\psi_g \mathcal{E}l_g]_g + c_2[\psi_g \mathcal{E}l_u]_u + c_3[\psi_u \mathcal{E}l_u]_g + c_4[\psi_u \mathcal{E}l_g]_u$$

We can imagine two ways to construct a coherent superpositon of the „g“ and „u“ state starting from this wavefunction

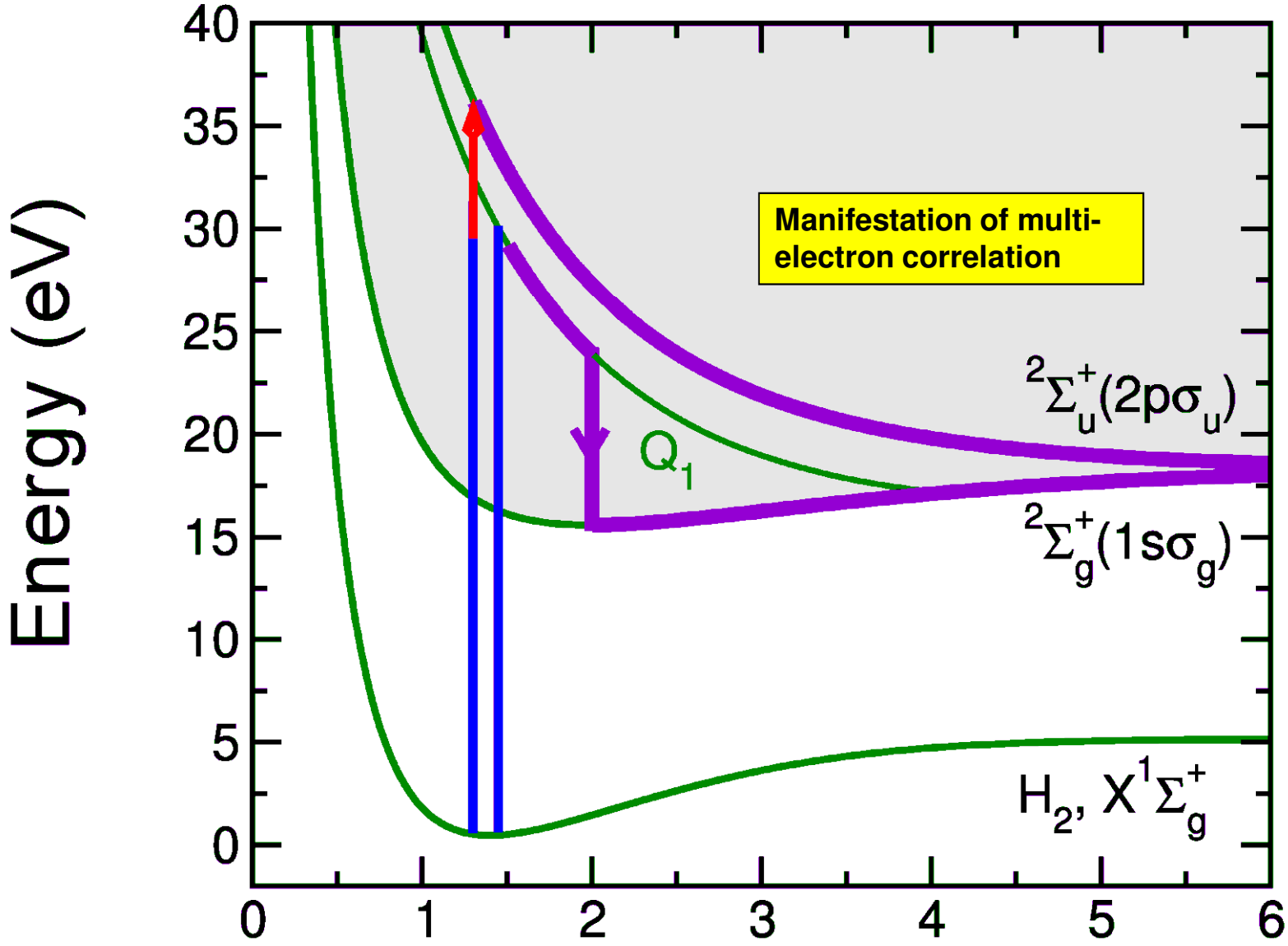
(1) Use the laser to convert „g“ into „u“ or „u“ into „g“ (remember electron localization with CEP-stable few-cycle pulses)

(2) Take one of the allowed terms in the expression shown above and change the angular momentum of the photoelectron from „u“ to „g“ or from „g“ to „u“

Mechanism I


Energy (eV)
40
35
30
25
20
15
10
5
0
0
1
2
3
4
5
6
Coupling of electronic and nuclear degrees of freedom
$^2\Sigma_u^+(2p\sigma_u)$
Q_1
$^2\Sigma_g^+(1s\sigma_g)$
H_2, $X\,^1\Sigma_g^+$

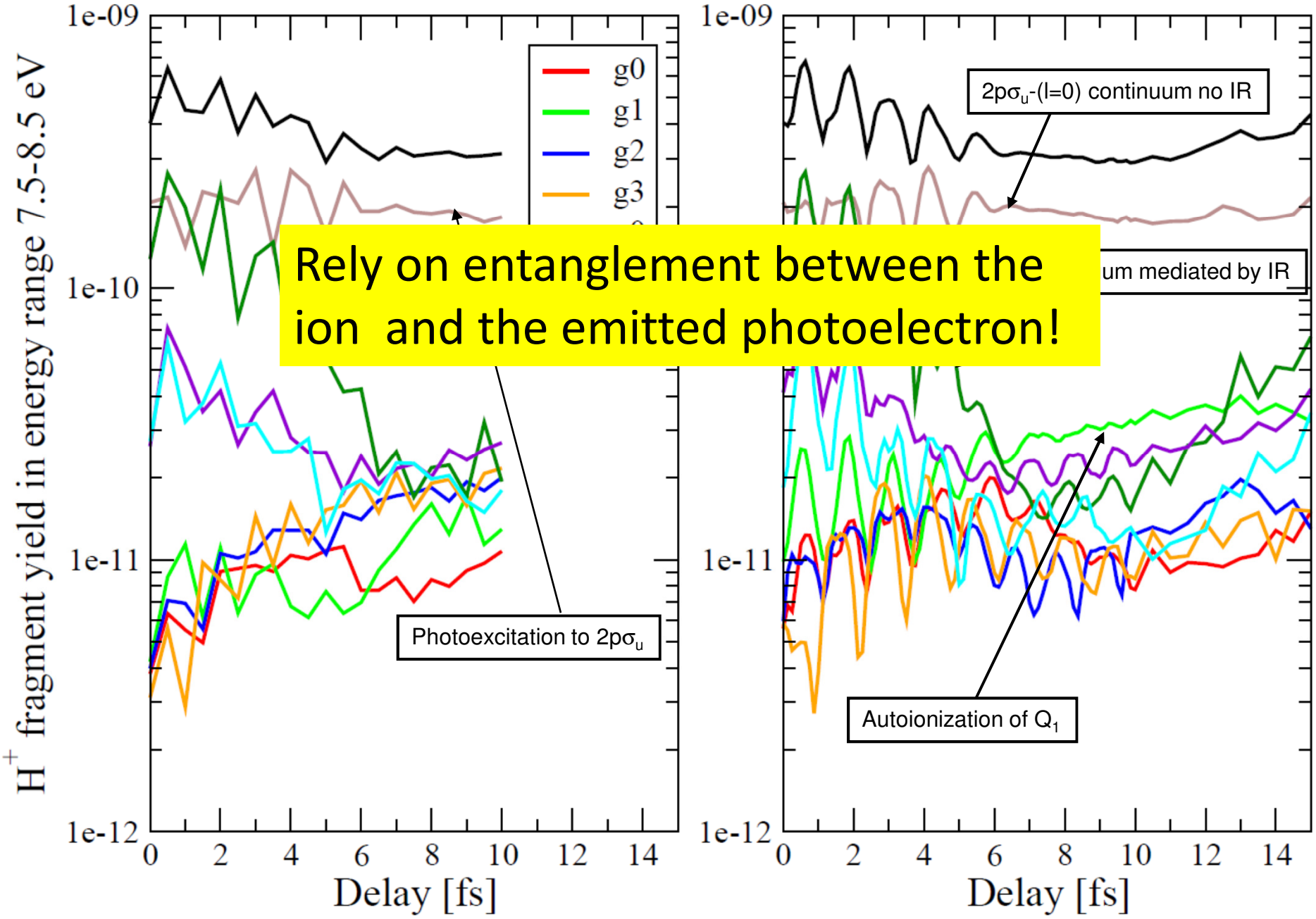
Mechanism II




Collaboration with F. Martin and co-workers
Copyright 2005. Barcelona Supercomputing Center - BSC

Without the auto-ionizing states

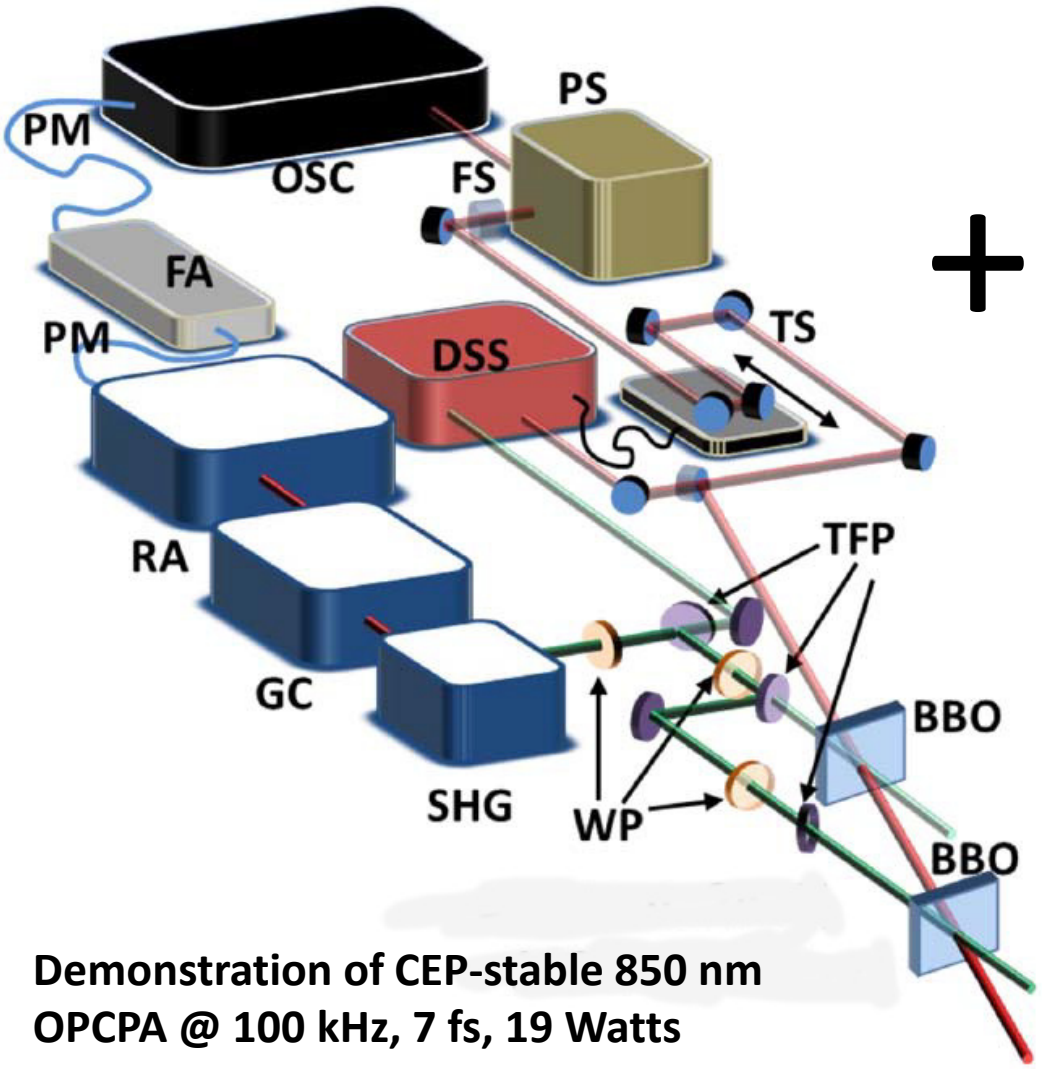
Full calculation



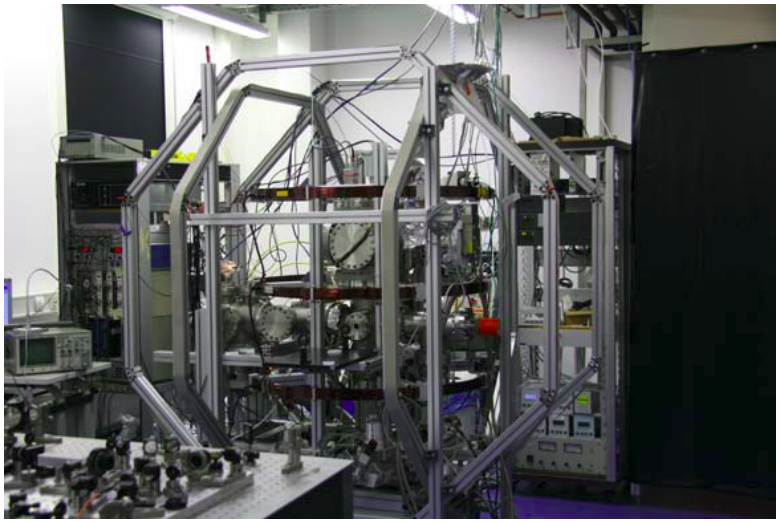
Rely on entanglement between the ion and the emitted photoelectron!

The future

Development of a high repetition rate attosecond XUV+IR experiment



+



COLTRIMS apparatus, commissioning completed, preparation for HHG + two-color, XUV+IR interferometer

Demonstration of CEP-stable 850 nm OPCPA @ 100 kHz, 7 fs, 19 Watts

Federico Furch et al., Optics Letters 42, 2495 (2017)