Attosecond Time-resolved Electron Dynamics

Marc Vrakking
FOM Institute for Atomic and Molecular Physics (AMOLF)
Kruislaan 407, 1098 SJ Amsterdam; m.vrakking@amolf.nl

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*** Prologue ***

#1 Single-active electron dynamics vs multi-electron dynamics

(my summer 2005 Pop Quiz)

#2 Photo-absorption on attosecond timescales

(another summer 2005 Pop Quiz)

#3 Perturbative vs non-perturbative electron dynamics

#4 Nuclear dynamics vs electron dynamics

#5 Isolated attosecond pulses vs attosecond pulse trains

1. Single-active electron dynamics vs collective electron dynamics

" What **IS** a plasmon resonance? "

2.2. The classical picture

The cooperative motion of electrons leading to giant resonances can be described classically. Suppose the photon 'strikes' one of the atomic electrons, which, after acquiring the photon's energy, starts moving relative to the nucleus. Owing to strong interelecronic repulsion, this motion must affect the other electrons, and eventually the whole electronic shell begins to move. Being attracted by the atomic nucleus, the shell is forced to return, and thus starts to oscillate with a characteristic frequency Ω. The magnitude of this frequency is clearly typical of a given atom, of the oscillating shell's position within the atom and of the number of electrons it contains. The oscillation, once excited, would exist forever if it were unable to lose its energy by transferring it either to the individual electrons of the oscillating shell or to electrons located outside it. This leads to a damping, which is described by the so-called half-width γ of the resonance. The important characteristic of a giant resonance [4] is the dimensionless ratio Ω/γ which, roughly speaking, tells us how many oscillations occur in a giant resonance before it dies out. Another important characteristic is its total oscillator strength F, which

M.Y. Amusia and J. P. Connerade, *The Theory of Collective Motion Probed by Light*, Rep. Prog. Phys. 63, 41 (2000)

But…

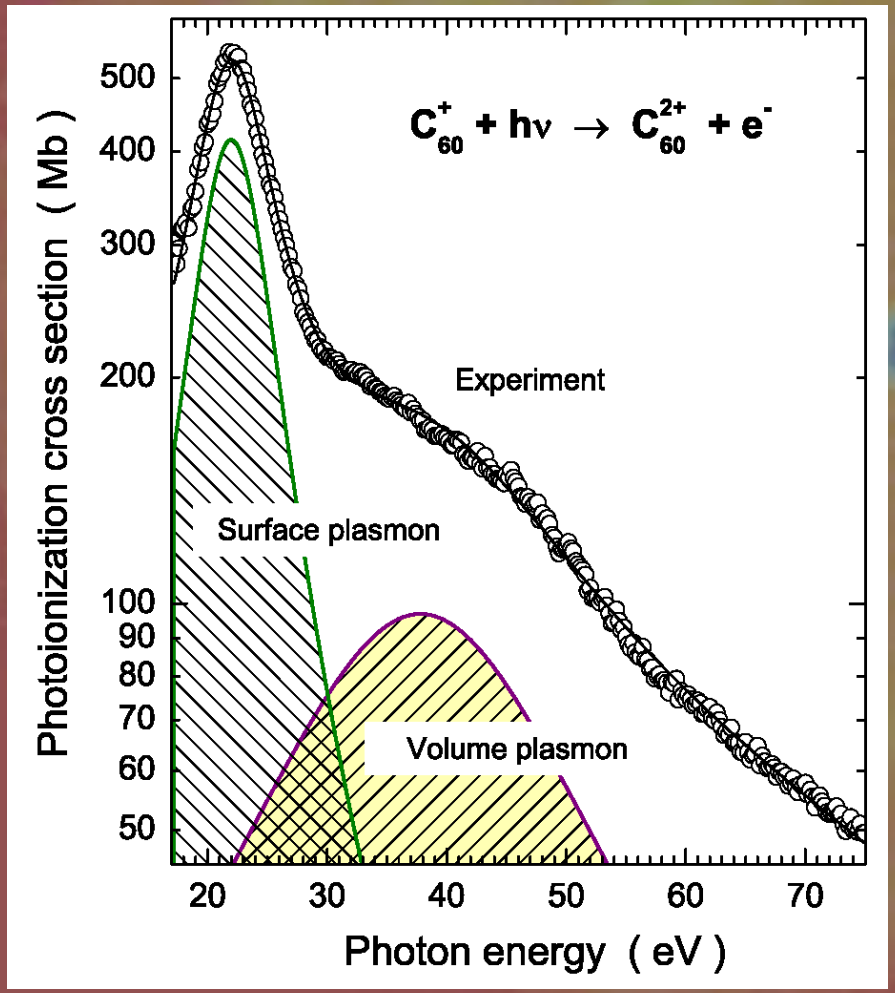
Aren’t all electrons involved in the plasmon resonance equivalent, experiencing the same interaction with the light source?

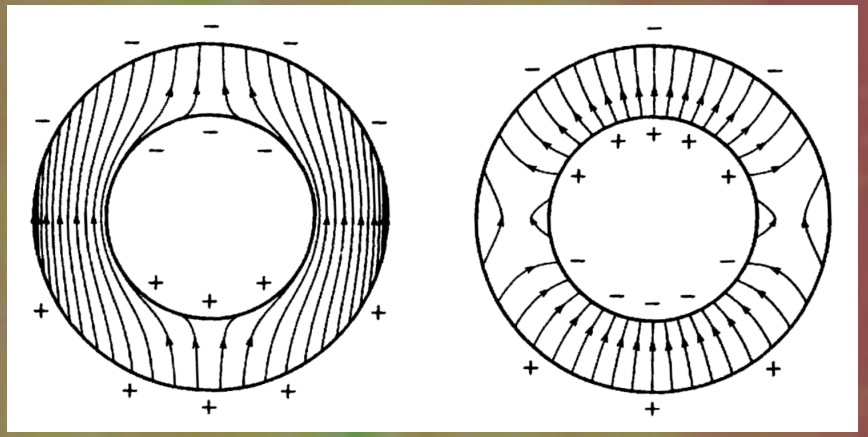
➔ Two possible, mutually exclusive scenarios for the collective electron excitation:

1) One electron absorbs a photon (dipole transition) and the collective electron oscillation is established as a result of subsequent electron-electron coupling

2) All electrons collectively interact with the light field and collectively “absorb the photon”

Studying collective electron excitations in C_{60}





Surface plasmon (22 eV)

Volume plasmon (38 eV)

How is the collectivity established?

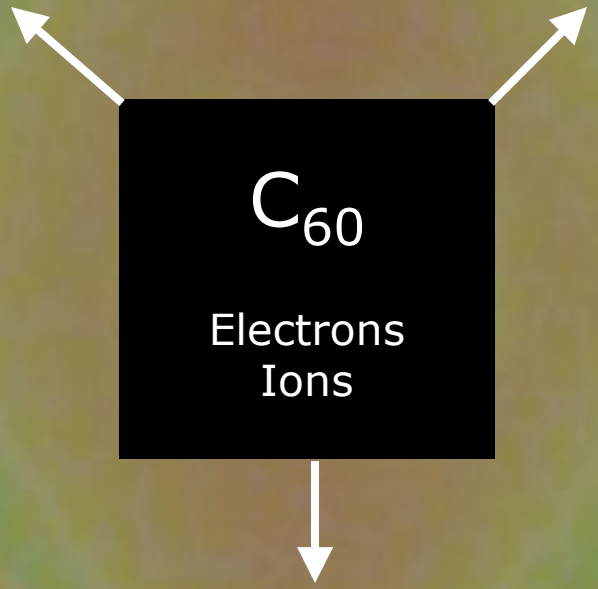
How is the collectivity lost?

S.W.J. Scully et al. Phys. Rev. Lett. 94, 065503 (2005).

Connecting Different Studies on C_{60}

XUV Photo-absorption @ 20-50 eV, establishment of volume & surface plasmons, with (sub)-femtosecond lifetime

IR femtosecond laser excitation (Campbell, Hertel) – 70-150 femtosecond timescales inferred for electron-electron and electron-phonon coupling



Far-infrared multi-photon excitation to total internal energy of ~40 eV followed by thermionic emission on timescale up to 100 μs

2. First: look at a two-level system

" What does simple resonant excitation look like on sub-cycle timescales? "

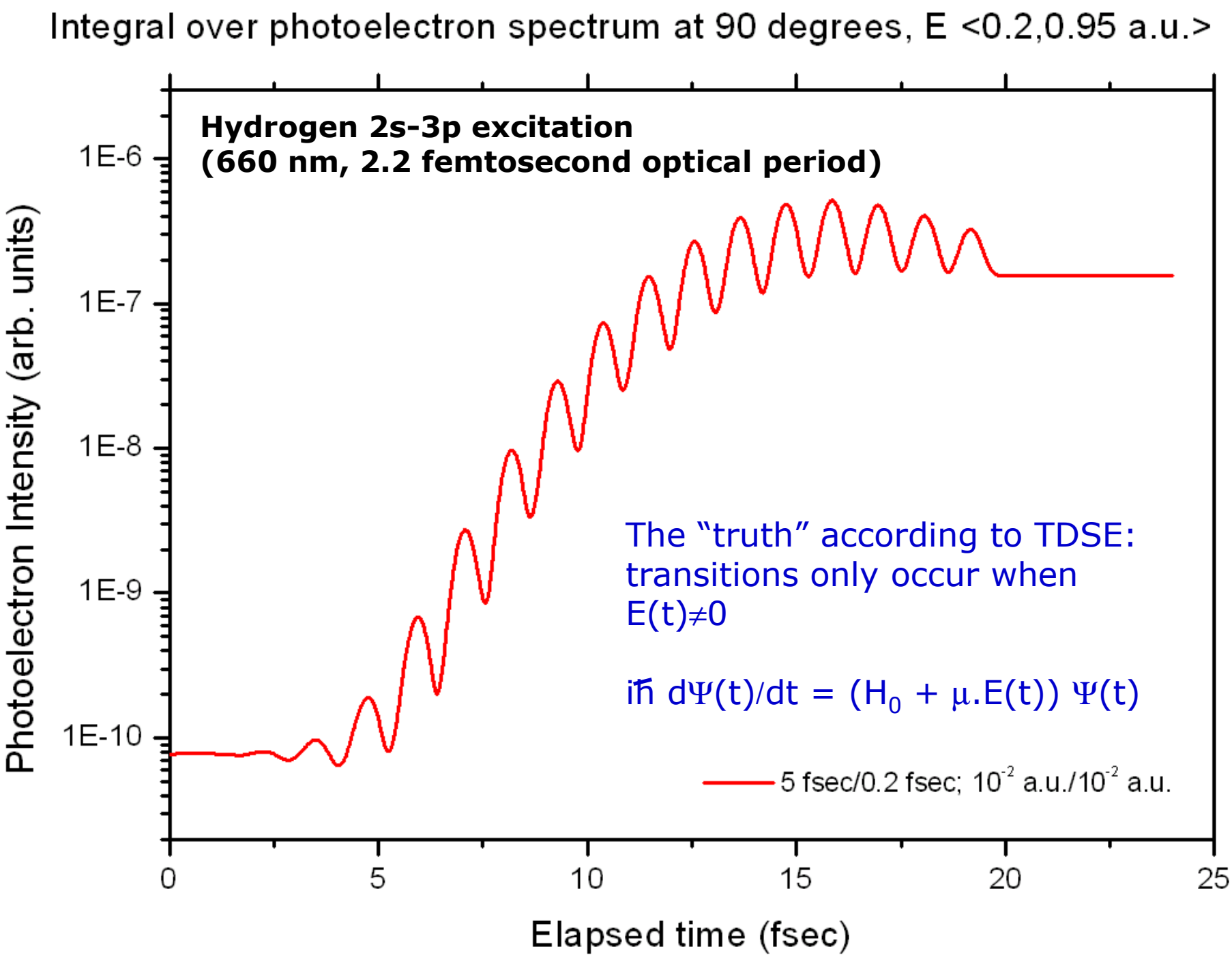
Conventional wisdom: a two-level system subjected to resonant monochromatic radiation will start to perform Rabi oscillations

$$\Gamma(t) = N(t)\exp\left(-\frac{\mathcal{E}^2 f^2(t)}{\omega_L^3}\Phi(\gamma(t),\theta(t))\right). \qquad (17)$$

The subcycle dependence in the preexponential factor $N(t)$ can be ignored (up to the electric-field envelope). Indeed, at $\gamma \ll 1$ and $\gamma \approx 1$, ionization is strongly peaked around $\phi(t) = \omega_L t + \varphi_0 = \pi k$ and we only need to know $N(\phi = \pi k)$. At $\gamma \gg 1$, the subcycle dependence disappears, and knowing $N(\phi = \pi k)$ is again sufficient. Hence, it is sufficient to include the time dependence in $N(t)$ via the envelope $\mathcal{E}f(t)$ only.

Sub-cycle dependence of ionization rates:

Yudin & Ivanov, Phys. Rev. A 64, 013409 (2001)


Integral over photoelectron spectrum at 90 degrees, E <0.2,0.95 a.u.>
Hydrogen 2s-3p excitation
(660 nm, 2.2 femtosecond optical period)
Photoelectron Intensity (arb. units)
1E-6
1E-7
1E-8
1E-9
1E-10
0
5
10
15
20
25
Elapsed time (fsec)
The "truth" according to TDSE:
transitions only occur when
E(t)≠0
iħ dΨ(t)/dt = (H0 + μ.E(t)) Ψ(t)
5 fsec/0.2 fsec; 10-2 a.u./10-2 a.u.

3. Perturbative vs non-perturbative electron dynamics

" When are our investigations relevant to the outside world, incl. real applications? "

Nature, real applications: linear regime

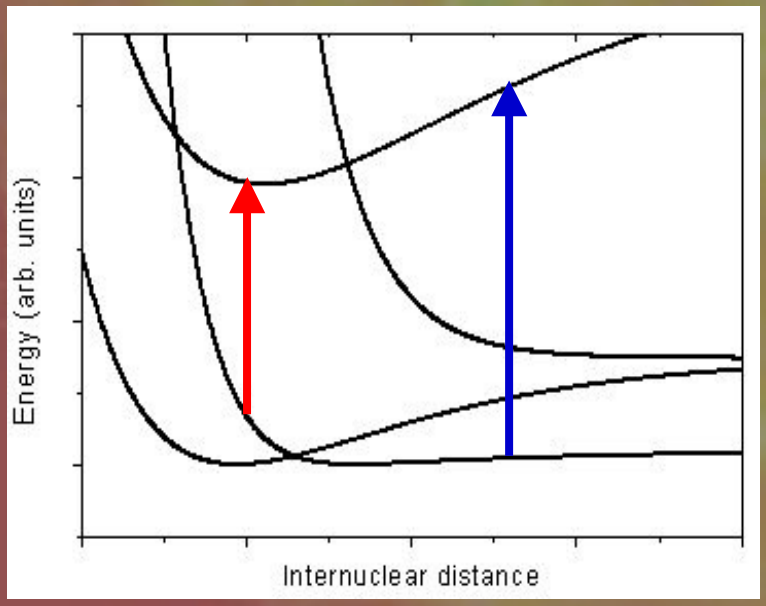
Photo-induced processes are one-photon processes, independent of pulse intensity, shape, phase, etc.

Femtosecond laser laboratory: non-linear regime

Multi-photon & Above-threshold ionization, High Harmonic generation, Enhanced ionization, Dynamic Alignment, Plasma resonances in large clusters, Coherent & optimal control

4. Nuclear dynamics vs electron dynamics

" Studying electron dynamics on short timescales requires a fundamentally different approach "

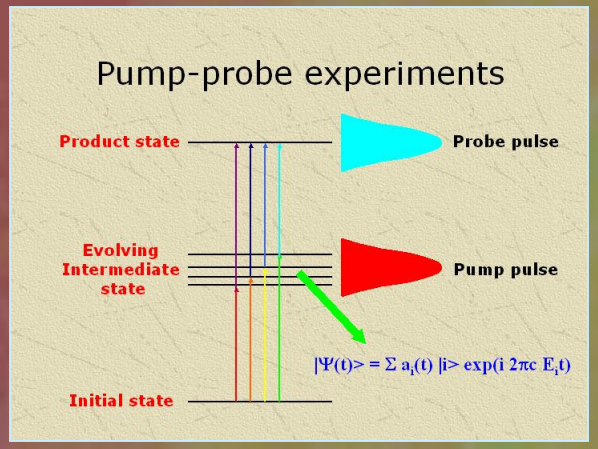


The electronic energy adapts to the configuration of the molecule and manifests itself in the time-dependent absorption spectrum of the molecules

In other words: the nuclear dynamics is guided by and is a consequence of electron dynamics

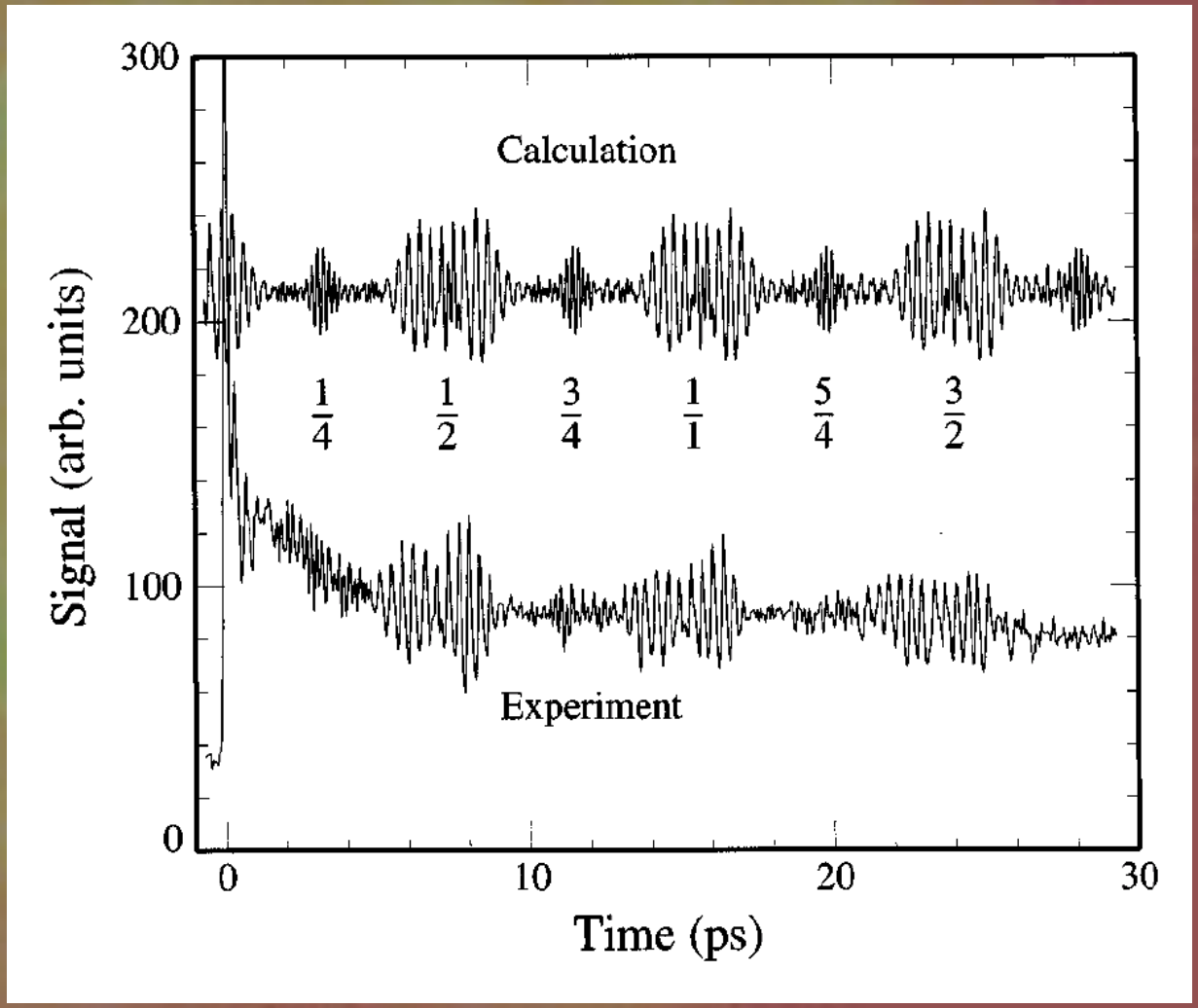
This has been exploited in many femtosecond time-resolved experiments

Femtosecond Pump-Probe Spectroscopy: vibrational wavepackets



Ionization of Br_2 by two time-delayed pulses (560 nm & 290 nm)

A. Stolow et.al.
Phys. Rev. A 54, R37 (1996)

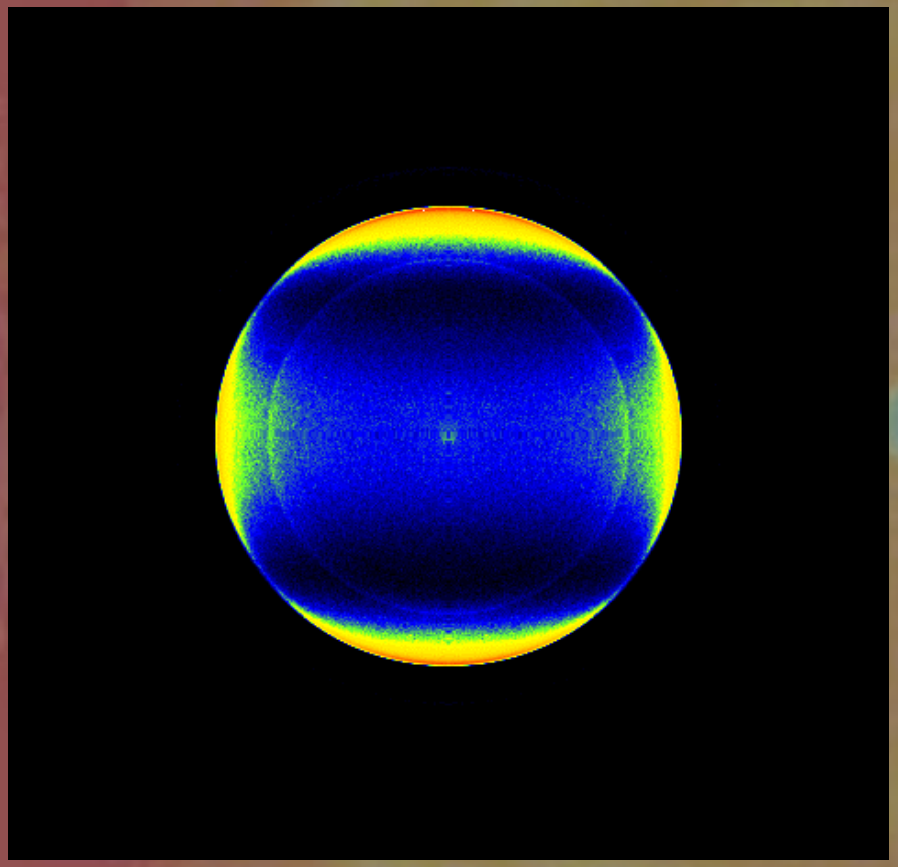


Current challenges to attosecond science

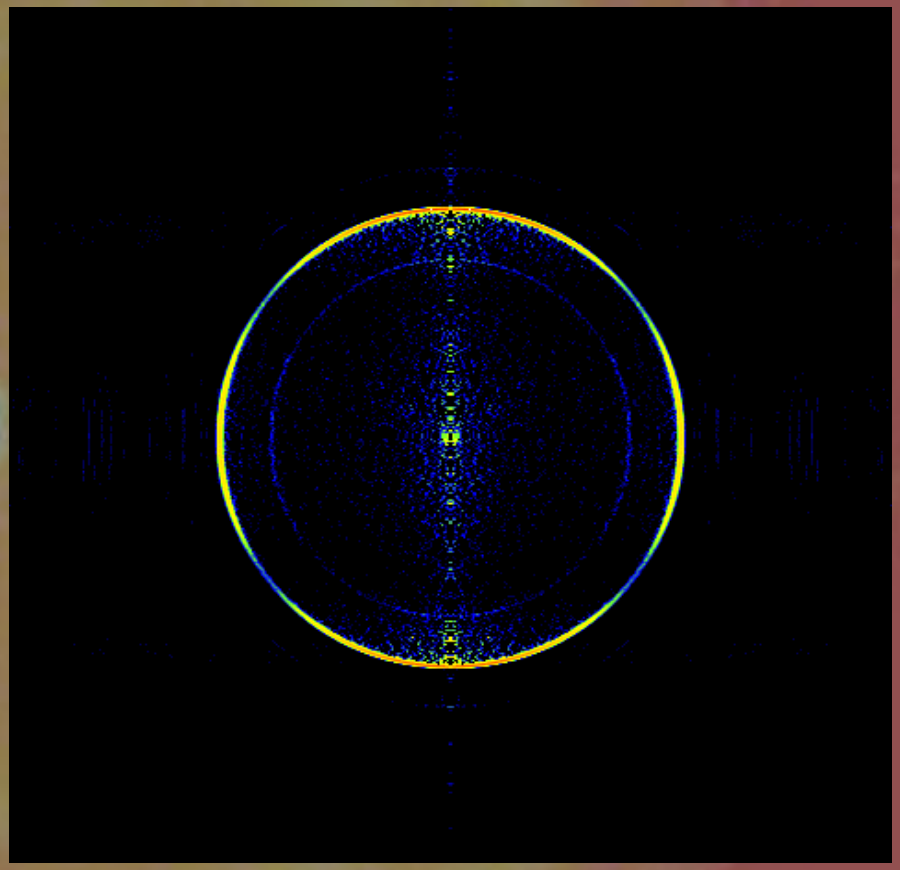
+ scheme to generate attosecond laser pulses
+ scheme to characterize attosecond laser pulses
+ XUV beam delivery optics
? scheme to initiate attosecond electron dynamics
  → coherent excitation over ≥5 eV bandwidth;
  → current attosecond pulses: XUV-soft x-ray
    photochemistry: UV/VUV

? scheme to probe the attosecond electron dynamics
  → ionization = "universal" outcome of attosecond
    experiment
  → signature in angle- and energy-resolved
    photoelectron and -ion spectroscopy

2D Imaging – a typical experimental result and the mathematical treatment

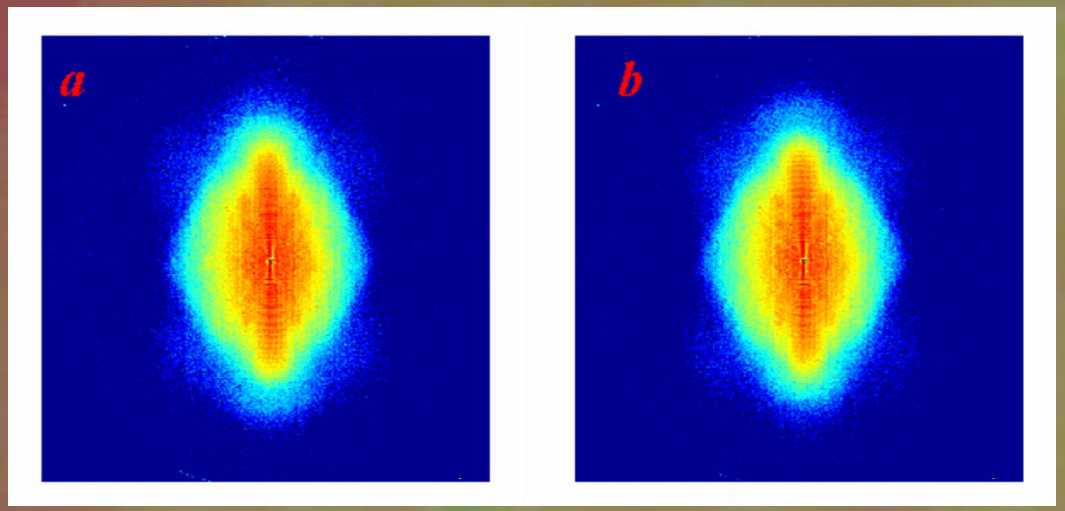


Raw image for 2-photon ionisation of Ar by 532 nm light

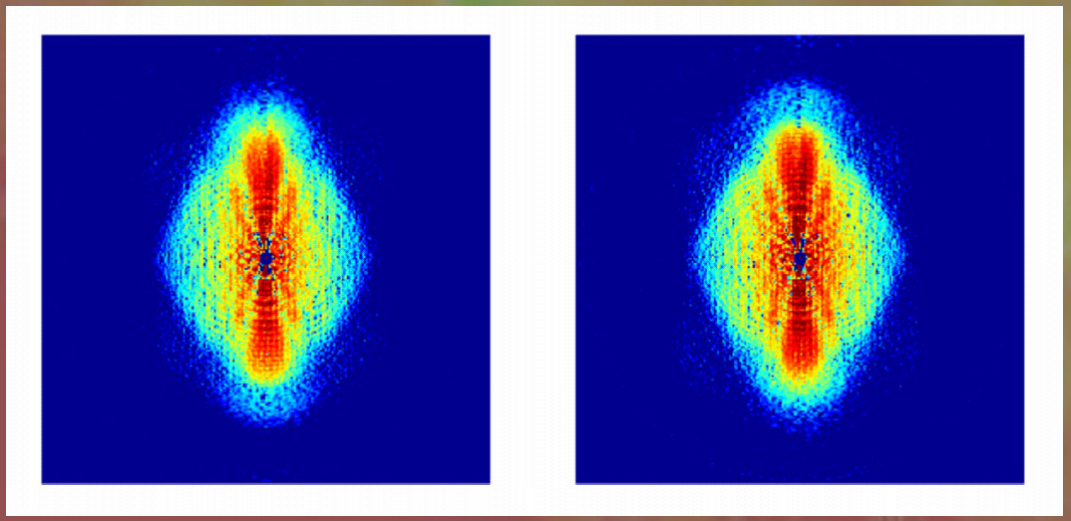


Slice through the 3D velocity distribution, obtained by Abel inversion of the image Dv/v = 1% (N.B. also use iterative technique)

2D Imaging as a tool for the determination of the few-cycle CEP



Raw images for two values of the carrier envelope phase



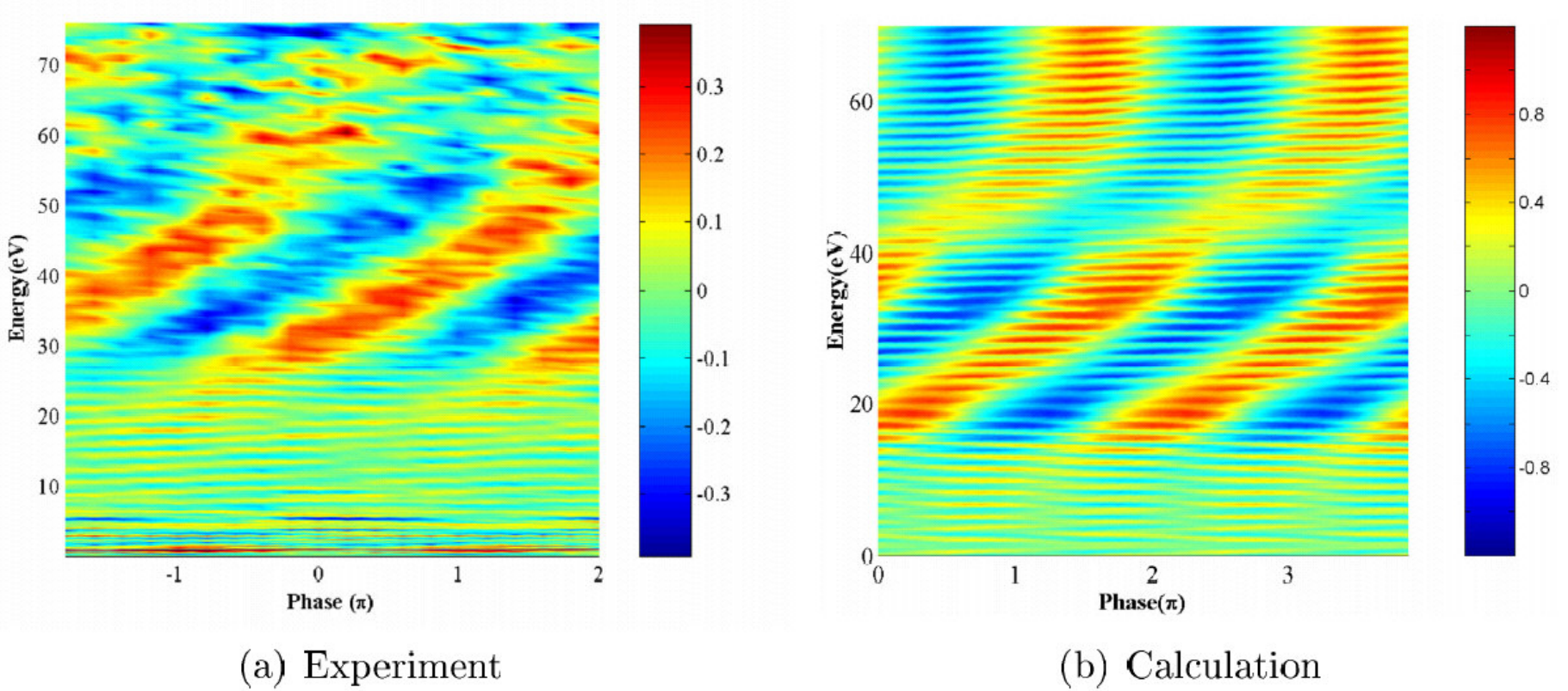
Corresponding inverted images (slice through the 3D velocity distribution)

Ar, 6 fs, 1.2×10^{14} W/cm^2

(collaboration w. Ferenc Krausz)

2D Imaging as a tool for the determination of the few-cycle CEP

Asymmetry = $(P(E)_{up}-P(E)_{down})/(P(E)_{up}+P(E)_{down})$



Ar, 6 fs, 1.2×10^{14} W/cm^2

5. Isolated attosecond pulses vs attosecond pulse trains

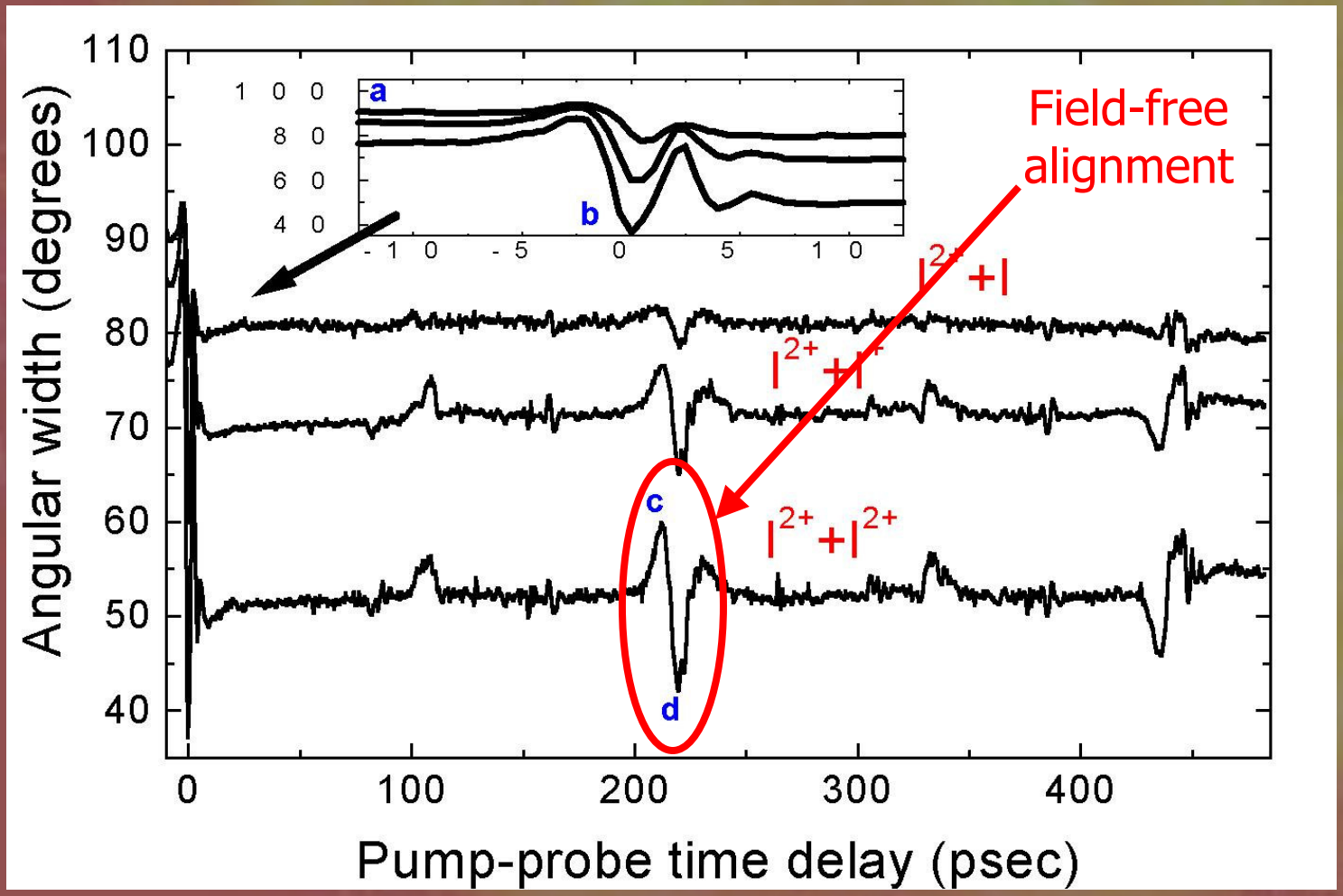
A. High harmonic generation using a few-cycle (CEP-stabilized) laser pulse or using a pulse with a time-varying polarization → *isolated attosecond pulses*

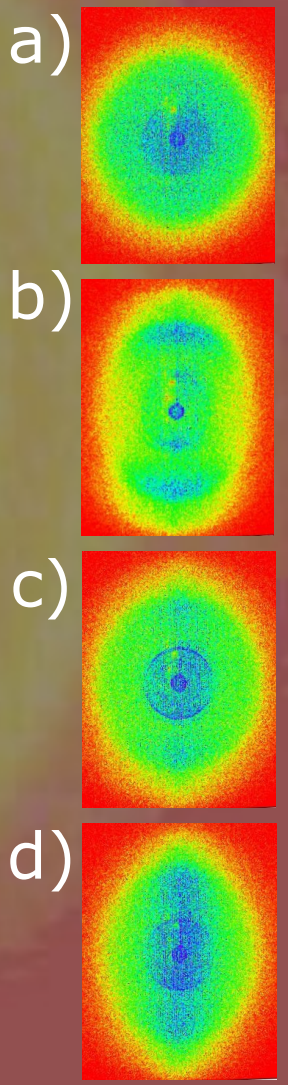
Good for pump-probe experiments

B. High harmonic generation using a many-cycle laser pulse → *train of attosecond laser pulses*

Good for interferometry experiments (Anne L'Huillier)

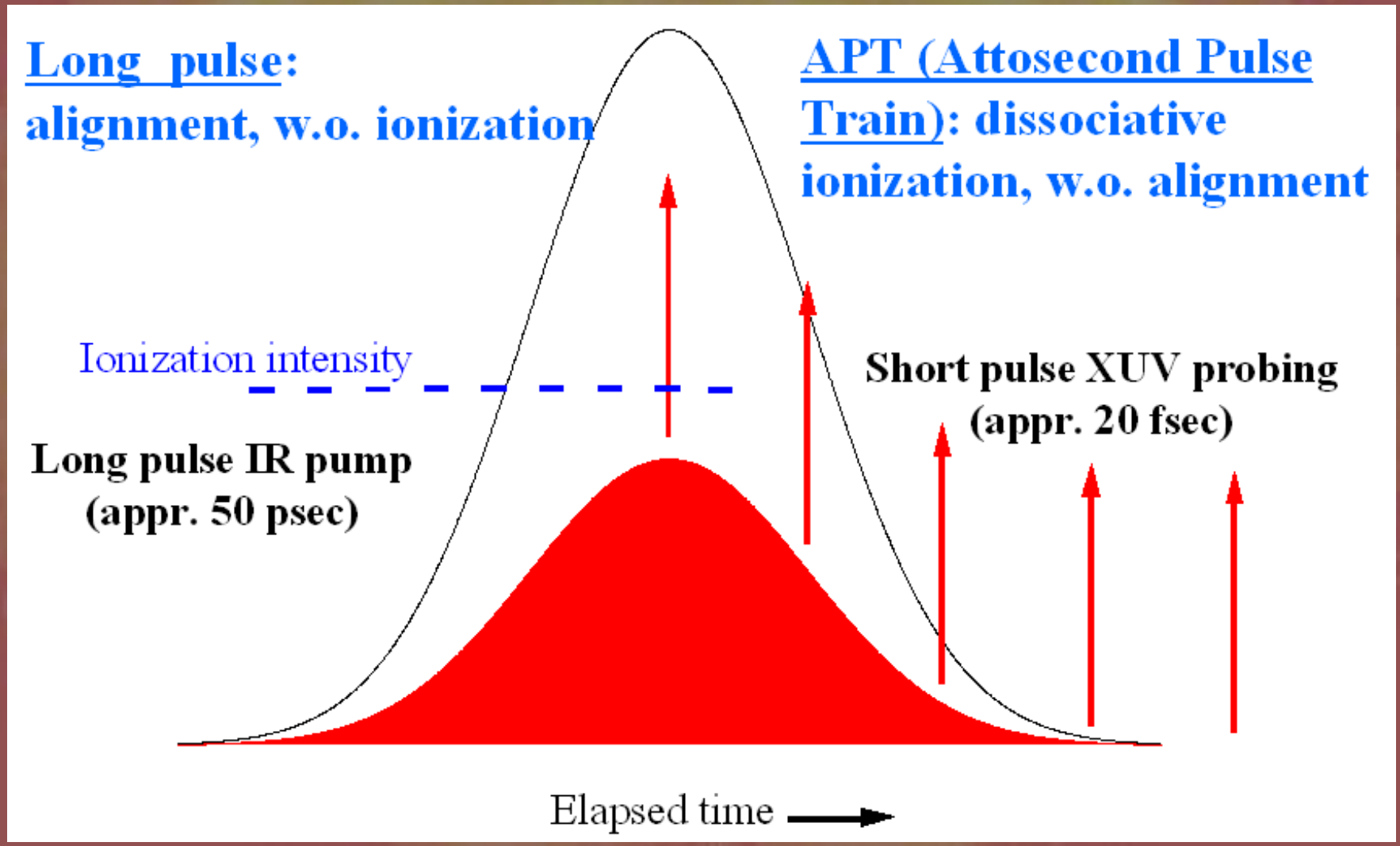
Strongly Driven Electrons in Molecules:
Dynamic alignment





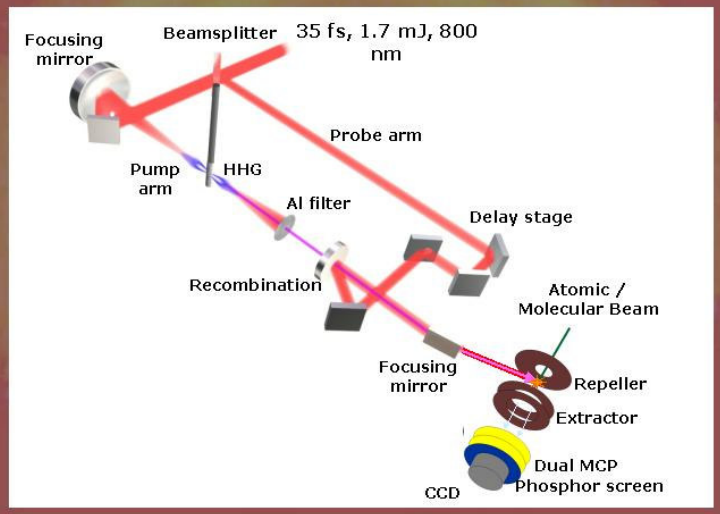
F. Rosca-Pruna and M.J.J. Vrakking, Phys. Rev. Lett. 87, 153902 (2001).

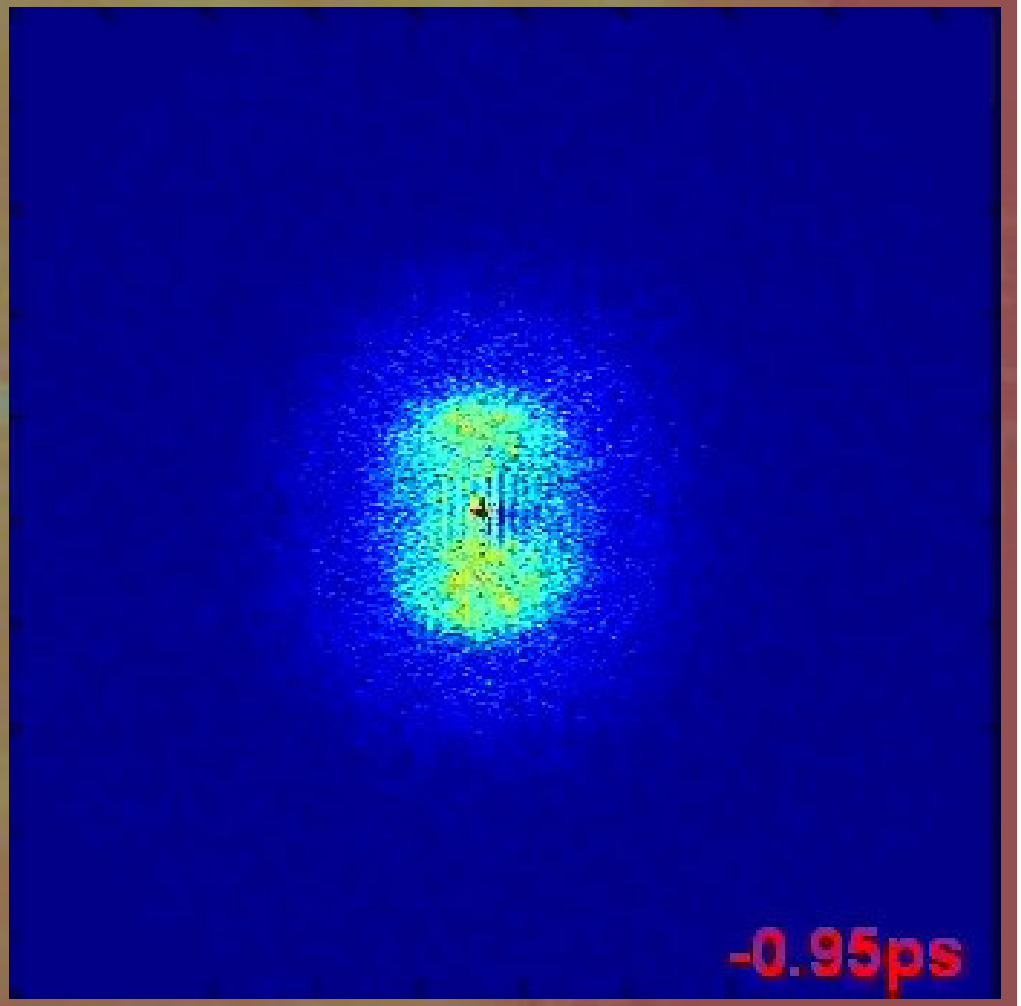
Towards probing electron dynamics during molecular alignment



Towards probing electron dynamics during molecular alignment (with Lund)

Experimental Scheme (with Lund):
strong IR field (10^{13} W/cm^2)

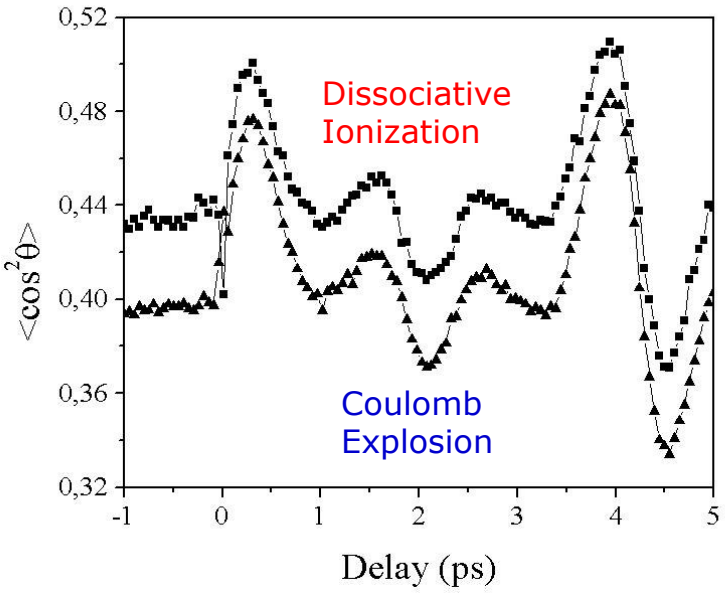


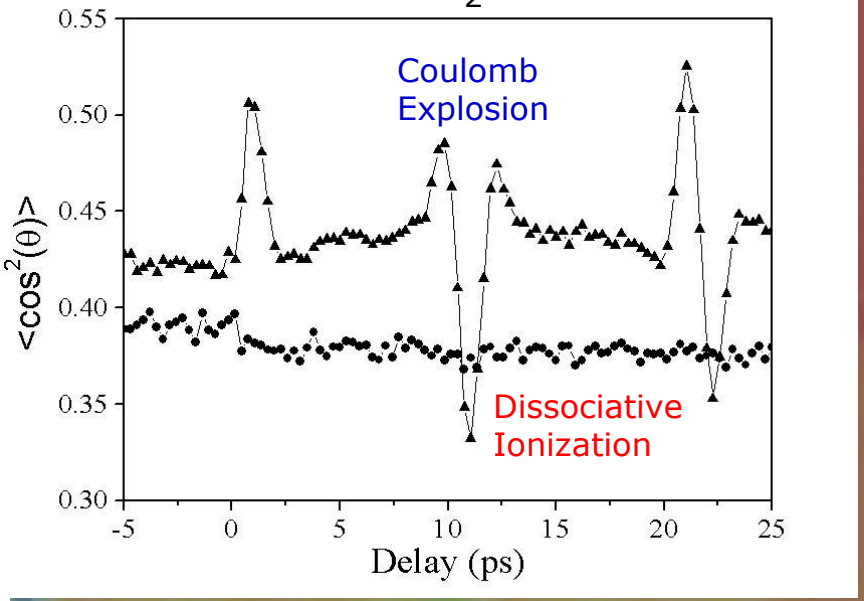


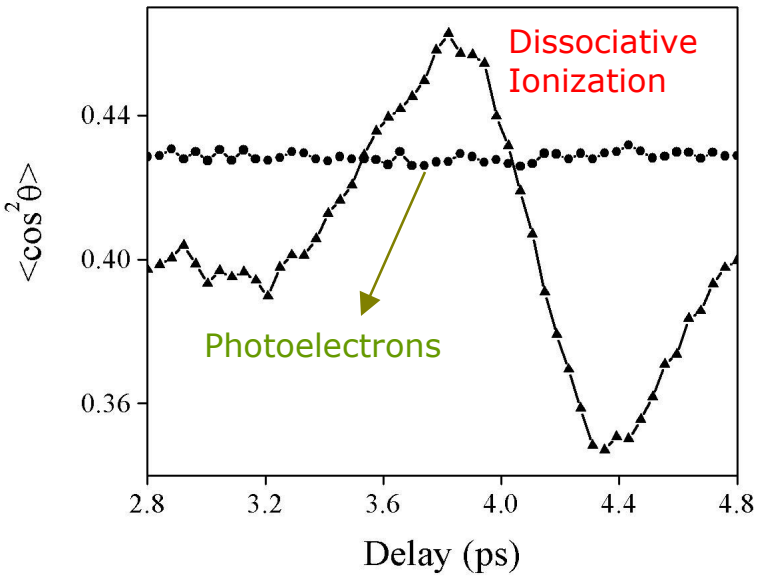
Images of N^+ formation as a function of the time delay between the IR pump and the XUV attosecond pulse train (APT)

F. Lépine et al., (submitted for publication)

(collaboration w. Anne L'Huillier)



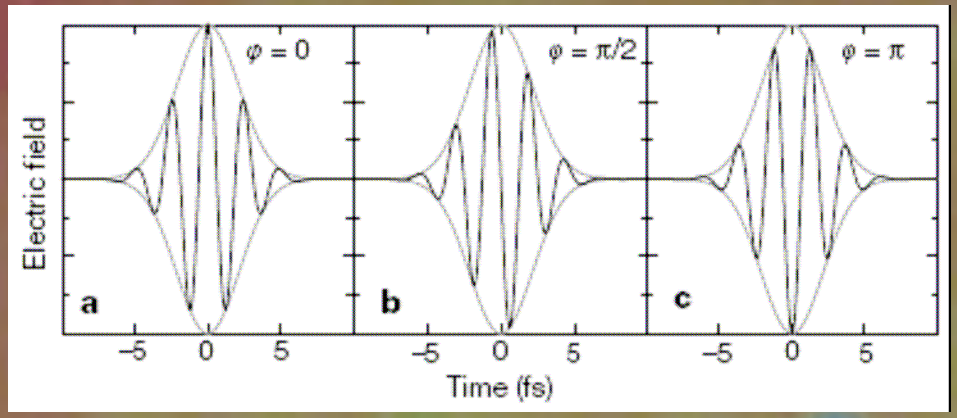




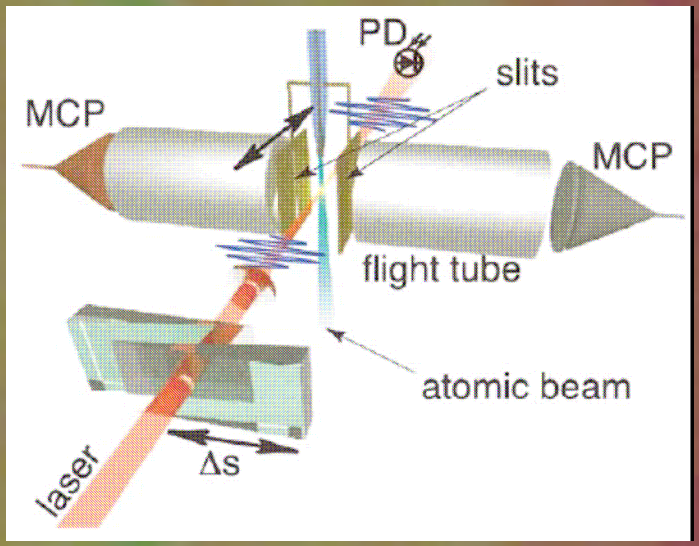
Future:

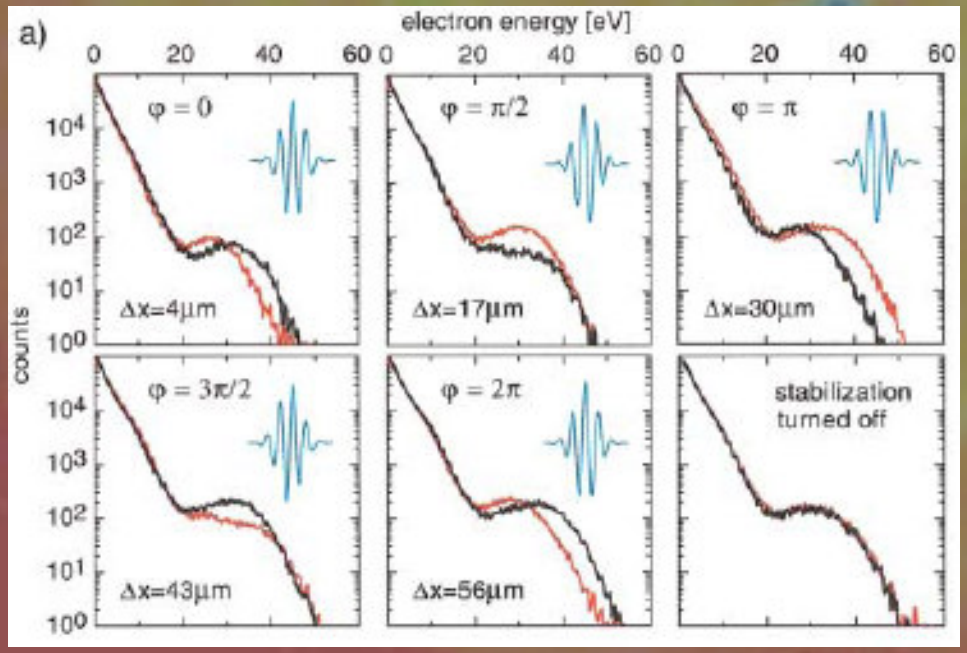
- ✓ Using attosecond pulses to probe electron dynamics responsible for molecule alignment
- ✓ Using XUV pulses to monitor chemical reactions by time-dependent electron diffraction

Carrier-Envelope Phase Effects in Atomic Ionization



ATI in Xenon

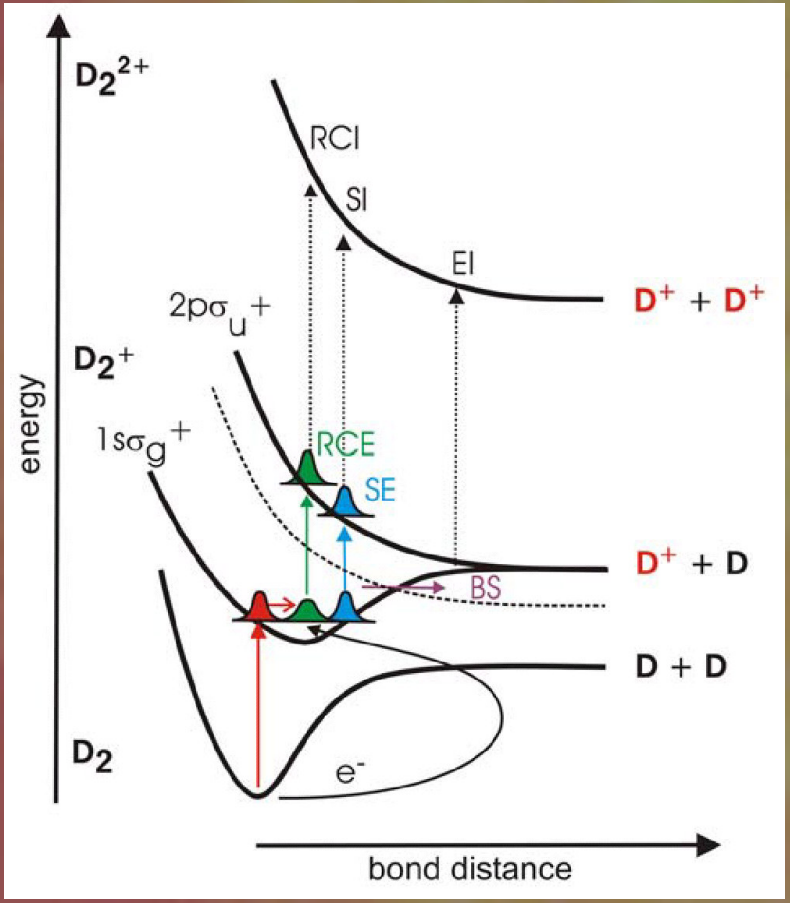


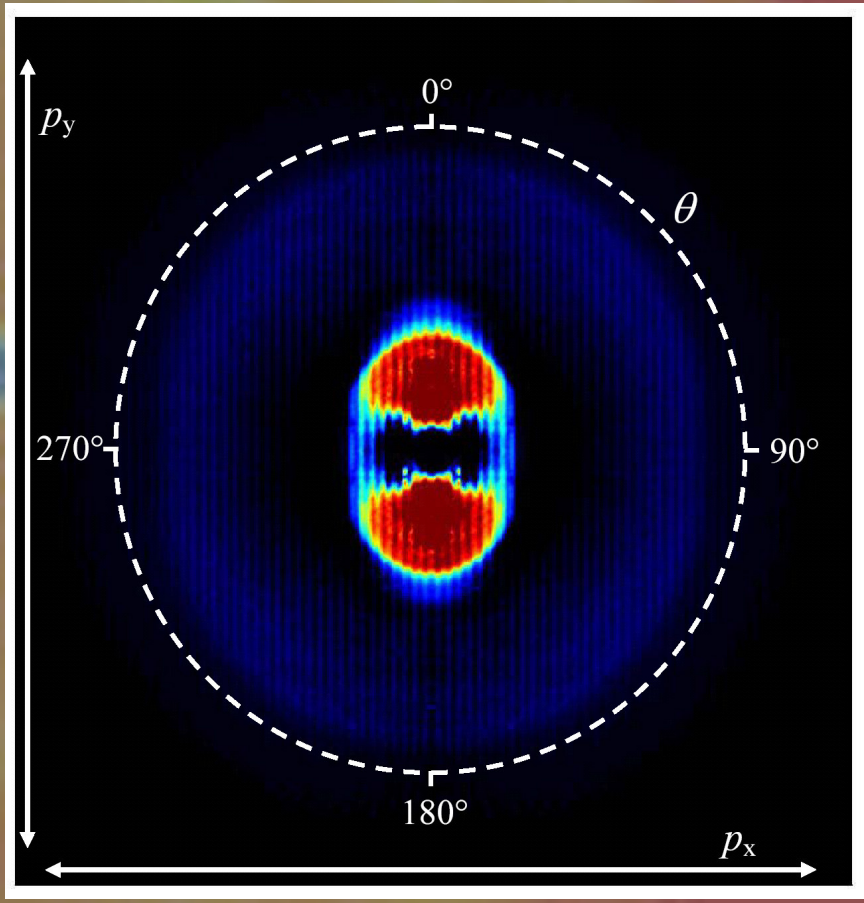


Can carrier envelope phase control electron motion *inside* molecules?

Paulus *et al*, *Nature* **414** (2001) 182;*PRL* **91** (2003) 253004

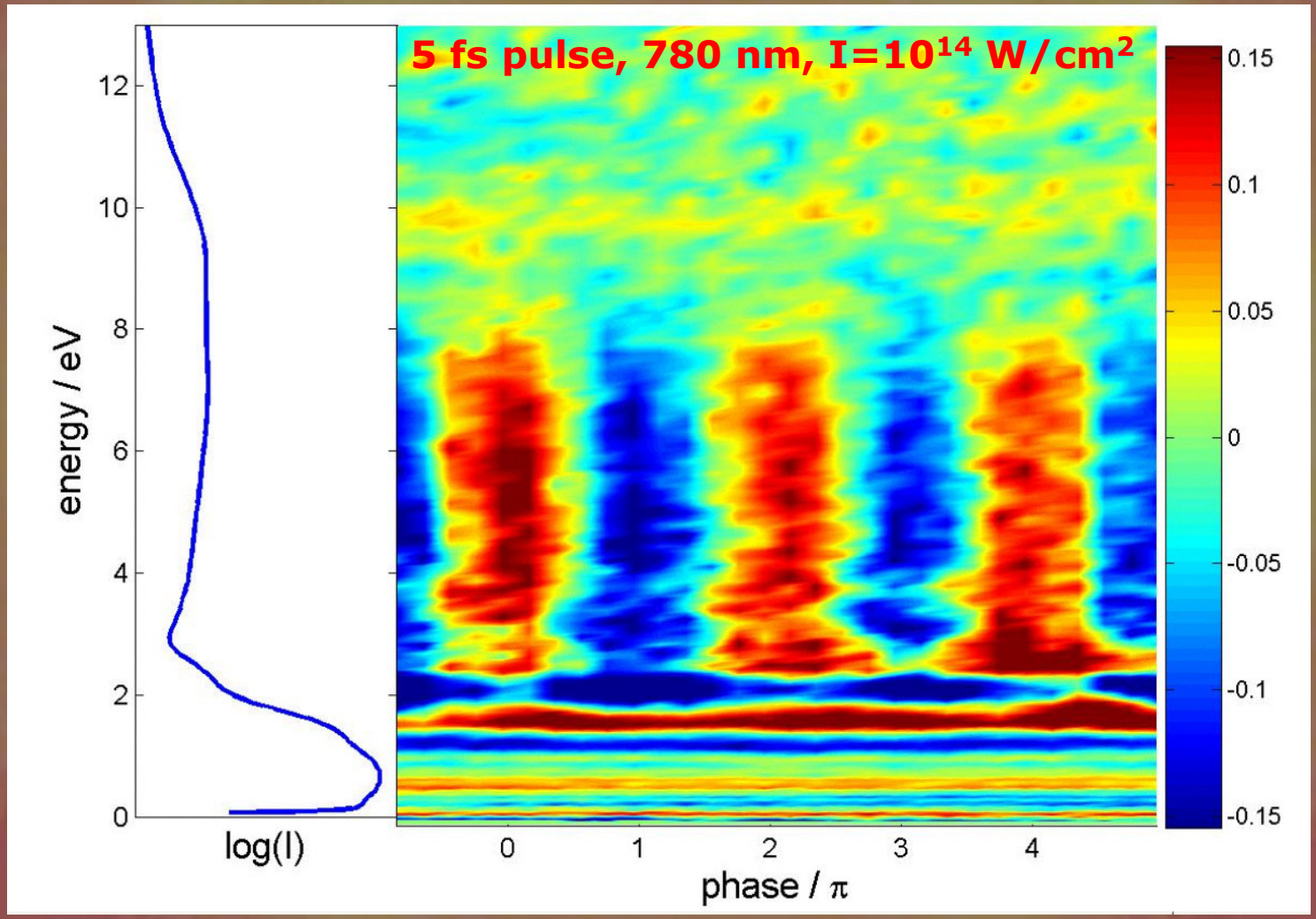
Attosecond electron dynamics in molecules
CEP control of electron localization





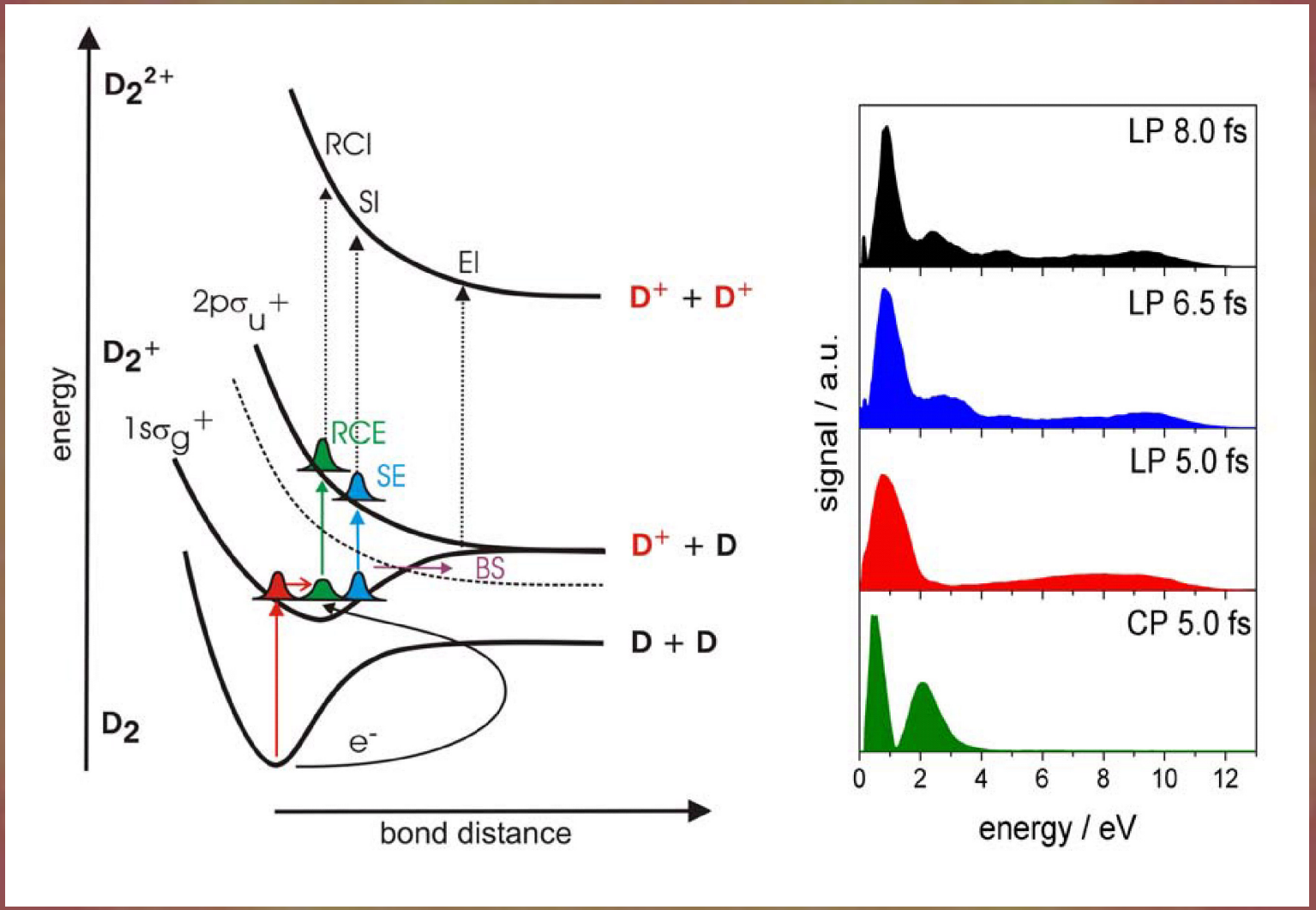
Recent experiment: angle-resolved D^+ ion imaging using CEP-locked few-cycle laser pulses (w. Ferenc Krausz)

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



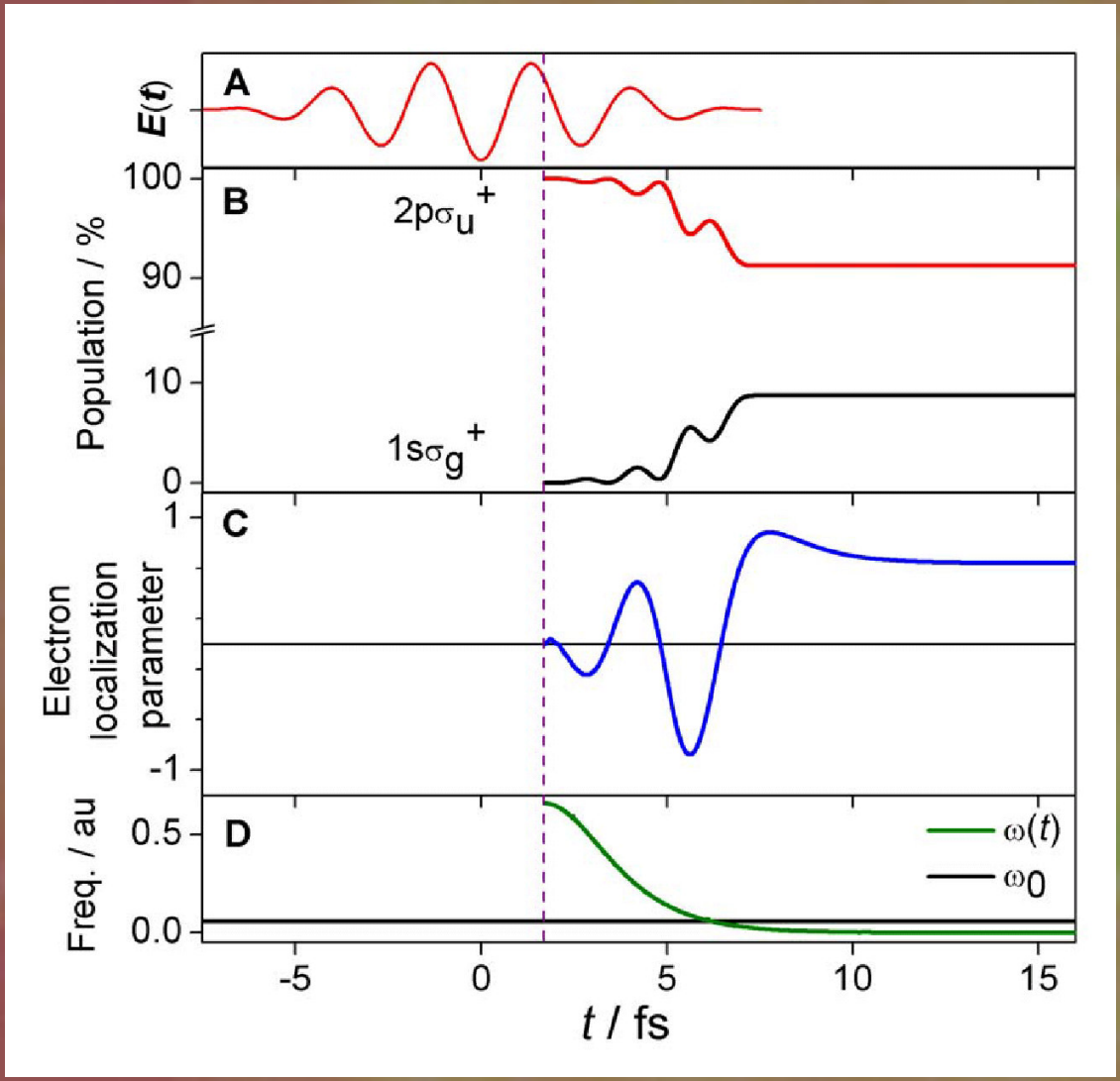
M. Kling et al., Science *312*, 246 (2006)

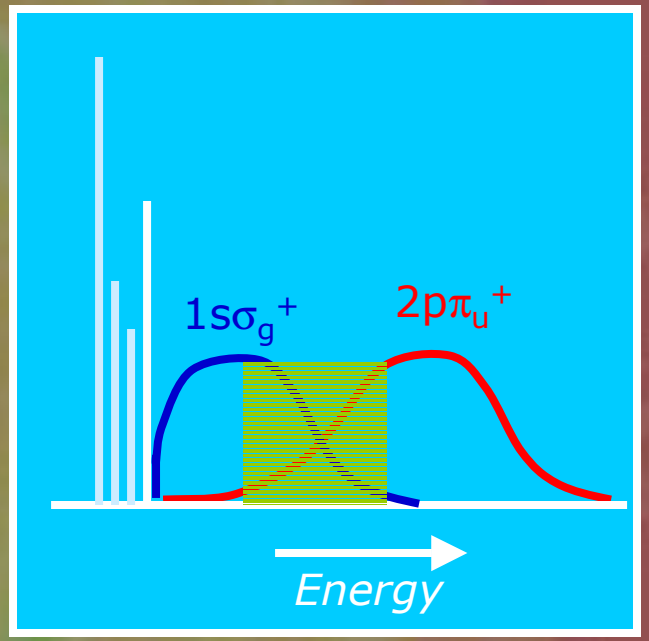
Phase Control Mechanism -1



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

Phase Control Mechanism -2

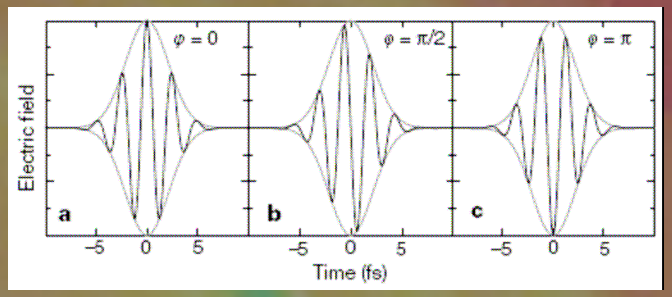


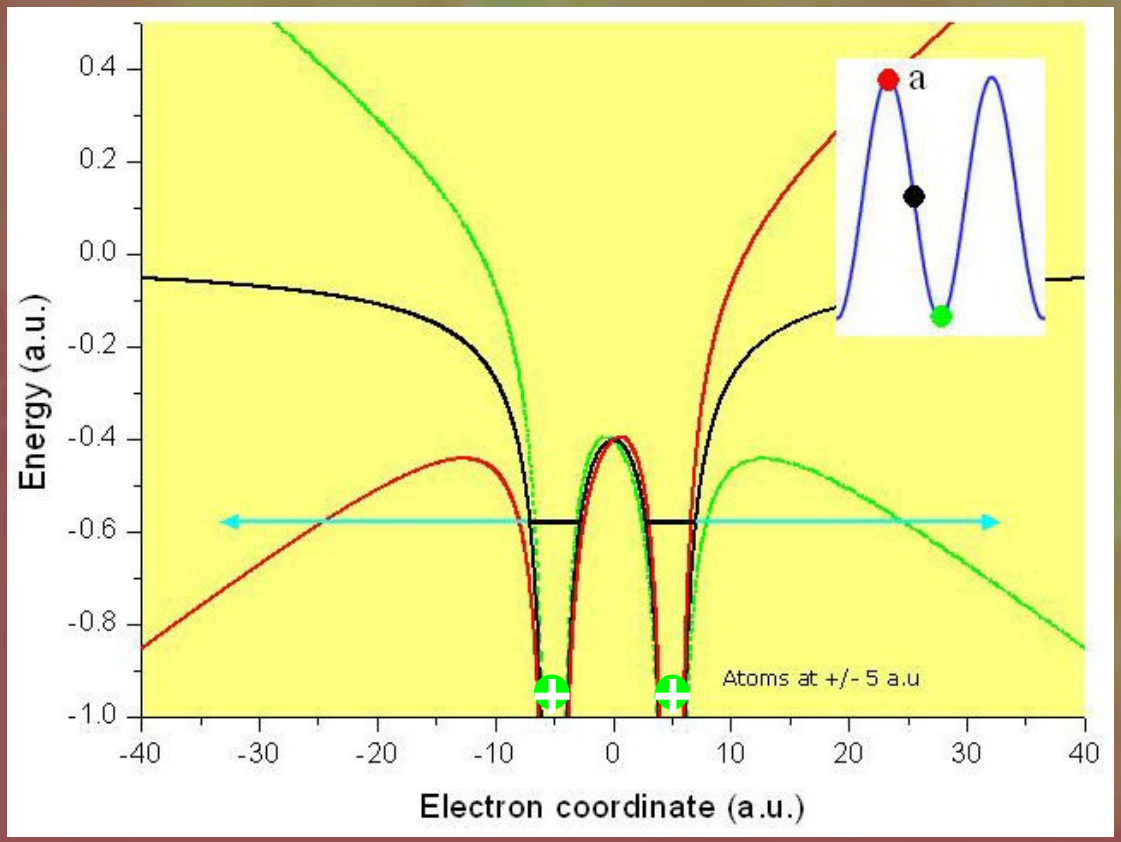


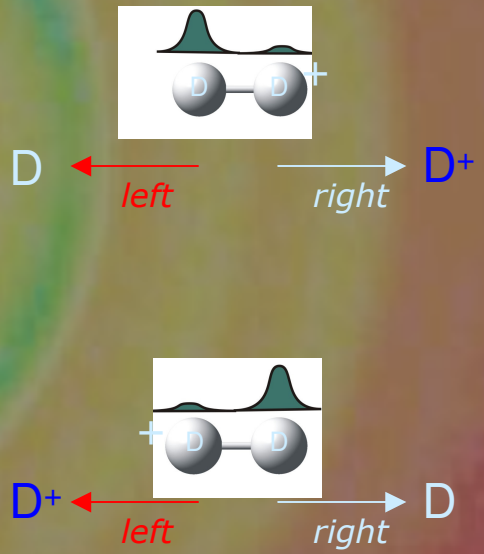
→ Preparation of coherent superposition state by stimulated emission to $1s\sigma_g^+$ state

Alternative Time-domain Picture

Carrier-envelope phase of a few-cycle laser allows *subcycle (attosecond) control of electron dynamics*







The electron oscillates between the two D+ ions, until this oscillation is impeded by the onset of a barrier between the two ions

Conclusions and Outlook

- Discussed a number of aspects relevant to attosecond science and distinguish it from earlier femtosecond work addressing nuclear dynamics (single/multi-electron excitation, photoabsorption, RWA, BO, (non)-perturbative excitation, methodology)

- Useful applications exist both for attosecond pulse trains and for isolated attosecond pulses.

- **Attosecond pulse trains**: used in attosecond electron wave packet interferometry
- **Isolated attosecond pulses**: monitoring of electron dynamics in pump-probe experiments → first steps: probing of dynamic alignment and CEP control of electron dynamics in D_2

- Eventually this work may provide insight into the elementory electronic processes that occur during photo-absorption and that accompany chemical rearrangements