Ultrafast XUV Sources and Applications

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Workshop “Emerging Sources”
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Overview: Attosecond Science from a "user" perspective

- What do we want?
- What do we use as our starting point to get this?
- What – as a result - can we presently do?
- What can we presently <u>not</u> do?
- What do we plan to do to get around this?

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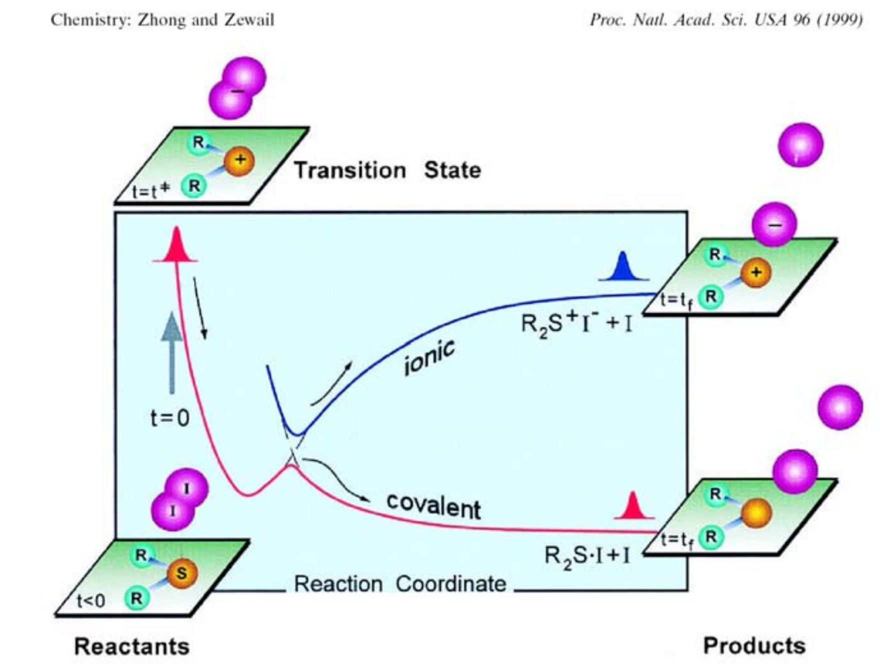
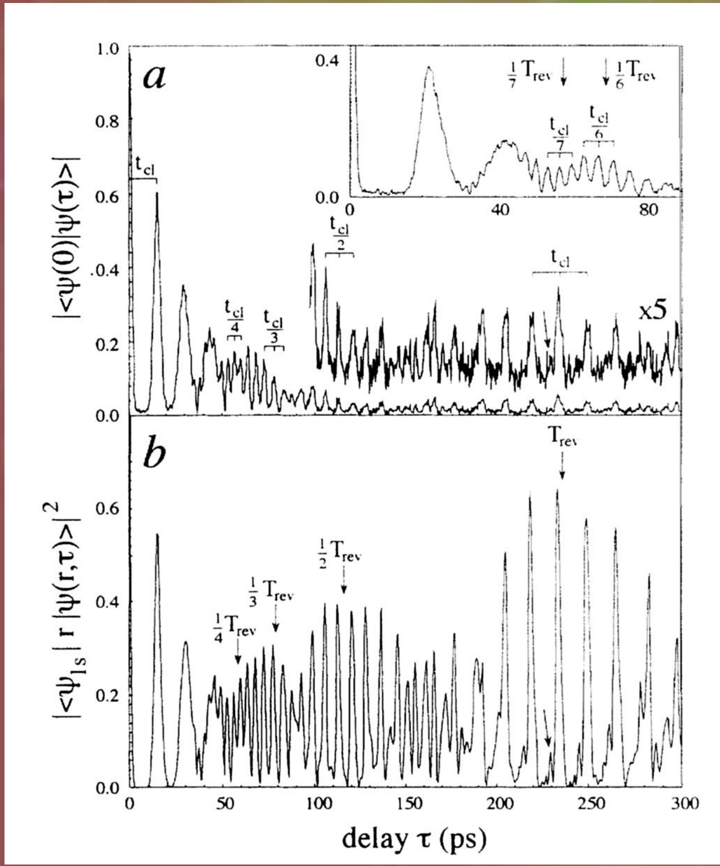
Femtosecond lasers are appropriate for studying nuclear dynamics





Electron dynamics in the ground or excited states of atoms or molecules requires the use attosecond lasers

N.B. Studying electron dynamics with femtosecond lasers requires slowing down the dynamics by working with Rydberg atoms or molecules

Wals. et.al. Phys. Rev. Lett. 72, 3783 (1994).

What do we want?

Fundamental understanding of photo-excitation

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

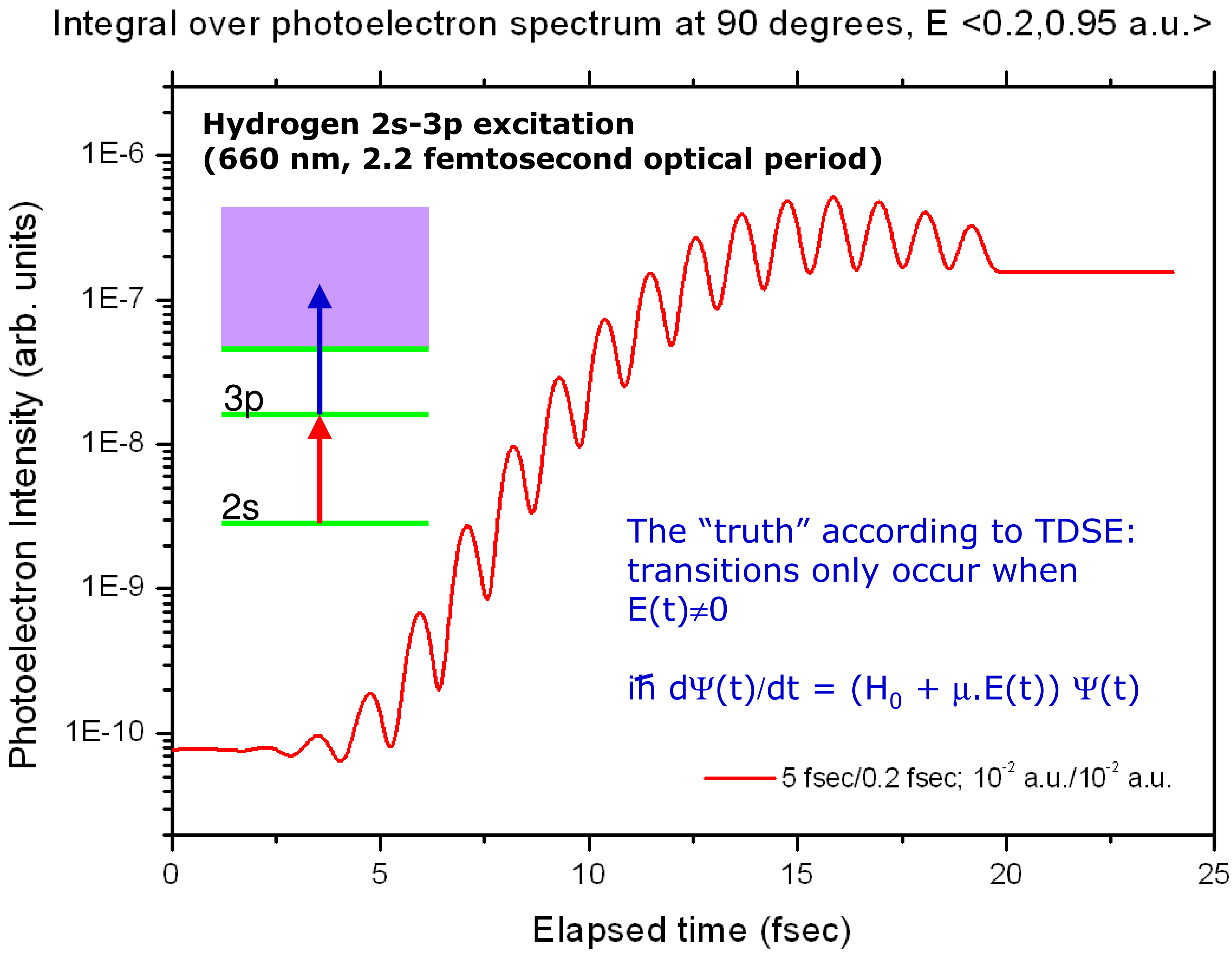
"Textbook" answer: a two-level system subjected to resonant monochromatic radiation will start to perform Rabi oscillations

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

The subcycle dependence in the preexponential factor $N(t)$ can be ignored (up to the electric-field envelope). Indeed, at $\gamma\leqslant 1$ 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:

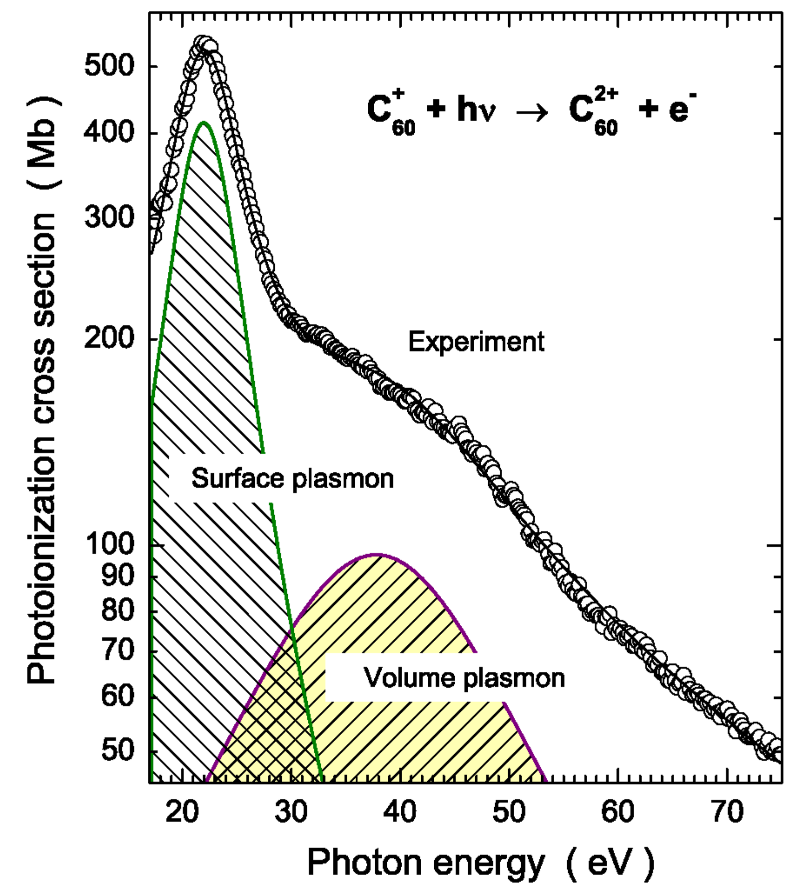
Yudin & Ivanov, Phys. Rev. A 64, 013409 (2001)


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)
3p
2s
The "truth" according to TDSE: transitions only occur when $E(t) \neq 0$
$i\hbar\, d\Psi(t)/dt = (H_0 + \mu.E(t))\, \Psi(t)$
5 fsec/0.2 fsec; 10^{-2} a.u./10^{-2} a.u.
Photoelectron Intensity (arb. units)
1E-6
1E-7
1E-8
1E-9
1E-10
0
5
10
15
20
25
Elapsed time (fsec)

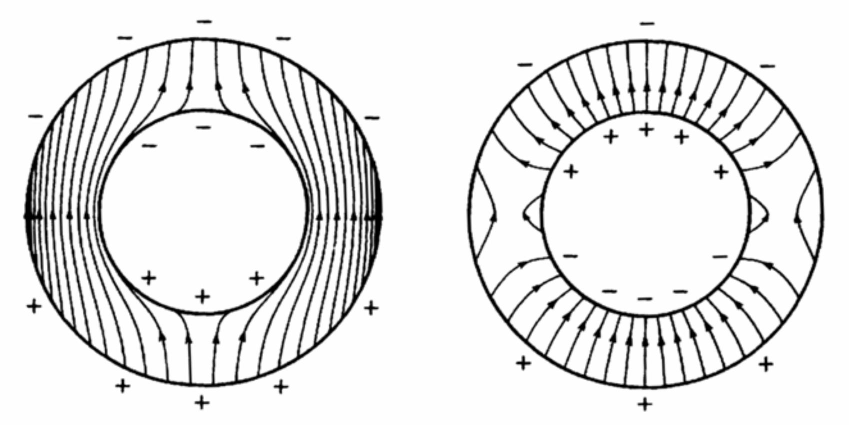
What do we want?

Fundamental understanding of electron correlation

Studying collective electron excitations in C_{60} and nano-particles



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



Surface plasmon (22 eV)

Volume plasmon (38 eV)

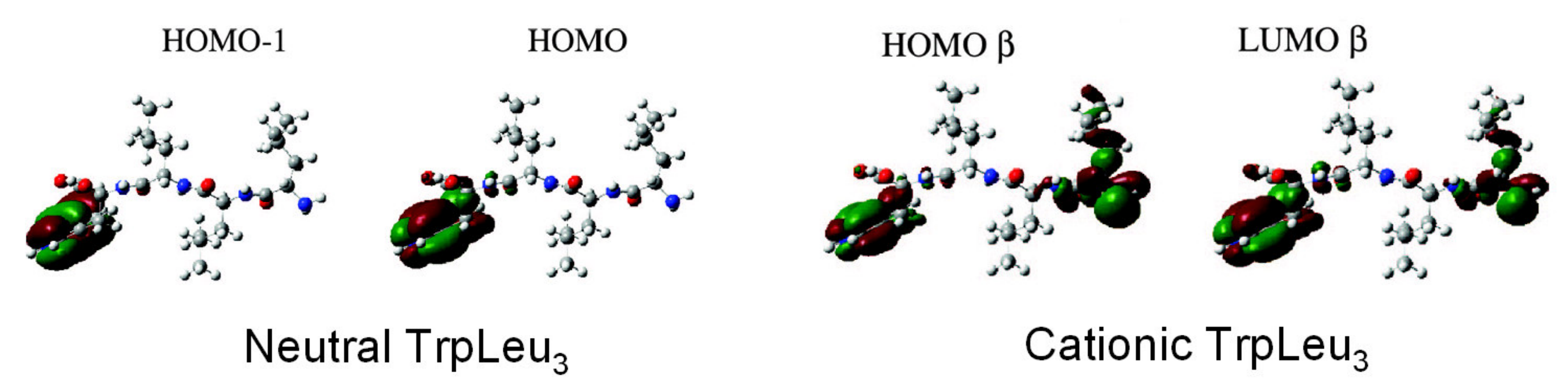
How is the collectivity established?

How is the collectivity lost?

What do we want?

Coupling of electronic and nuclear degrees of freedom

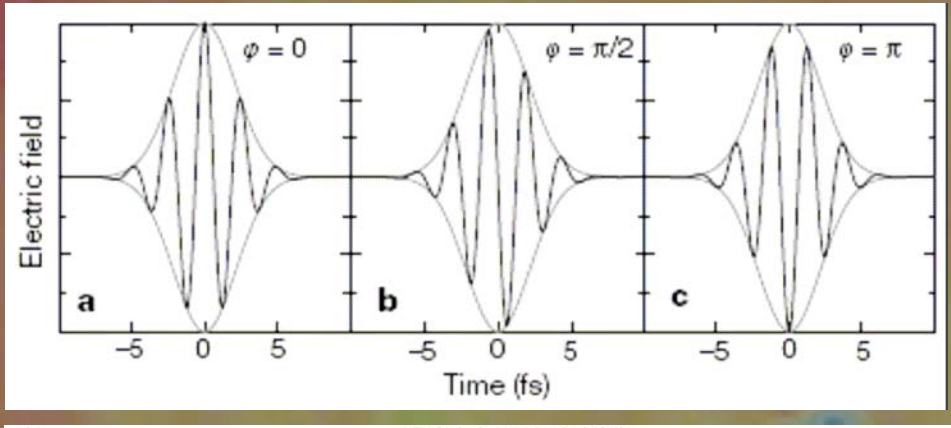
Prediction by F. Remacle and R. Levine (PNAS 103, 6793 (2005)): Ultrafast electron transfer is possible in large bio-molecules.

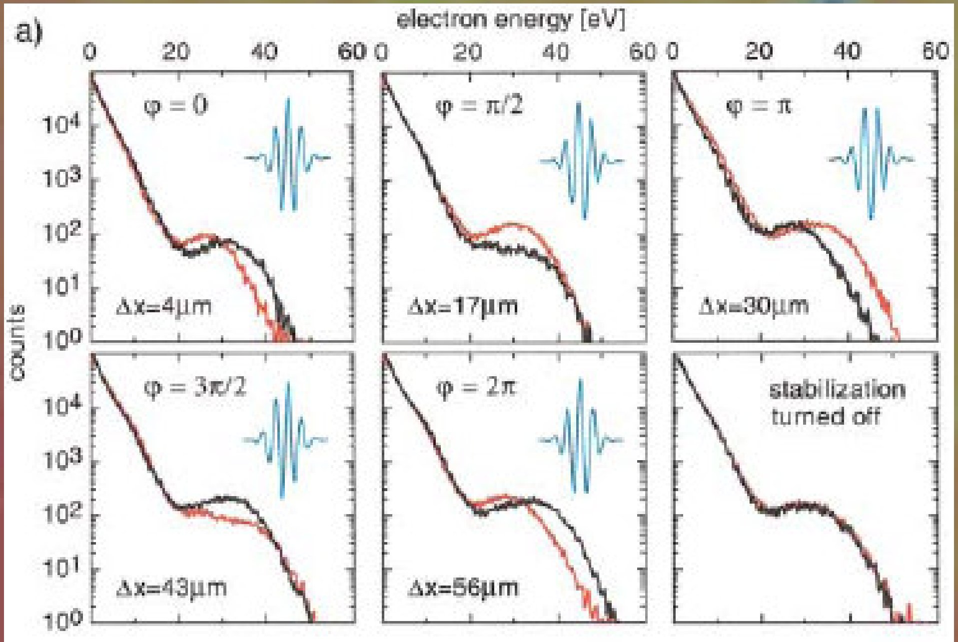


Also: electron dynamics in strong laser fields

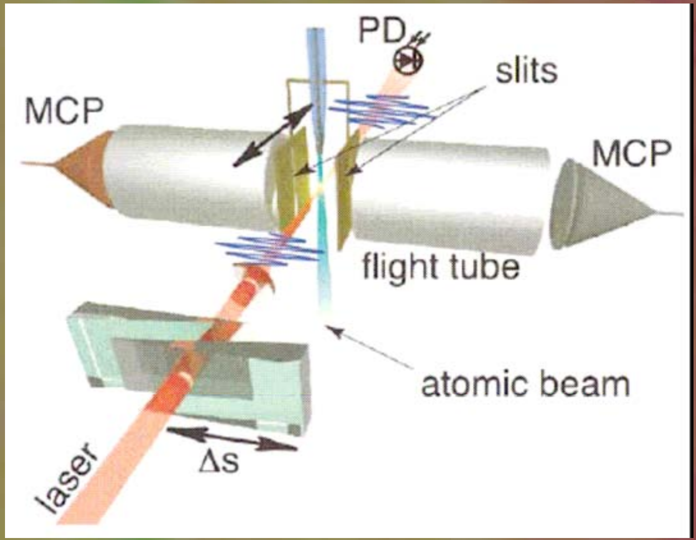
- dynamic alignment
- Coulomb explosion
- control of electron localization

Carrier-Envelope Phase Effects in Atomic Ionization





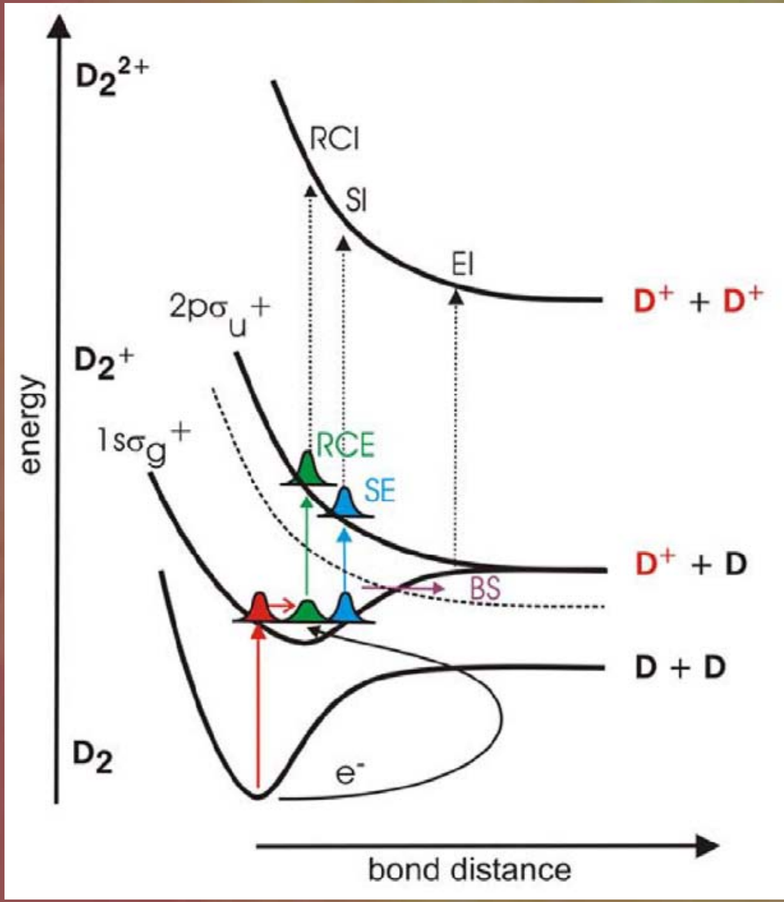
ATI in Xenon

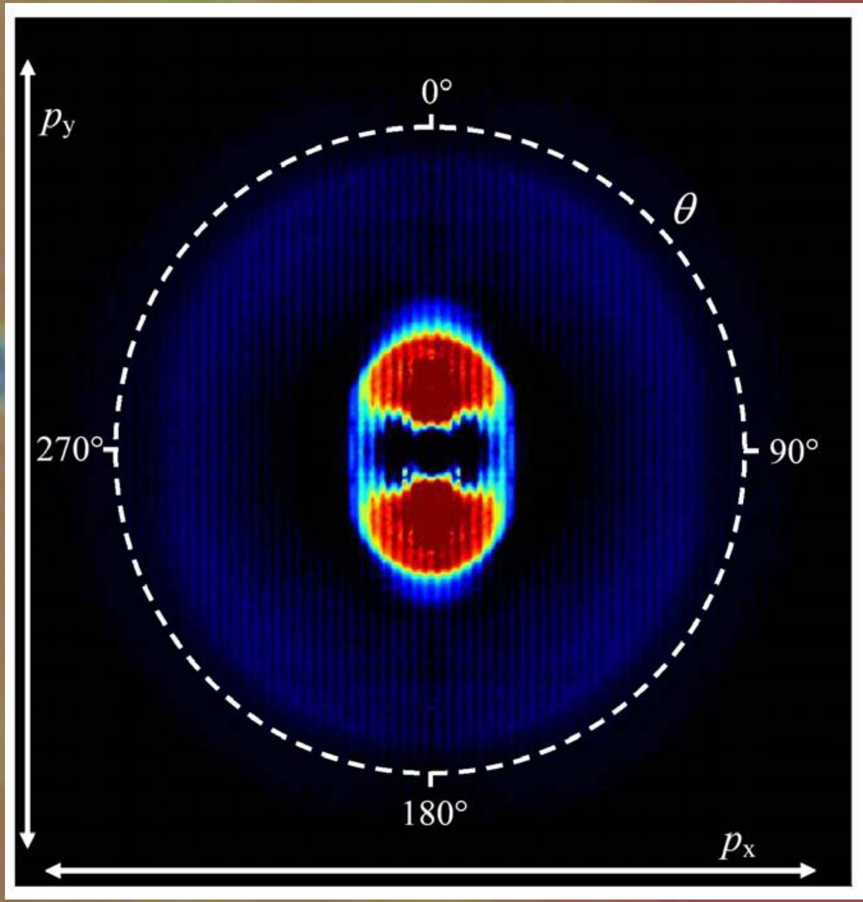


Can carrier envelope phase control electron motion *inside* molecules?

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

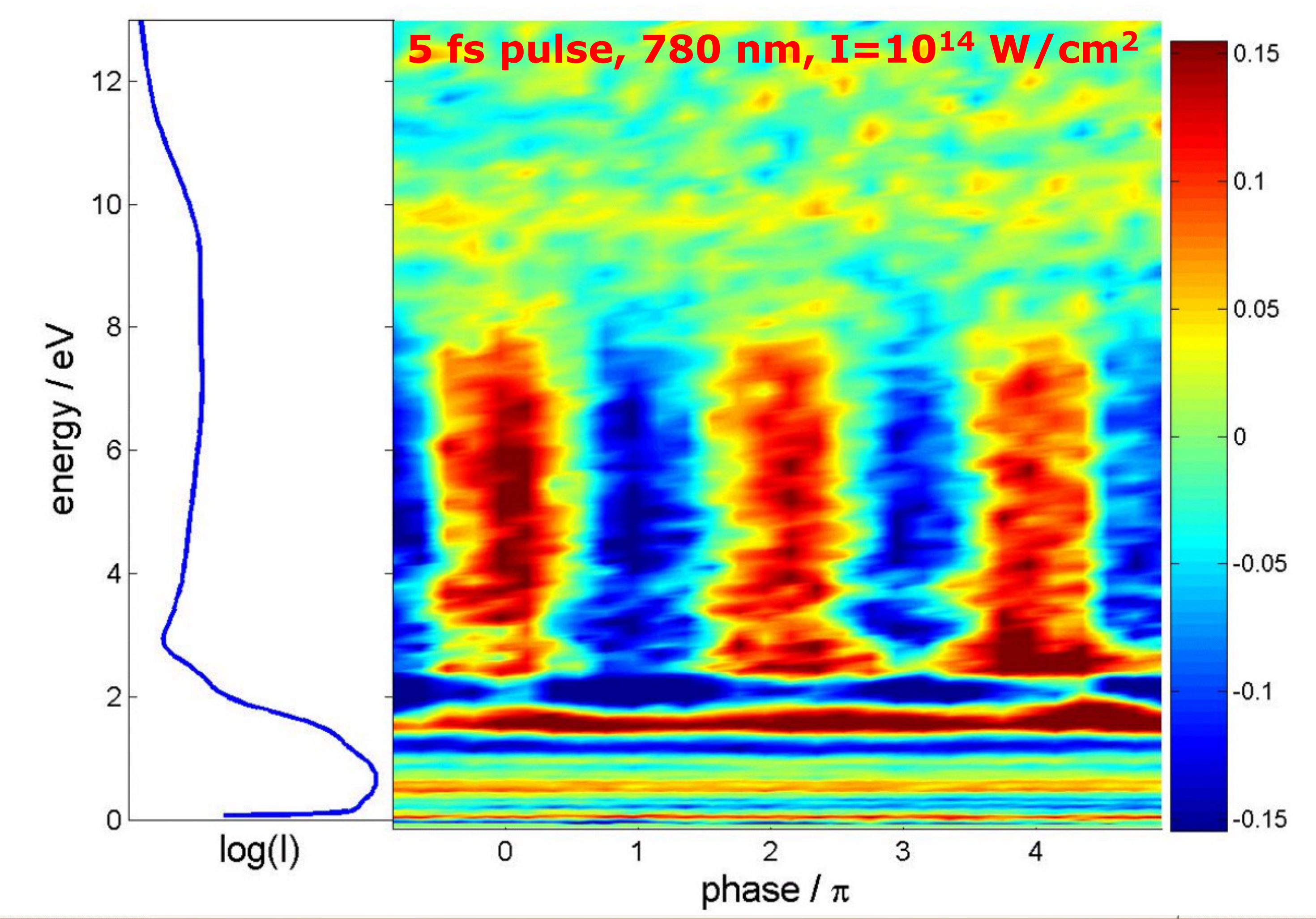
Attosecond electron dynamics in molecules
CEP control of electron localization





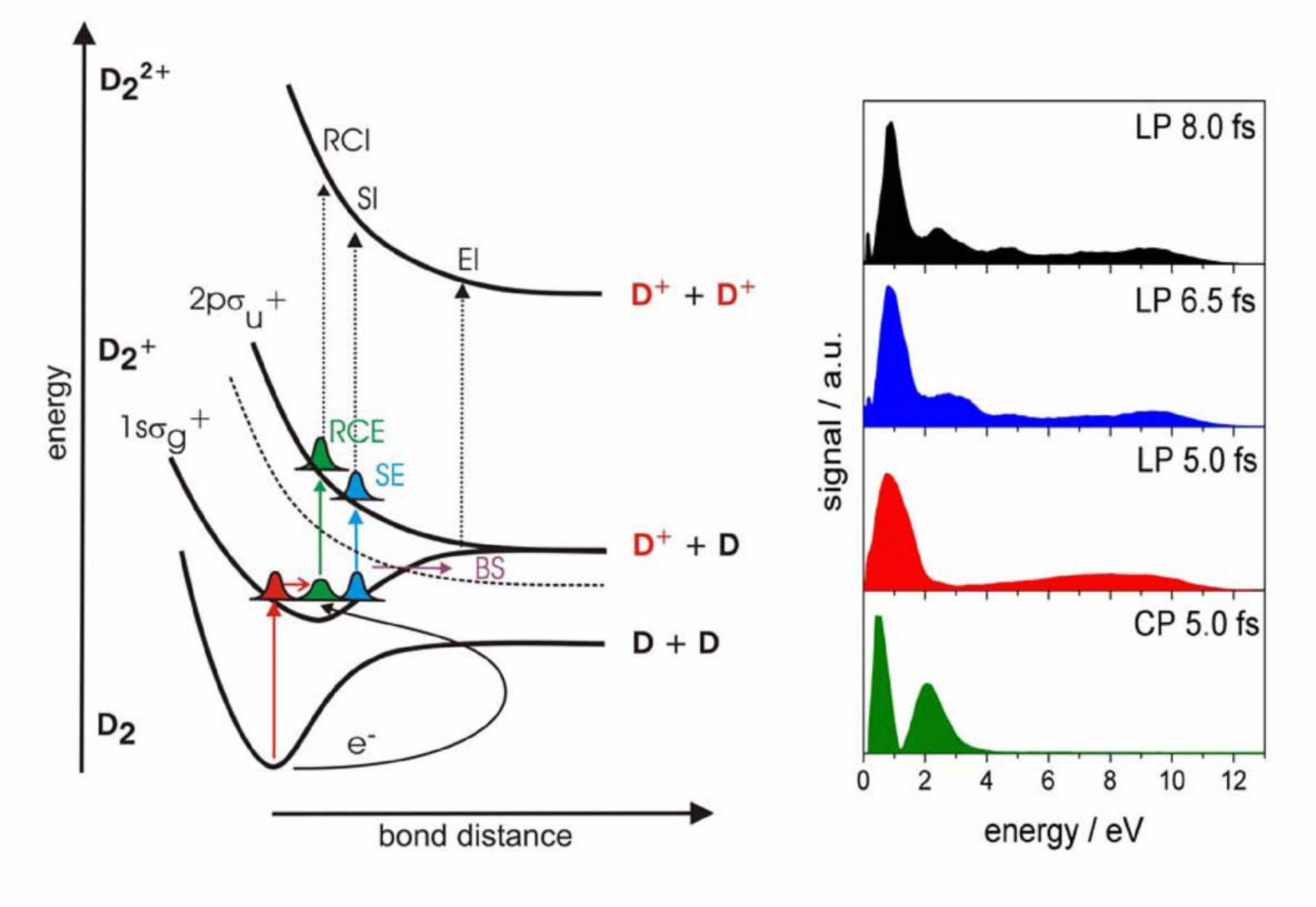
Recent experiment: angle-resolved D^+ ion imaging using CEP-locked few-cycle laser pulses (w. Ferenc Krausz)

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



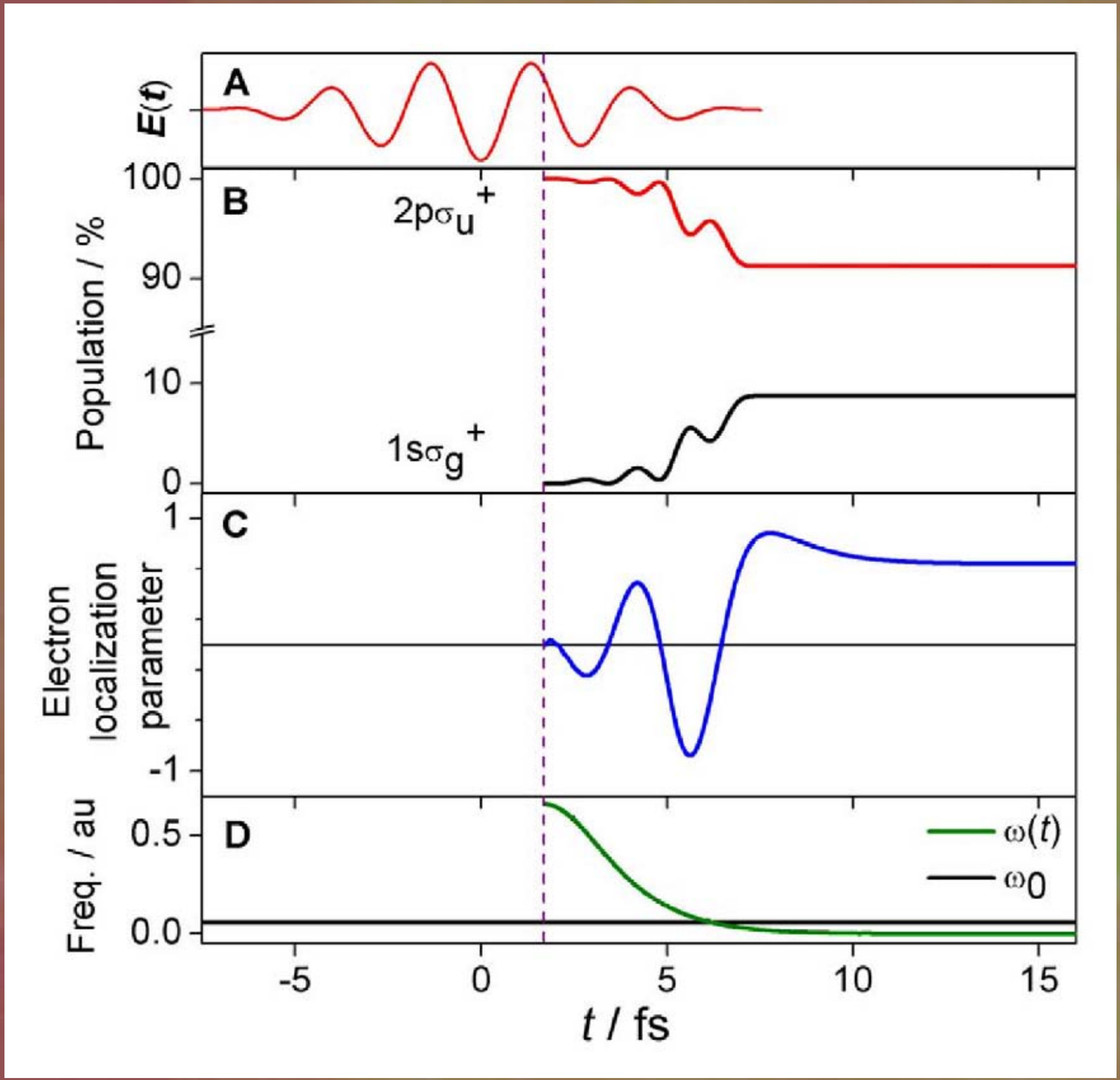
M. Kling et al., Science *312*, 246 (2006)

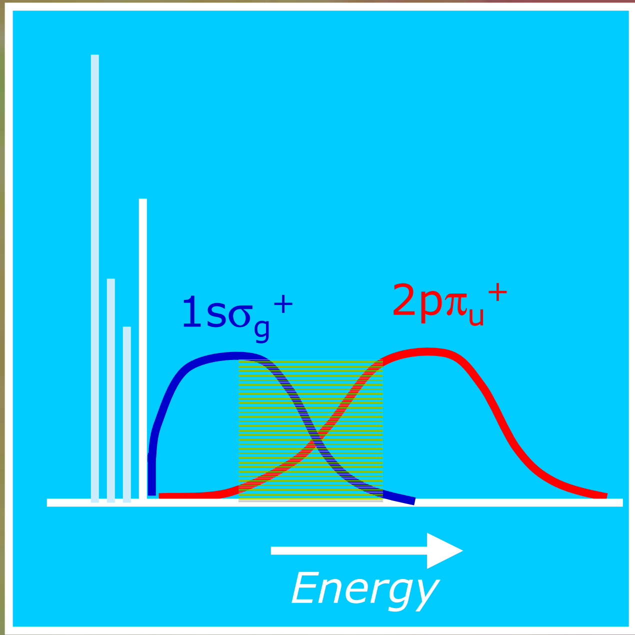
Phase Control Mechanism -1



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

Phase Control Mechanism -2

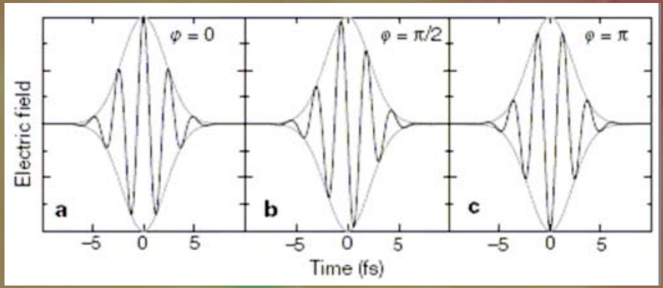


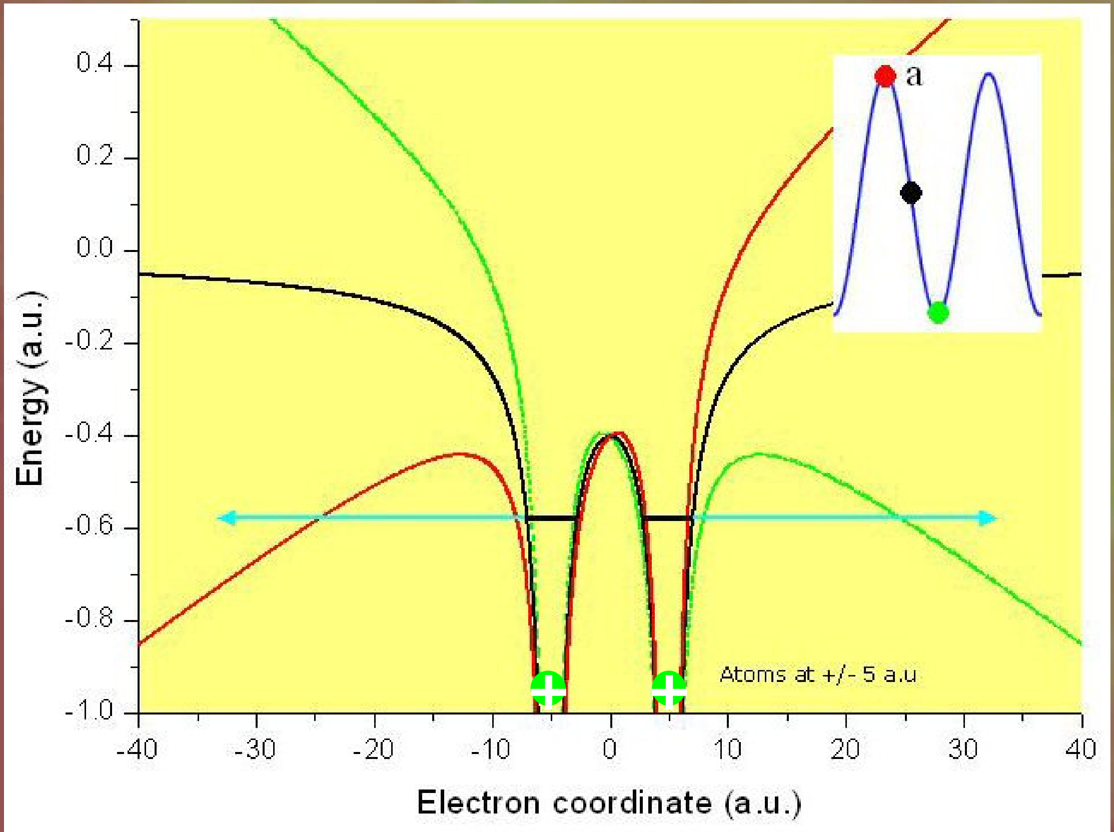


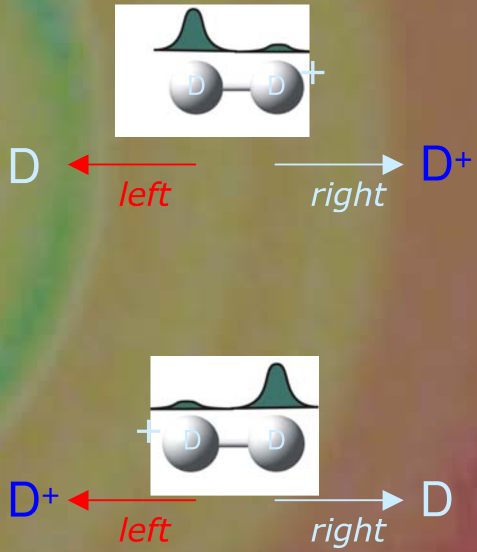
→ Preparation of coherent superposition state by stimulated emission to $1s\sigma_g^+$ state

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

Overview: Attosecond Science from a "user" perspective

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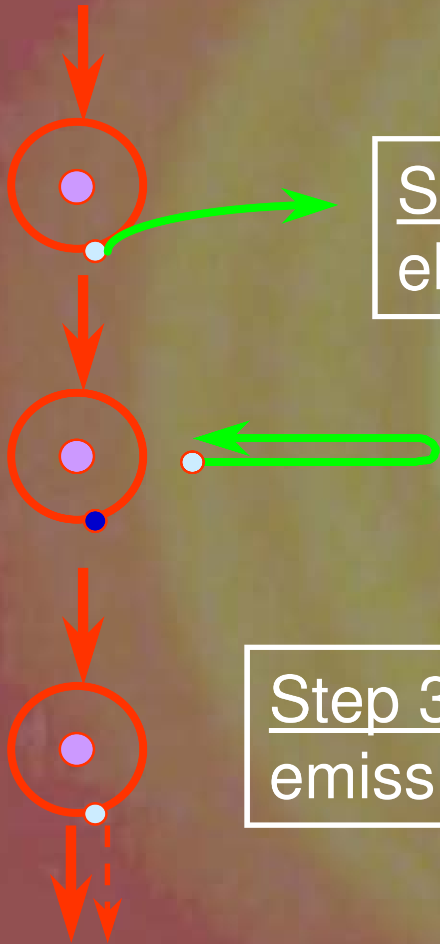
High Harmonic Generation

Intense near-infrared femtosecond laser



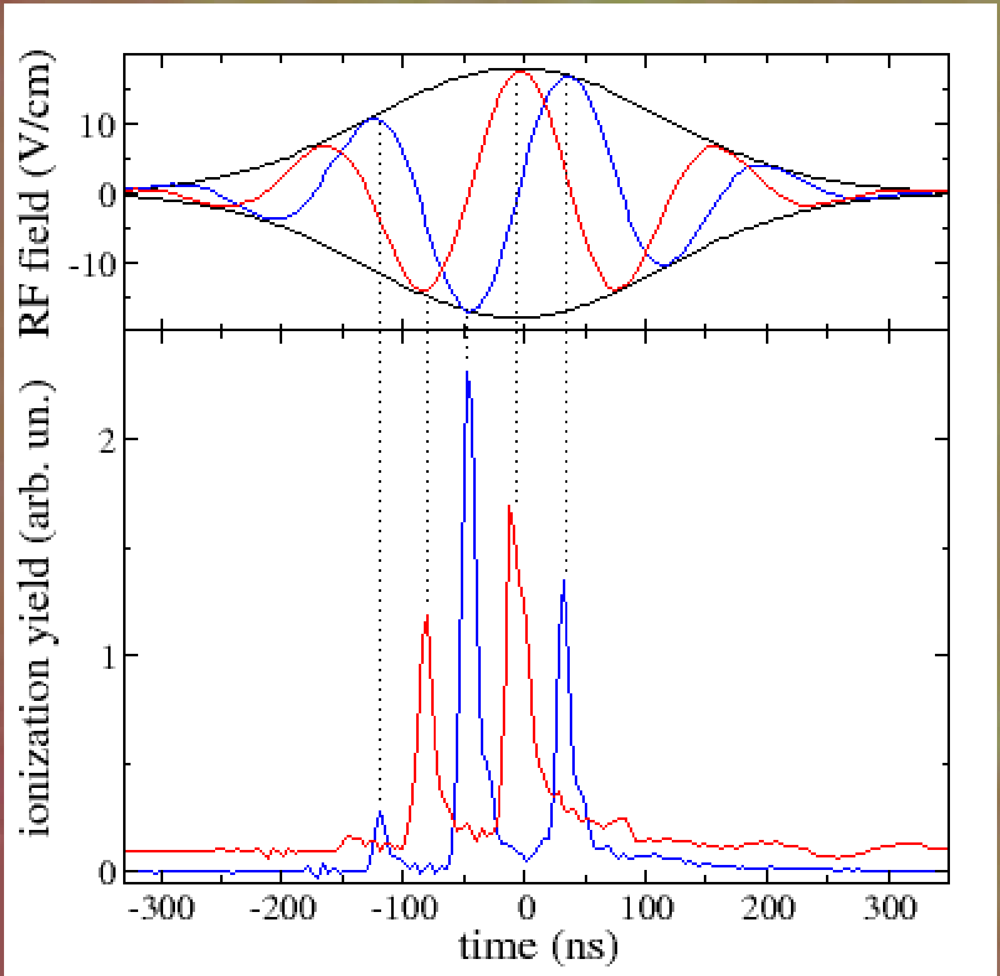
Step 1: ionization and removal of an electron from the positive ion core

Step 2: acceleration of the electron in the oscillatory laser field

Step 3: recombination, accompanied by the emission of an XUV photon

Intense near-infrared femtosecond laser + XUV radiation
(repetitive nature gives odd harmonics)

Dependence of few-cycle photoionization on the carrier envelope phase



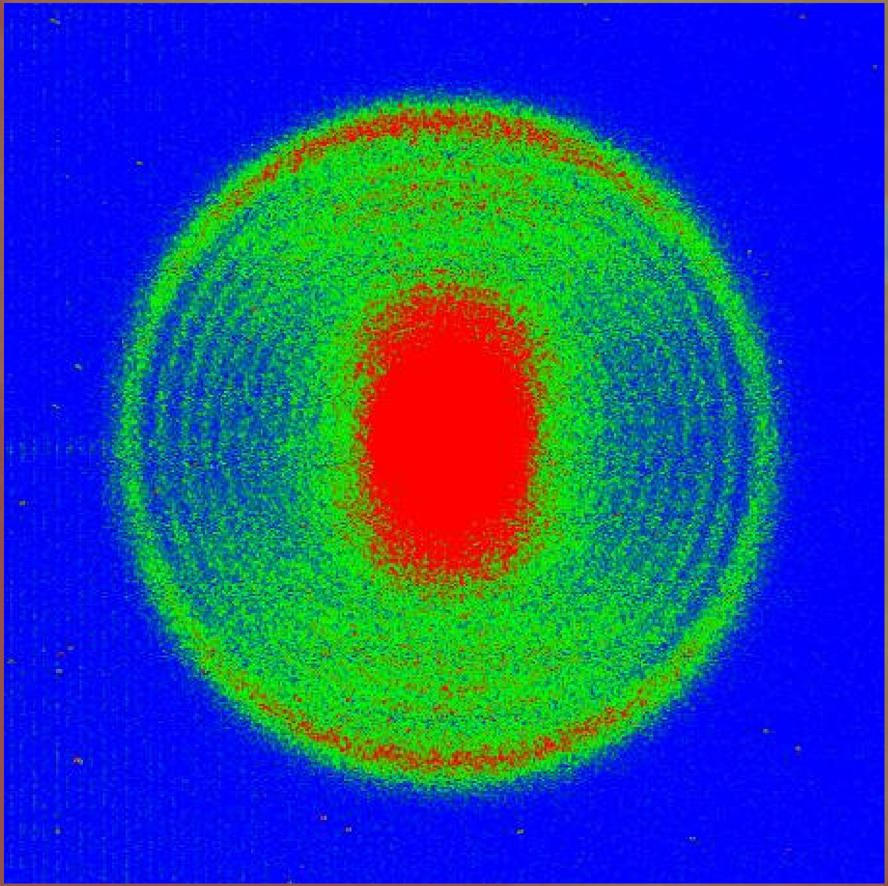
Temporal emission measured with sub-cycle resolution

$\Rightarrow$ emission with sub-cycle time dependence and strongly depending on carrier phase

Guertler et.al., Phys. Rev. Lett. 92, 063901 (2004)

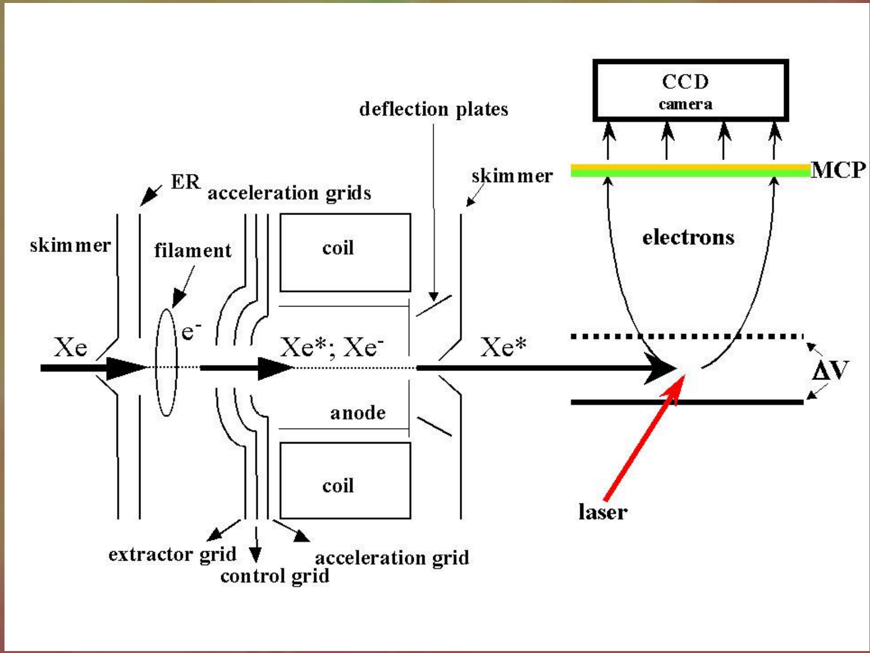
The role of electron trajectories in photoionization - 1

Experiment: Ionization of Xe metastable atoms with a small excess kinetic energy, with a nanosecond laser



Nicole et.al., Phys. Rev. Lett. 88, 133001 (2002)

Narrowband excitation near the field-free ionisation limit (above the saddle-point in the combined DC field + Coulomb potential) → mono-energetic photoelectrons



High Harmonic Generation

Intense near-infrared femtosecond laser

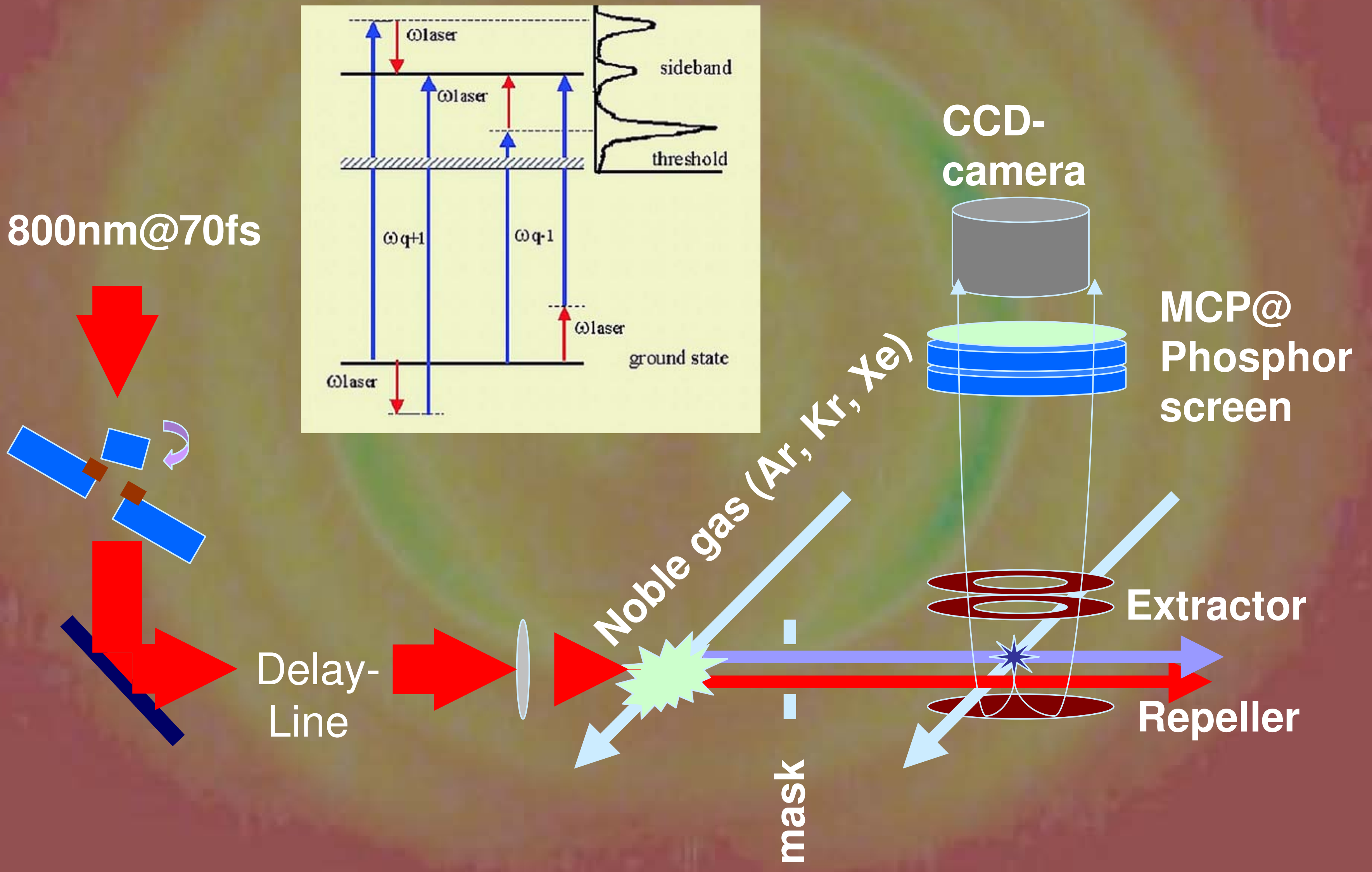
<u>Step 1</u>: Ionization near maximum of amplitude of laser electric field

<u>Step 2</u>: The electron follows a well-defined trajectory in the field

<u>Step 3</u>: The electron returns and recombines during a very finite part of the IR cycle

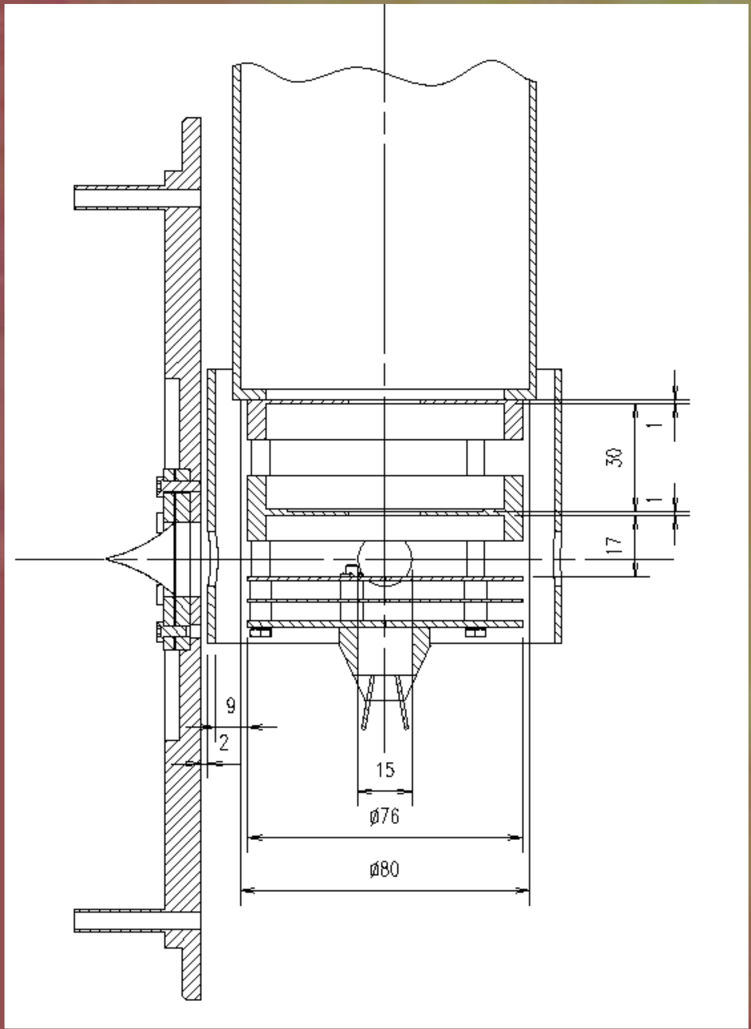
Intense near-infrared femtosecond laser + XUV radiation

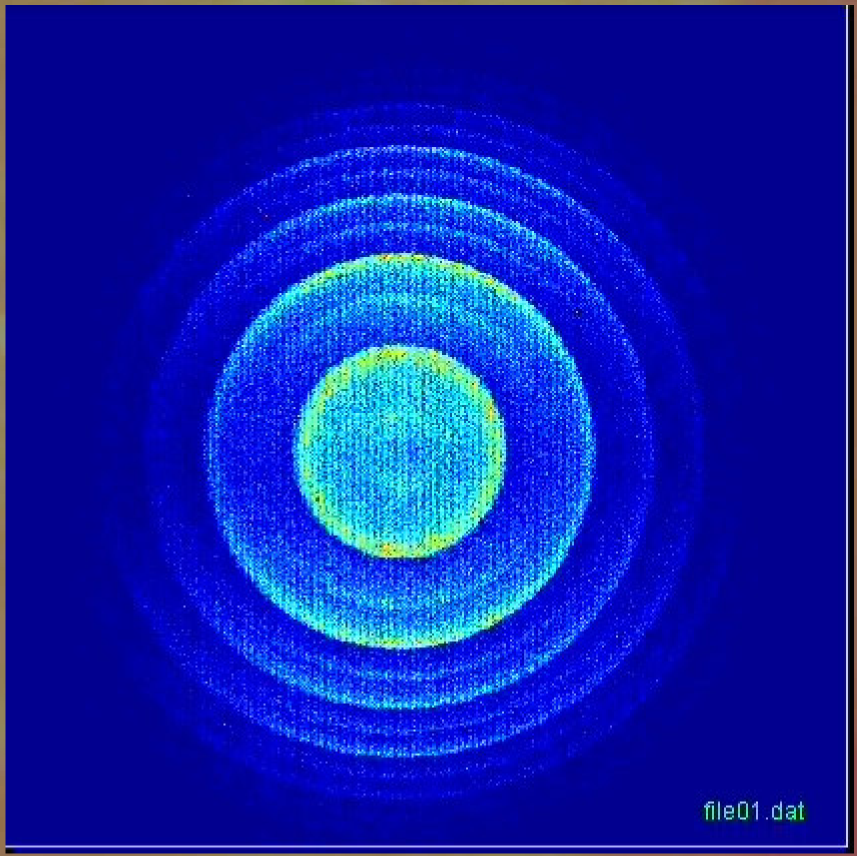
Characterization of attosecond laser pulses:
XUV + weak IR field (10^{11} W/cm^2)



Raw Photoelectron Images vs Time-delay

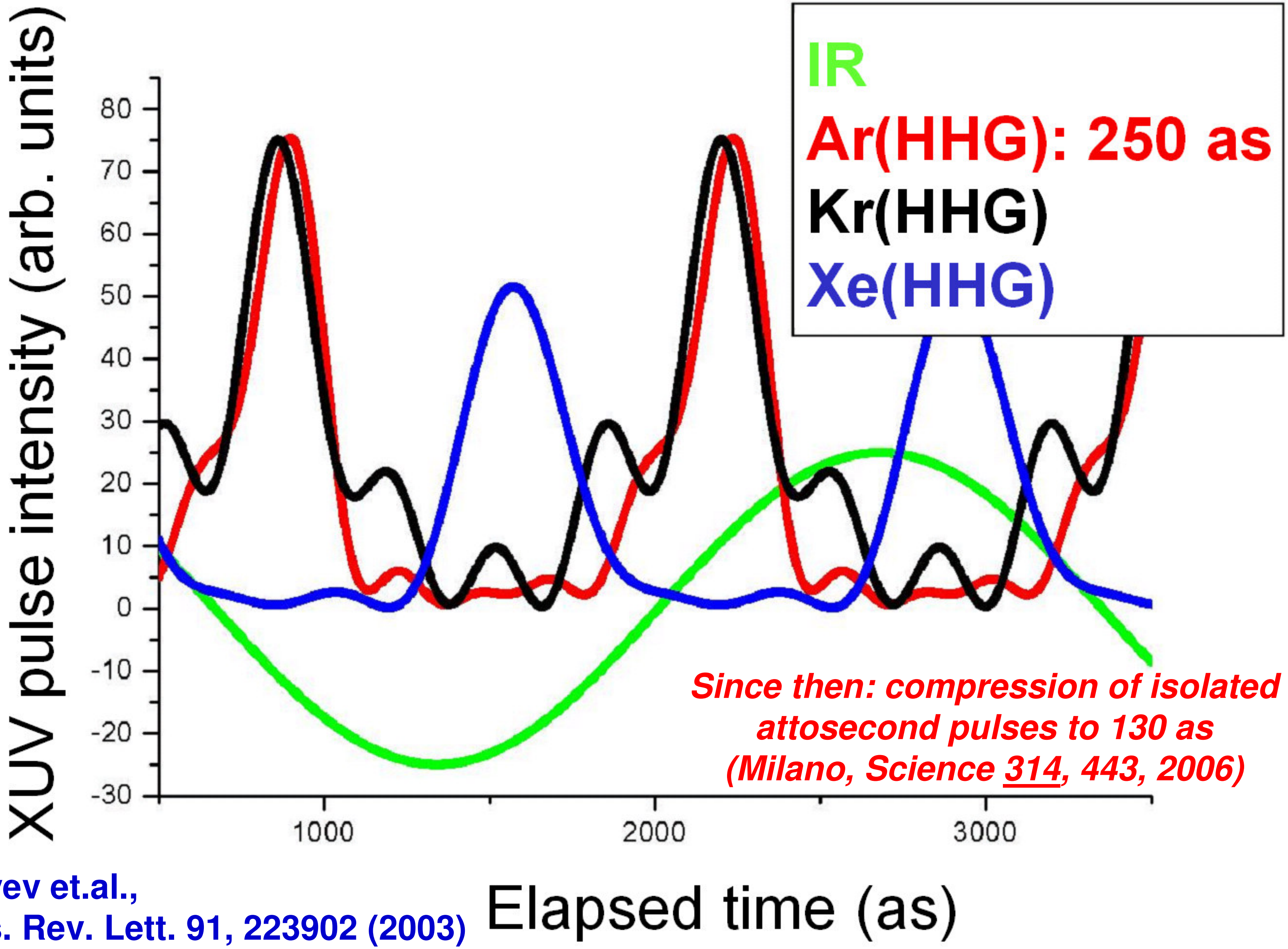
Velocity map imaging





Aseyev et.al., Phys. Rev. Lett. 91, 223902 (2003)

Reconstructed attosecond pulses



Since then: compression of isolated attosecond pulses to 130 as (Milano, Science 314, 443, 2006)

Aseyev et.al.,
Phys. Rev. Lett. 91, 223902 (2003)

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Two Families of Attosecond Laser Experiments

1. 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

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

   Good for interferometry experiments

Two ways that we can sofar use these attosecond pulses in experiments

1. XUV ionization followed by acceleration of the ionized electron in a strong IR field (continuum)

   Used in attosecond pulse characterization

   Used in attosecond interferometry experiments

2. XUV excitation of bound states, followed by ionization in a strong IR field

   Used to study bound state dynamics and/or time-dependent ionization dynamics

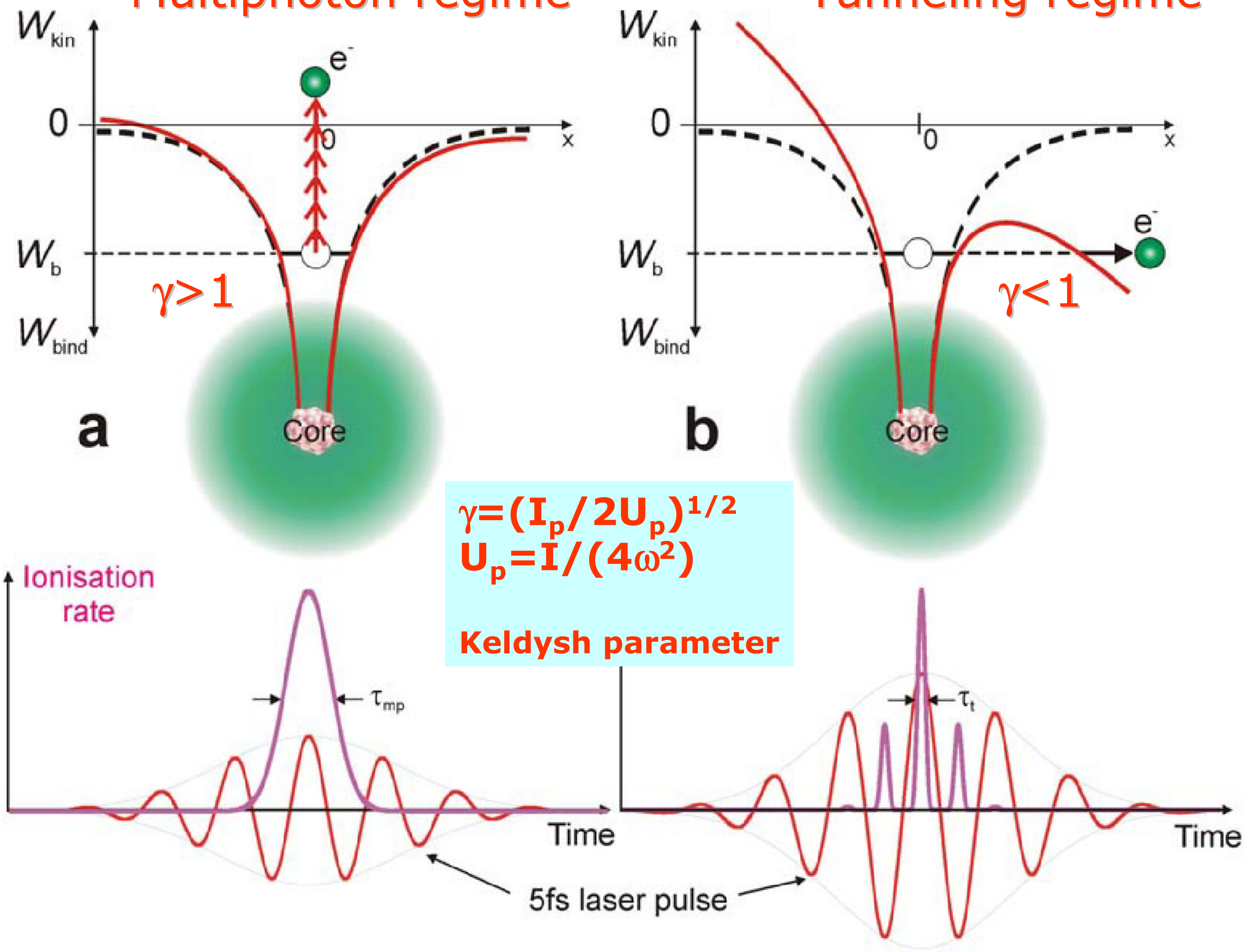
Example 1:

Photoabsorption on attosecond timescales

Pump-probe experiment
→ isolated attosecond pulses

Bound state dynamics
→ time-dependent strong-field ionization

Multiphoton regime   Tunneling regime

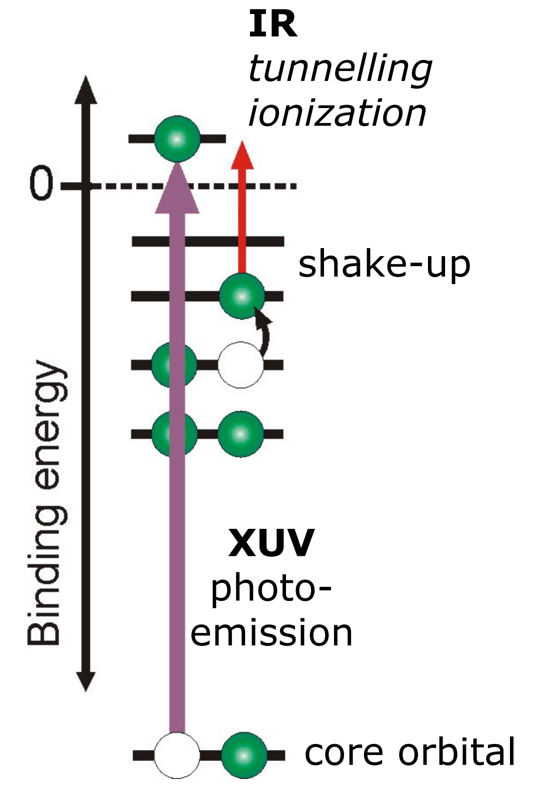


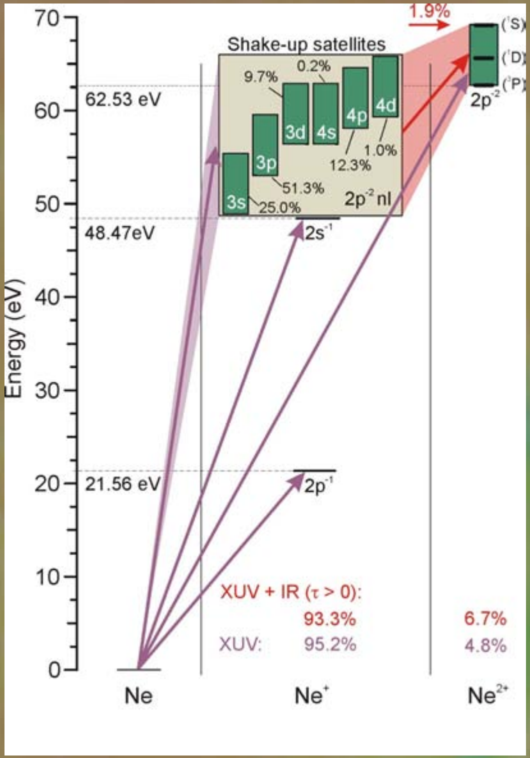
$\gamma=(I_p/2U_p)^{1/2}$
$U_p=I/(4\omega^2)$

Keldysh parameter

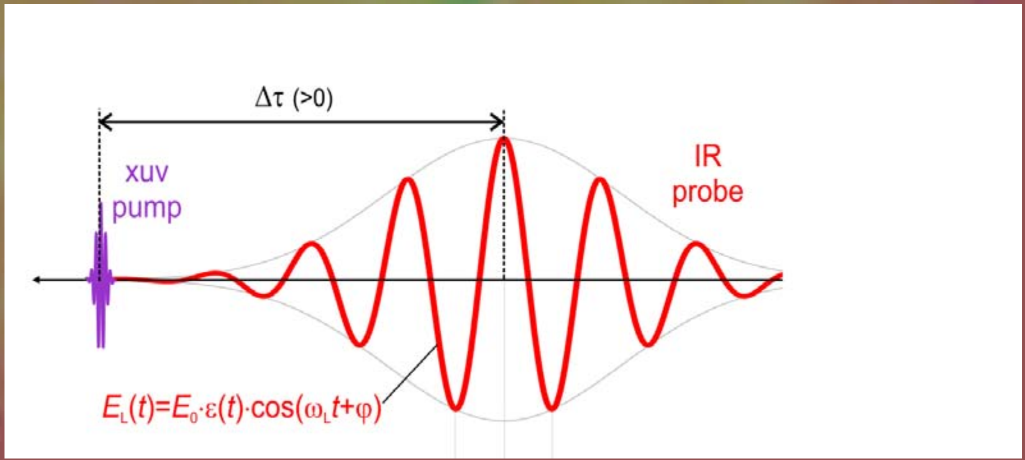
Time-resolving inner-atomic processes by tunnelling

Ne double ionization
(XUV+IR)

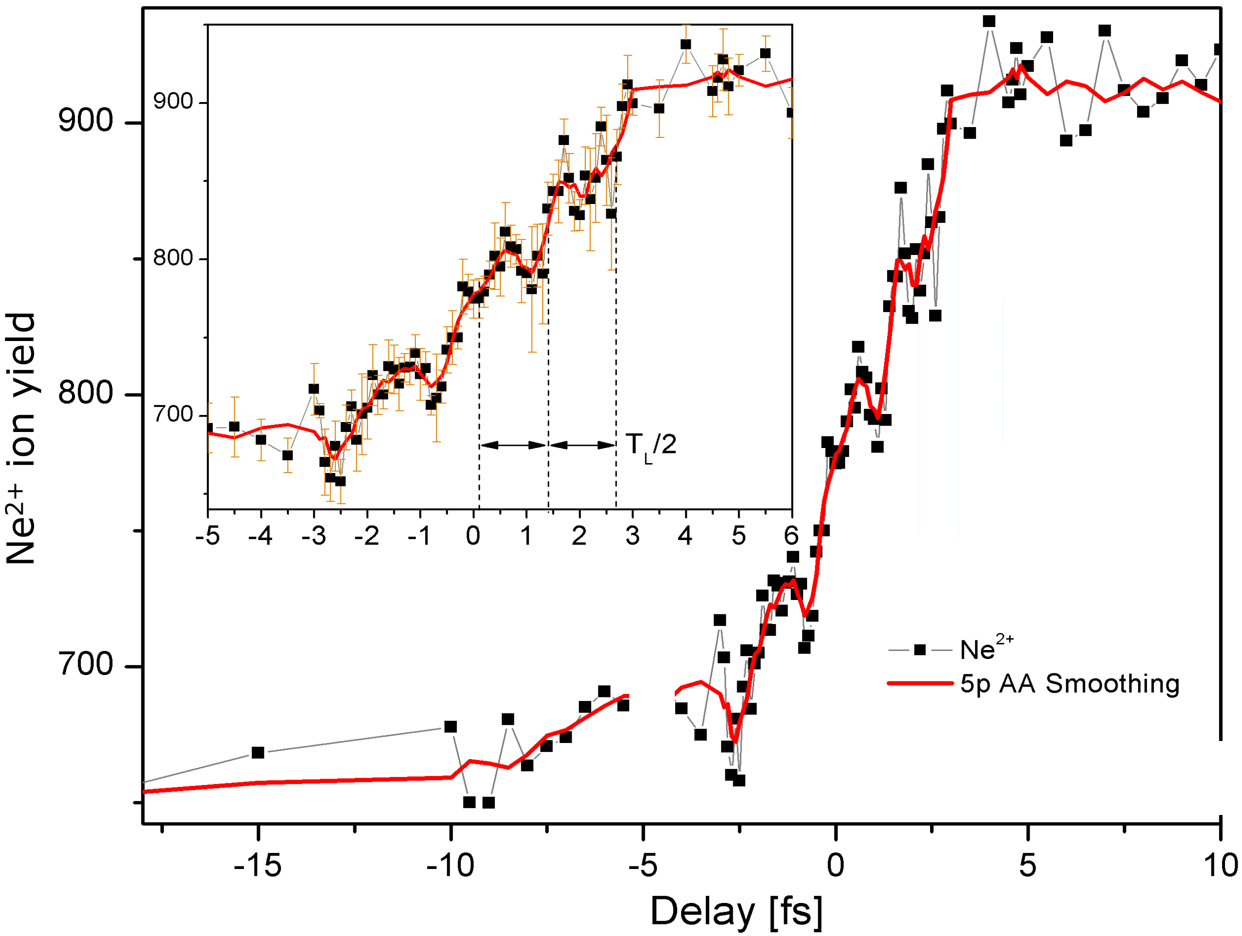




=> shake-up + tunnelling ionization probe < 400 as



Time-resolving inner-atomic processes by tunneling

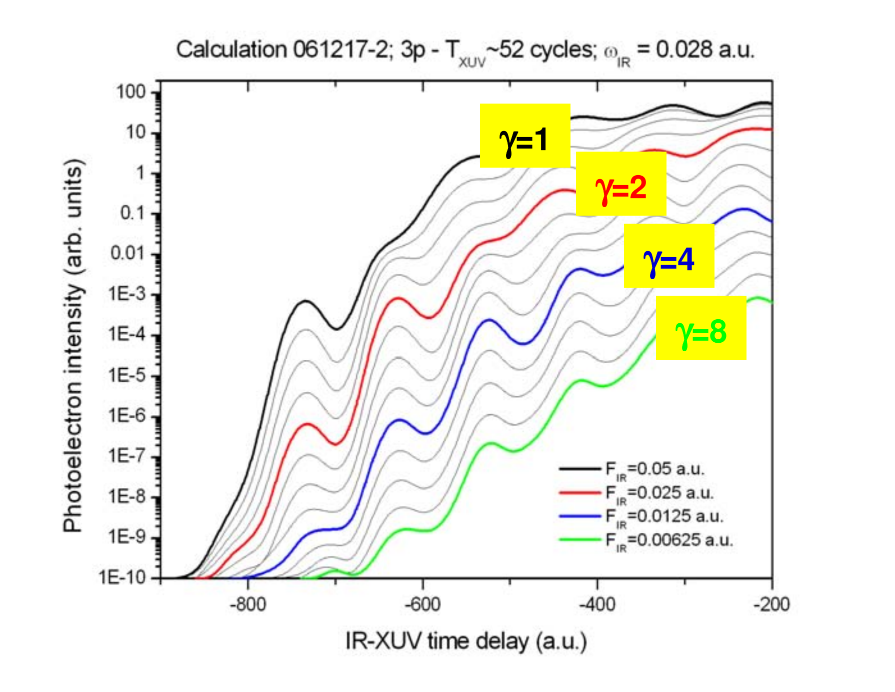


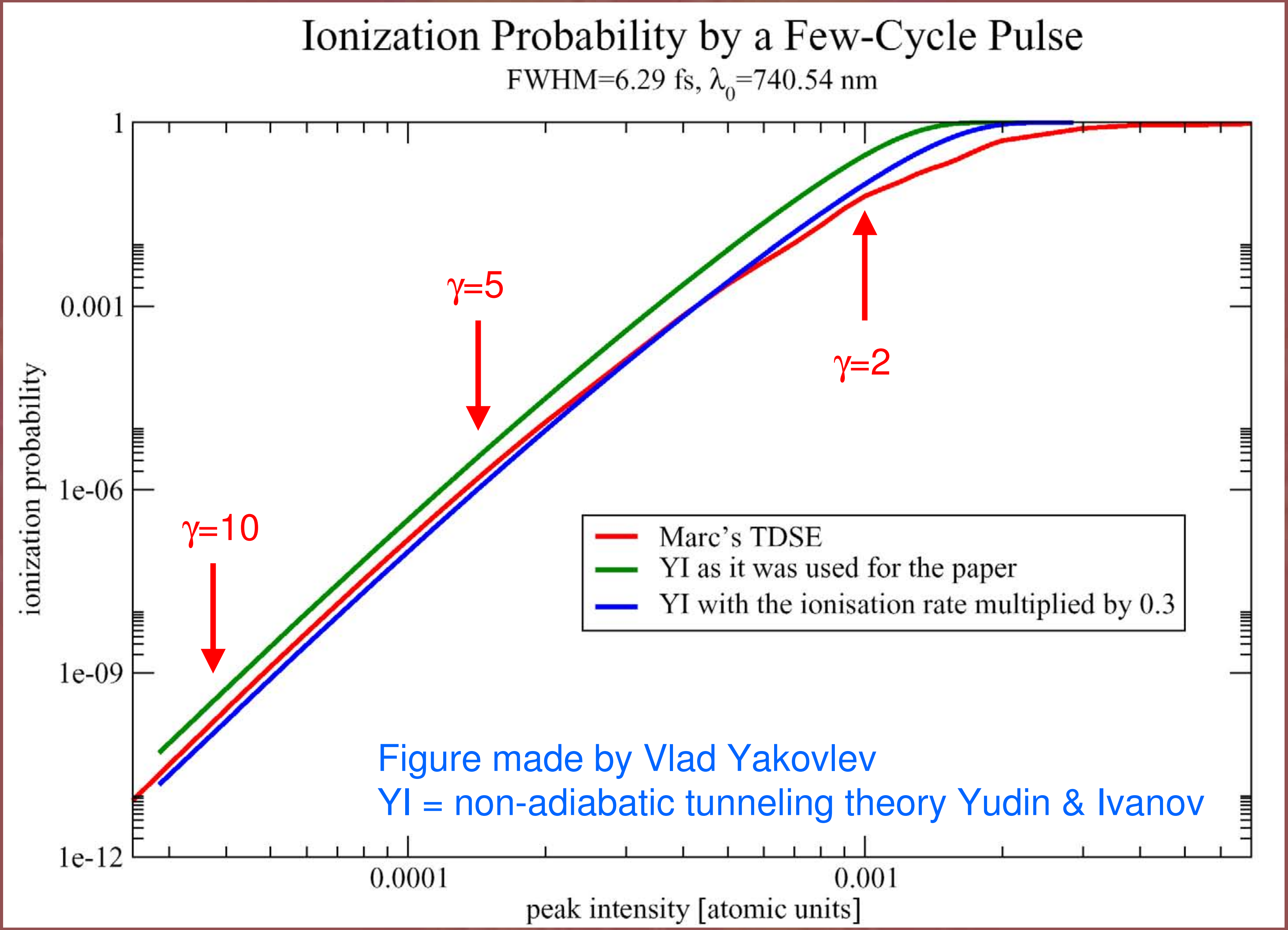
M Uiberacker et al., Nature 446, 627 (2007)

Changing the way people think about ionization … is the ‘conventional wisdom’ correct?

- Conventional wisdom: at low intensity ionization follows the envelope of the pulse, at high intensity it follows the field

- TDSE simulation of Ne^+ ionization at variable field strengths → the sub-cycle time dependence remains deep into the multi-photon regime!!!





Ionization Probability by a Few-Cycle Pulse
FWHM=6.29 fs, λ_0=740.54 nm
ionization probability
1
0.001
1e-06
1e-09
1e-12
0.0001
0.001
peak intensity [atomic units]
γ=10
γ=5
γ=2
Marc's TDSE
YI as it was used for the paper
YI with the ionisation rate multiplied by 0.3
Figure made by Vlad Yakovlev
YI = non-adiabatic tunneling theory Yudin & Ivanov

Example 2:

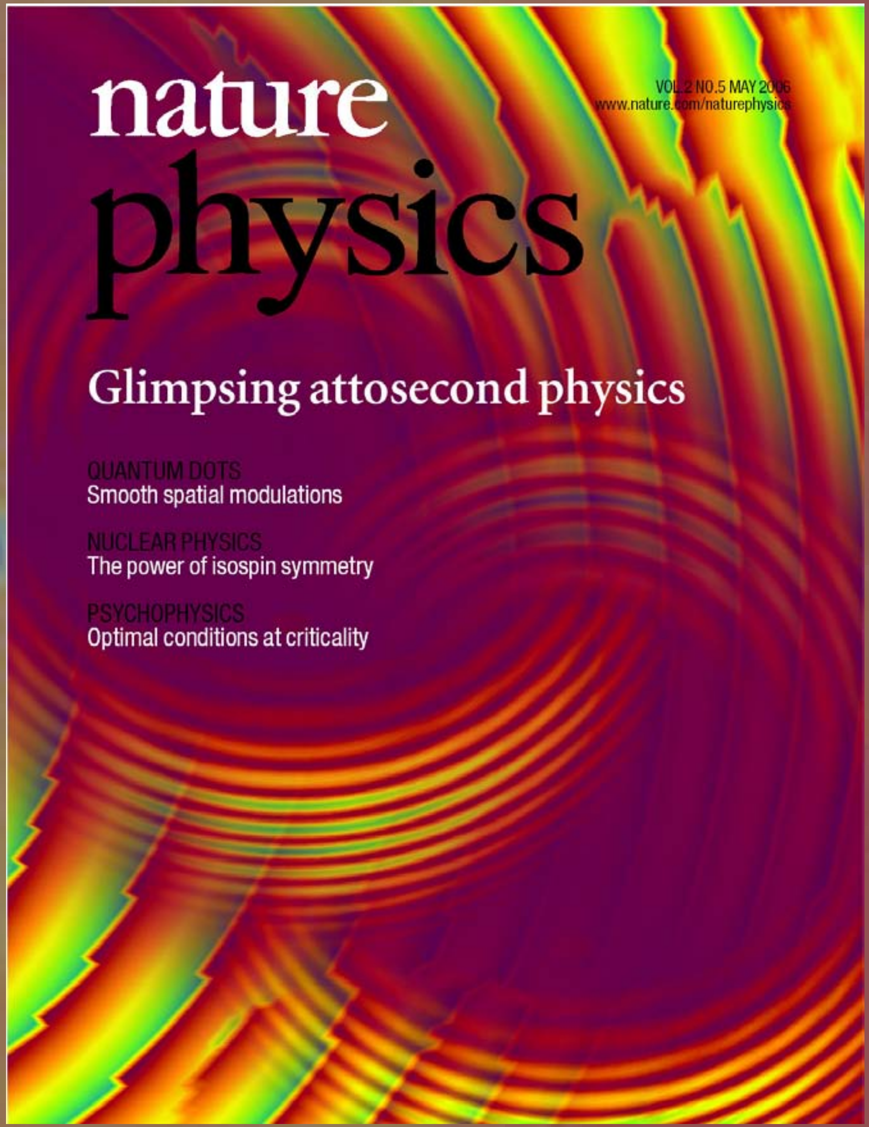
Attosecond electron wavepacket interferometry

Interferometry experiment
→ train of attosecond pulses

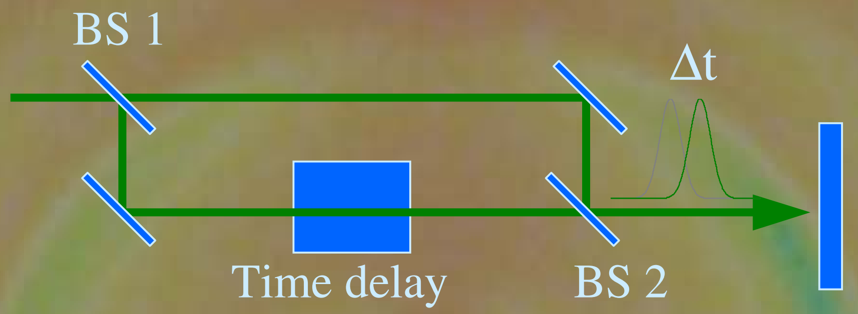
Continuum dynamics
→ acceleration of electrons by strong laser field

Double-slit interference of photon or electron waves is a phenomenon that "*is impossible, <u>absolutely</u> impossible, to explain in any classical way, and which has in it the heart of quantum mechanics. In reality, it contains the only mystery*".

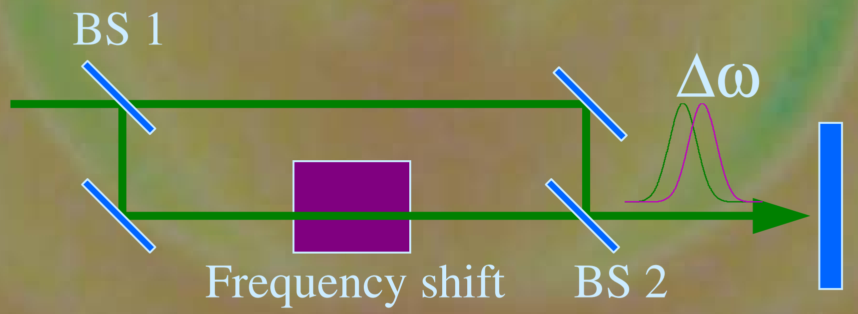
(Richard Feynman, "Feynman Lectures on Physics, vol. 3)



From optical interferometry…

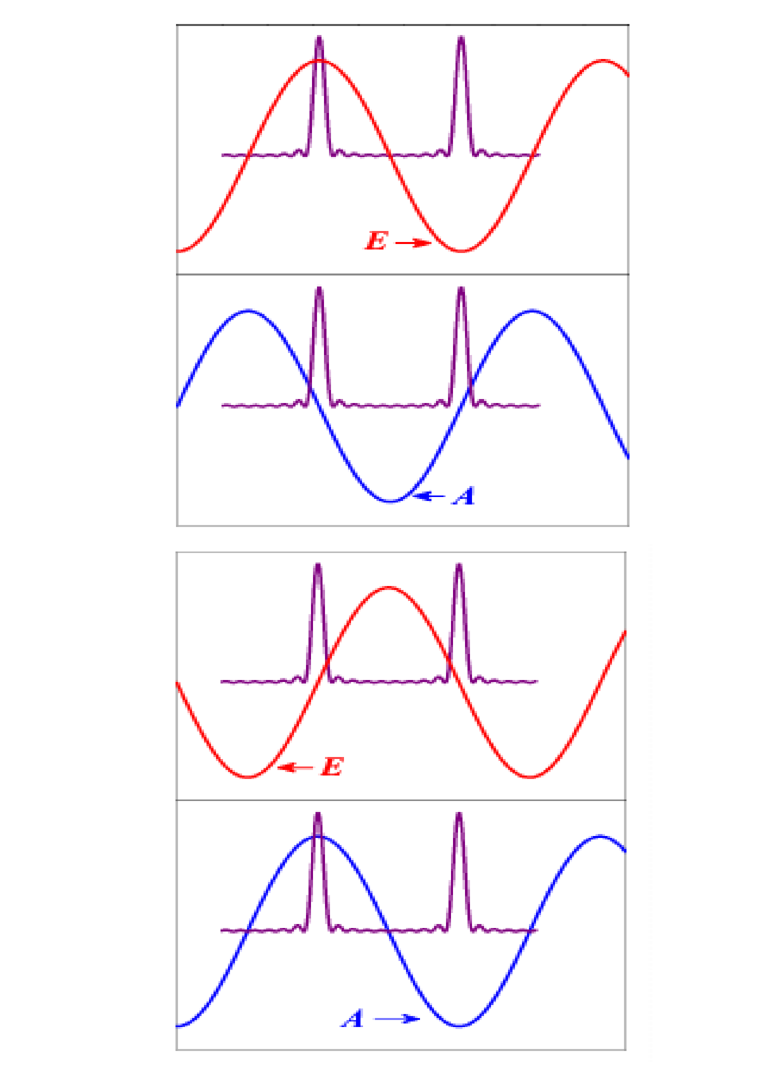


$$|\,E(t)+E(t+\Delta t)e^{i\phi(\Delta t)}\,|^2 \propto 1+\cos\phi(\Delta t)$$



$$|\,E(\omega)+E(\omega+\Delta\omega)\,|^2 \propto 1+\cos\left[\phi(\omega)-\phi(\omega+\Delta\omega)\right]$$

… to attosecond electron wavepacket interferometry

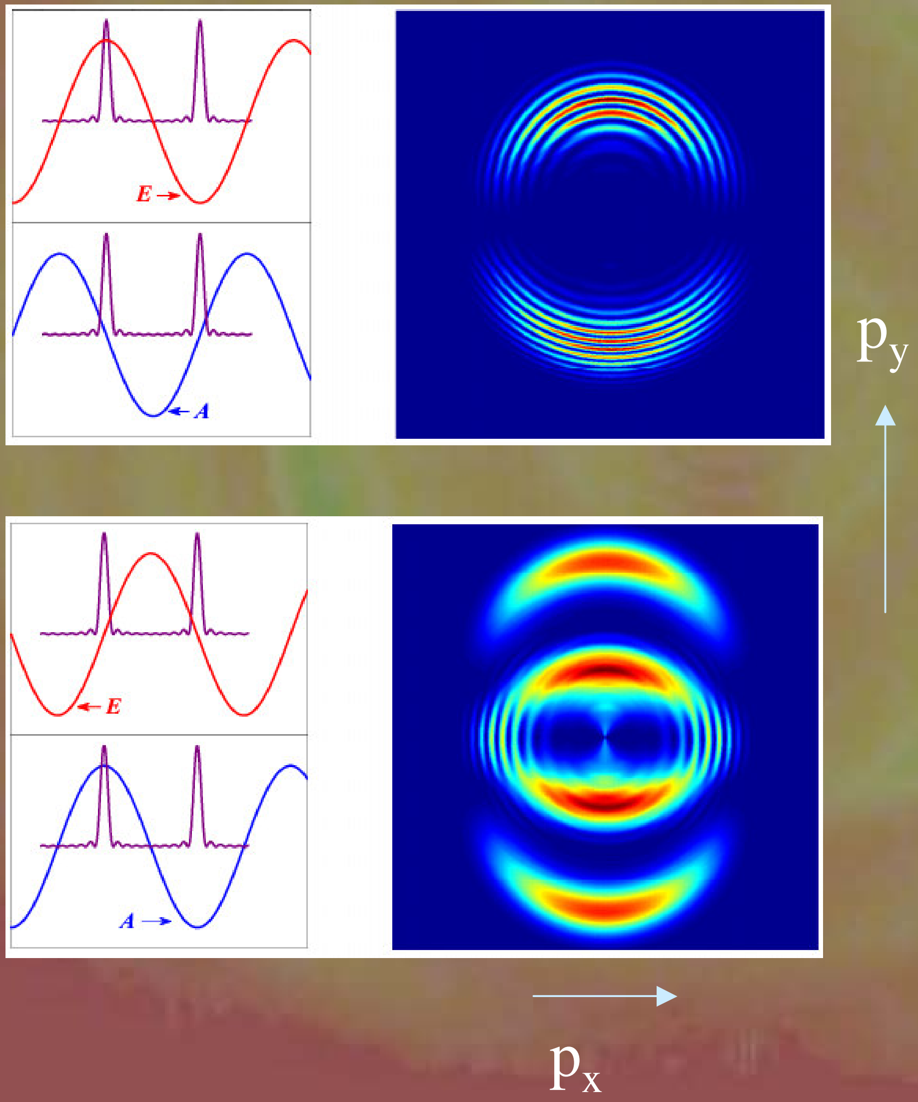


Analogy between optical fields and electronic wavefunctions

- ❖ Electron wavepackets are prepared with attosecond light pulses (replication)
- ❖ Wavepackets acceleration by a strong IR laser field (phase and/or frequency modulation)

$A(t) \sim \int E(t)\, dt$

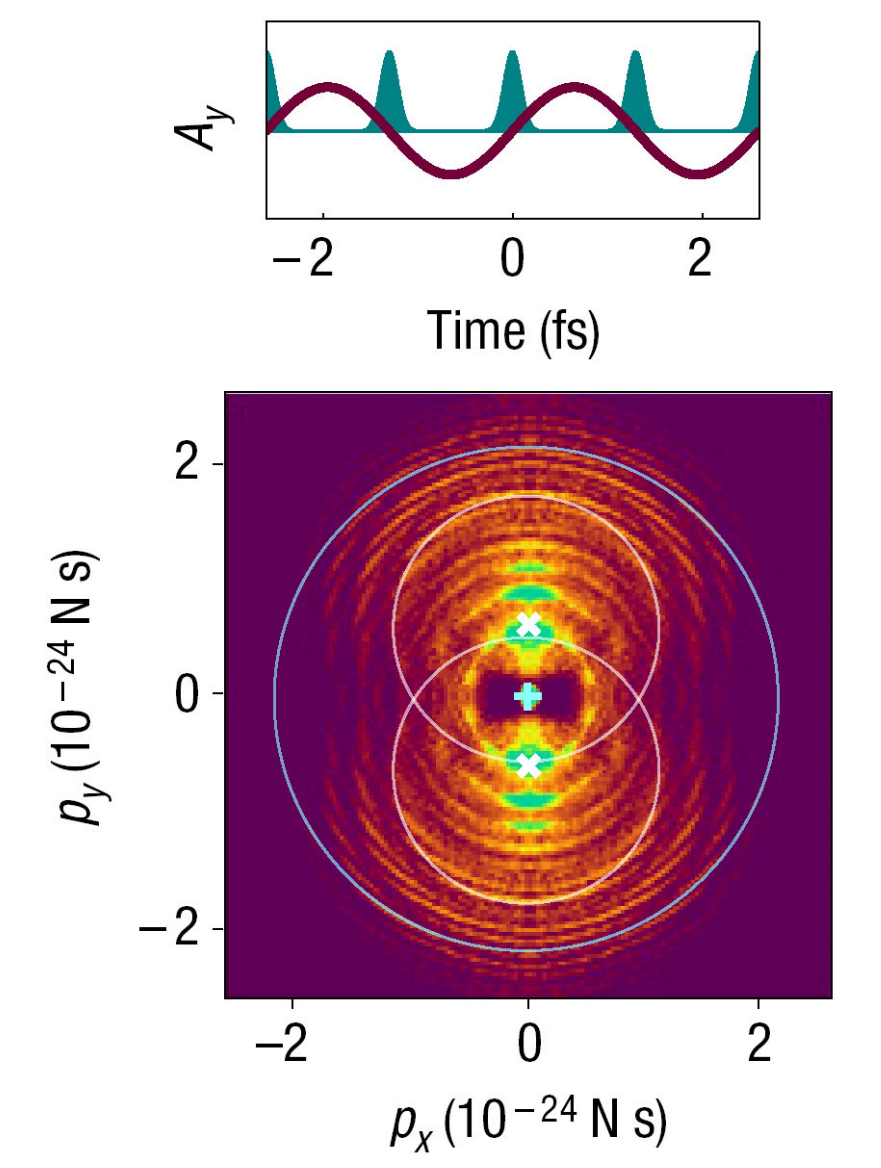
Simulation results:
Two-pulse Electron Wavepacket Interference



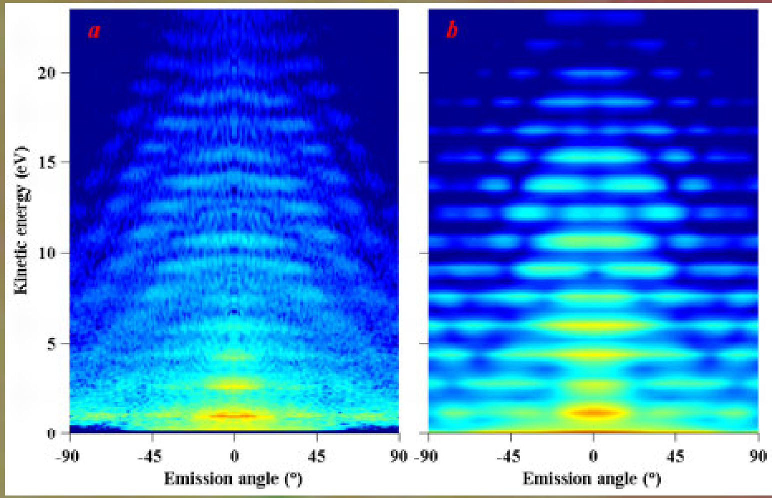
Can be used to measure properties of the infrared laser pulse

Can be used to measure the phase of the electron momentum wavefunction

Experimental results - 1



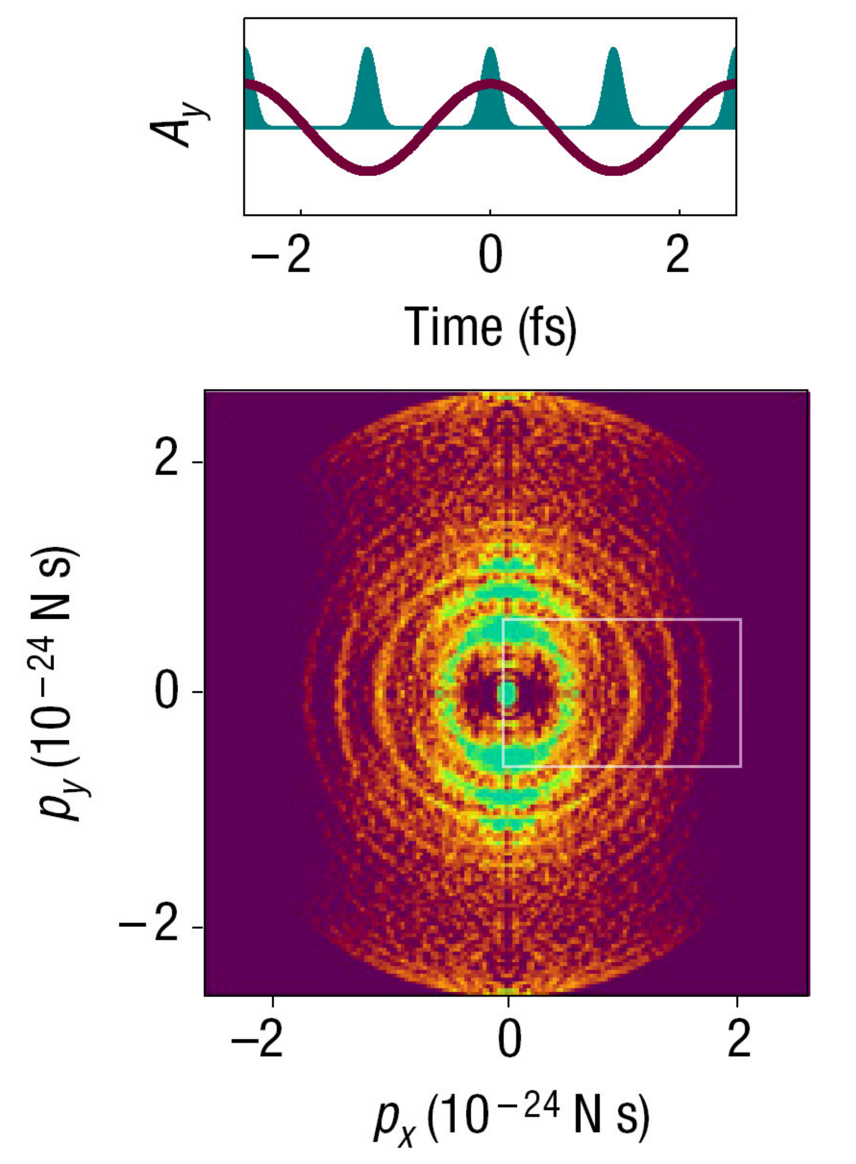
Experiment Calculation

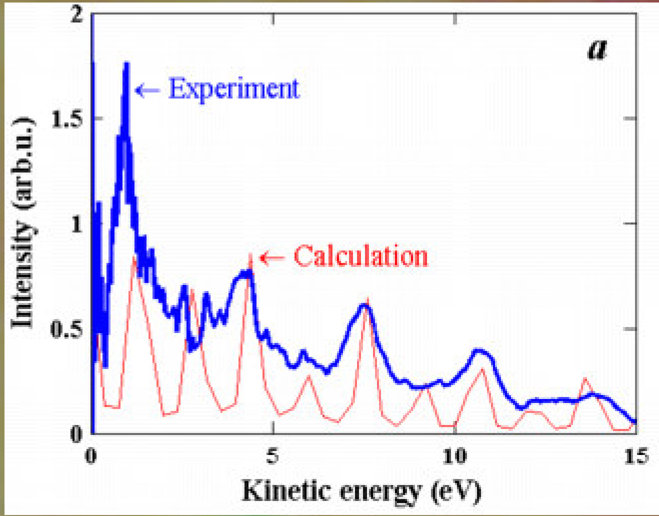


$$= W\frac{\pi}{\hbar\omega} + (-1)^k \frac{2eA_0\, p_y}{m\,\hbar\omega} \cos(\,\omega\tau)$$

Measuring the vector potential of the infrared laser pulse

Experimental results - 2





$$-\phi[\vec{p}+e\vec{A}(\frac{k\pi}{\omega},\tau)]+\phi[\vec{p}-e\vec{A}(\frac{k\pi}{\omega},\tau)]$$

Measuring the electronic wave function in momentum space

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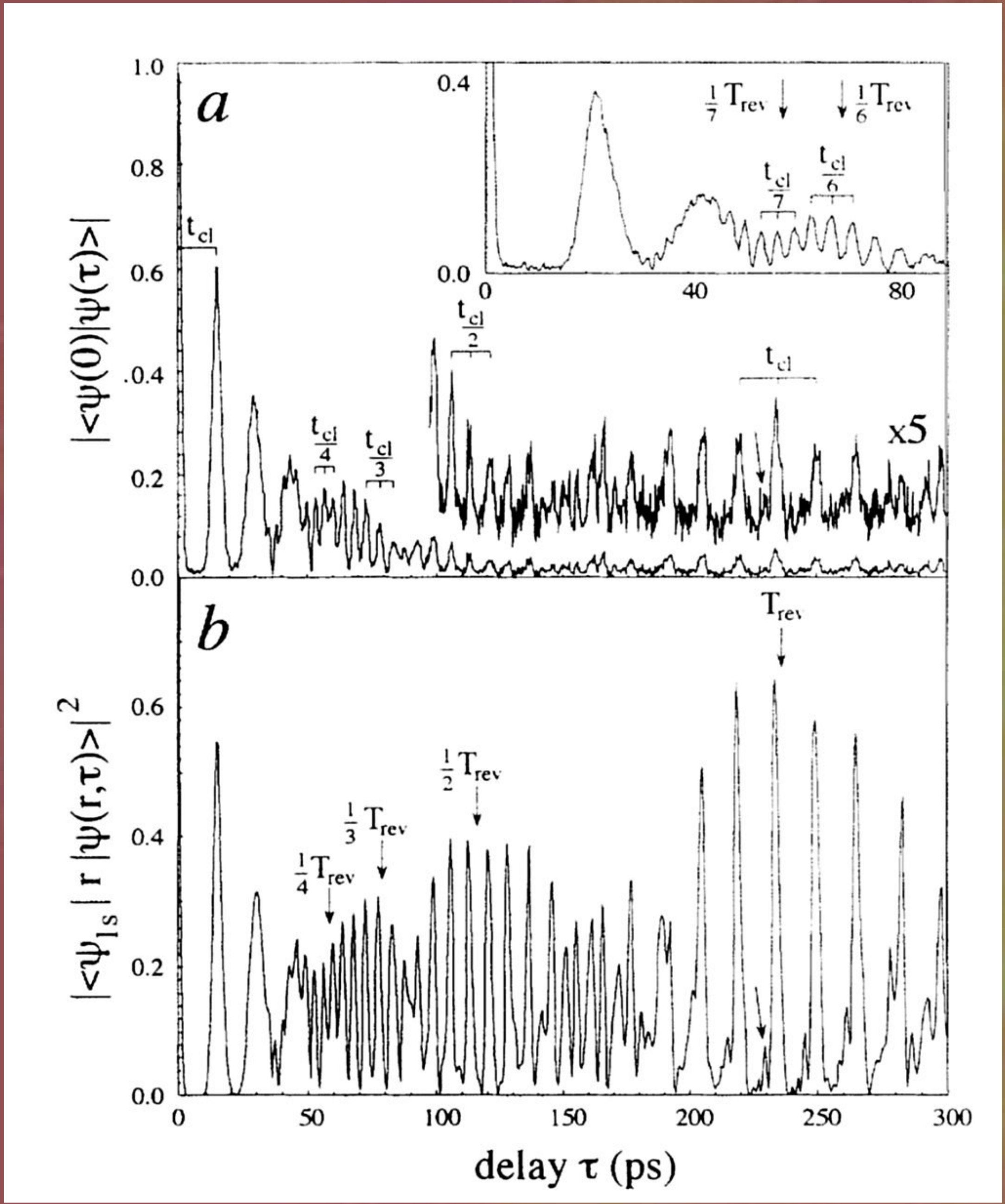

We can't do even the simplest attosecond XUV pump – attosecond XUV probe experiment!!

N.B. Studying electron dynamics with femtosecond lasers requires slowing down the dynamics by working with Rydberg atoms or molecules

Wals. et.al. Phys. Rev. Lett. 72, 3783 (1994).

Two Families of Attosecond Laser Experiments

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

   State of the art is ~ 10 μJoule/harmonic ($3x10^{12}$ photons/harmonic at 30 eV) → non-linear ionization "heroic"

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

   Typically ~ 10^7 photons/pulse or less

One “heroic” experiment

PRL **97,** 153904 (2006) PHYSICAL REVIEW LETTERS week ending 13 OCTOBER 2006

Interferometric Autocorrelation of an Attosecond Pulse Train in the Single-Cycle Regime

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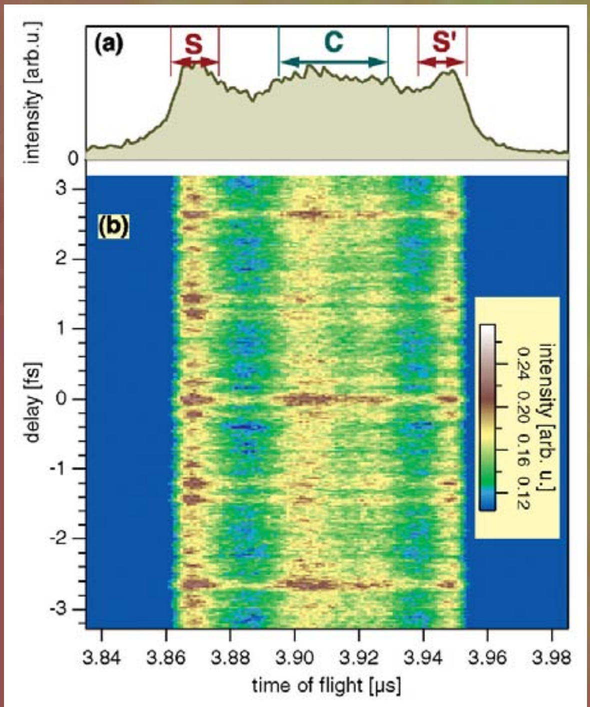
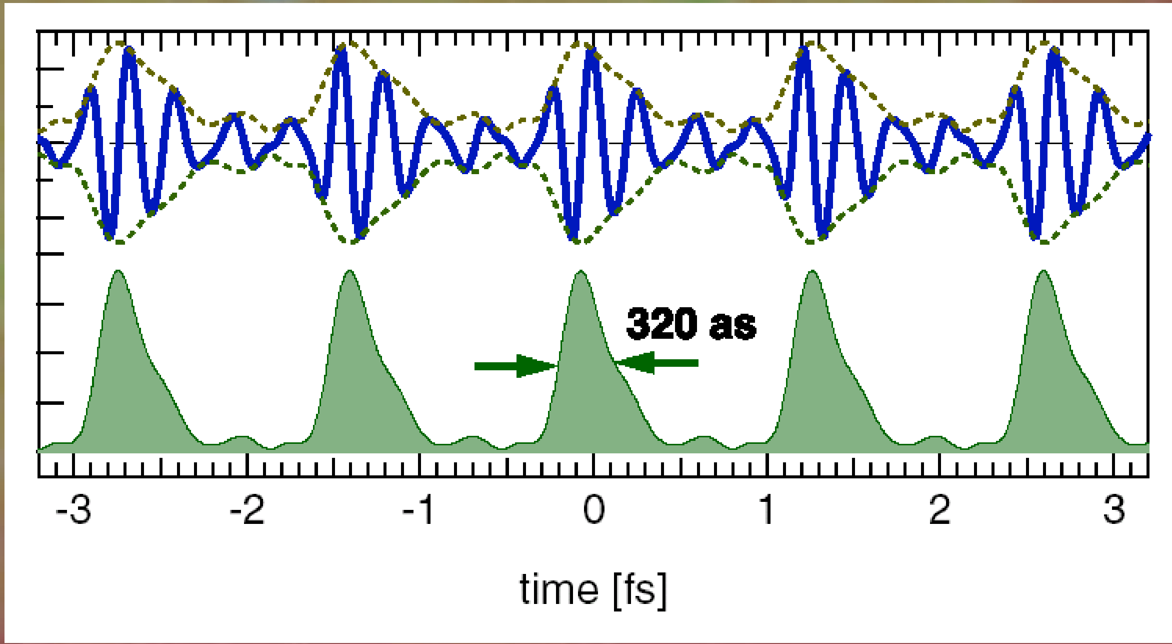




Overview: Attosecond Science from a "user" perspective

- What do we want?
- What do we use as our starting point to get this?
- What – as a result - can we presently do?
- What can we presently <u>not</u> do?
- What do we plan to do to get around this?

1) Our choices for going towards XUV-XUV experiments

Amplification of CEP-stable 30 fsec pulses to TW-level

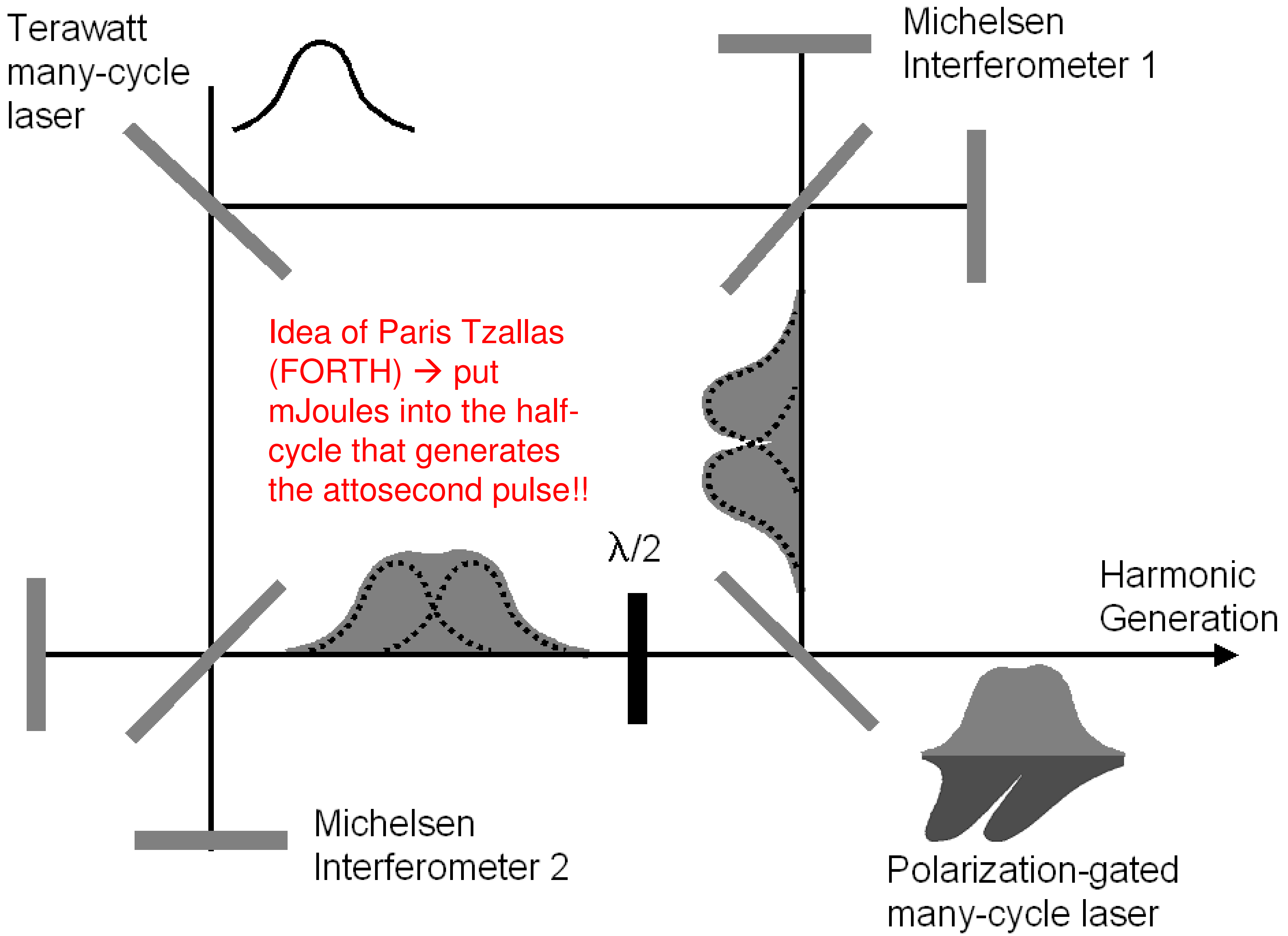
Specialized detectors: Development of hydrid COLTRIMS/Velocity Map Imaging detector

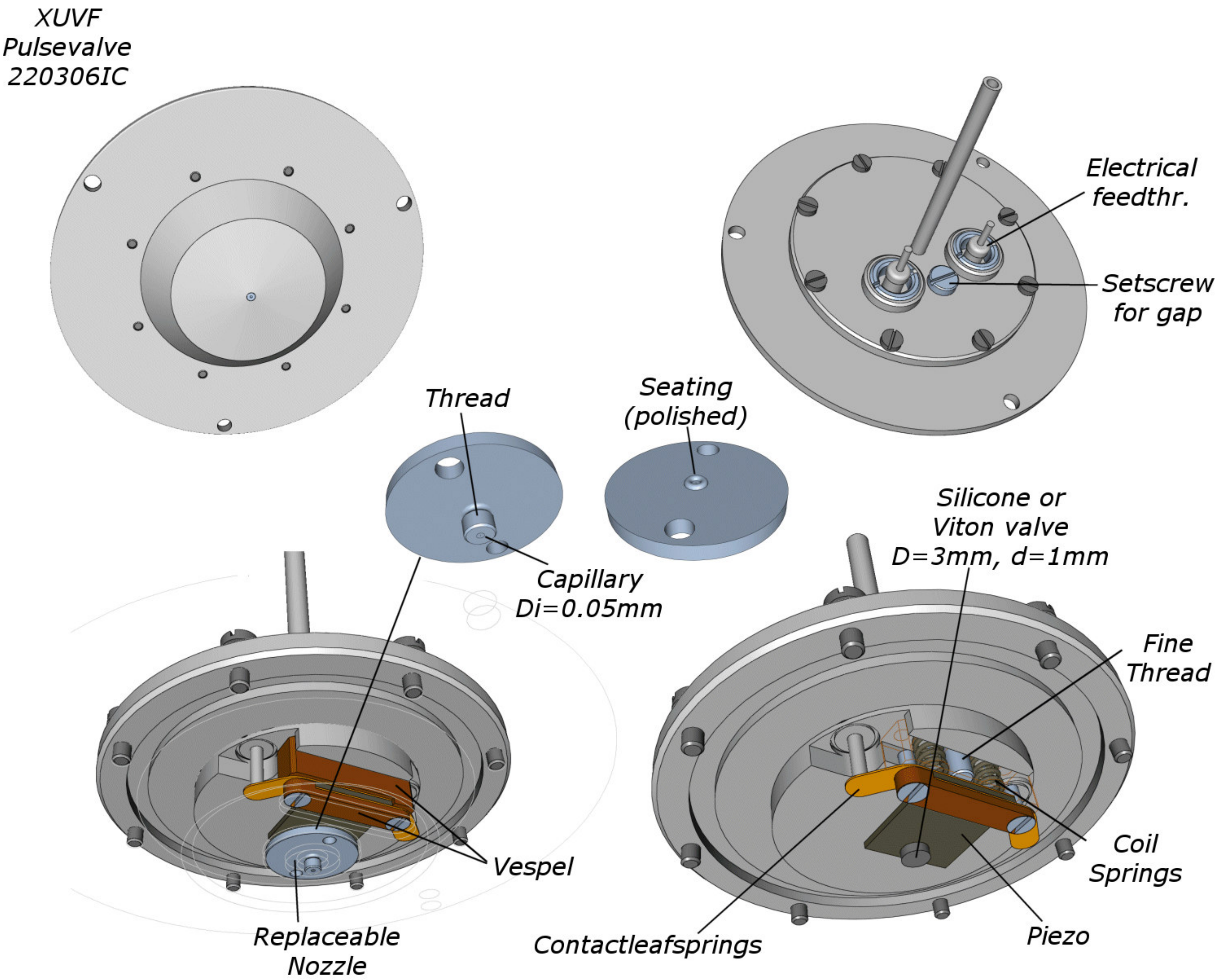
Successful Attosecond Experiment

Development of (chirped) XUV multilayer optics

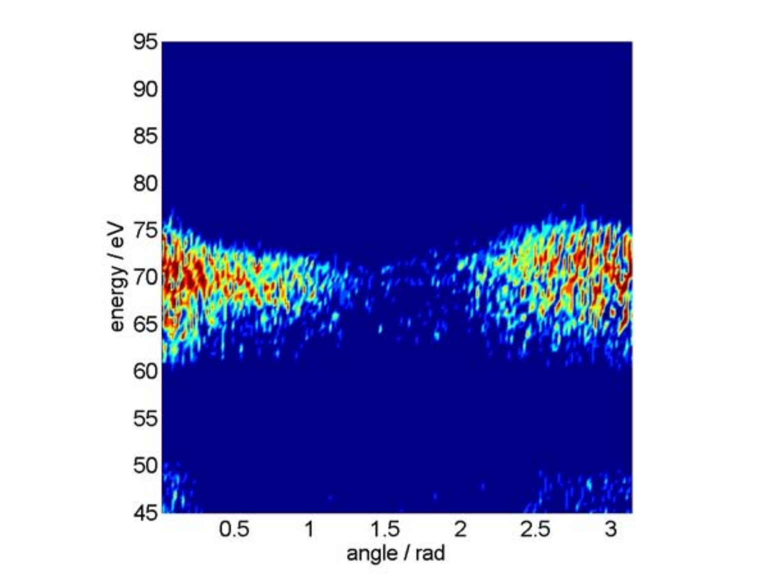
Special target injection

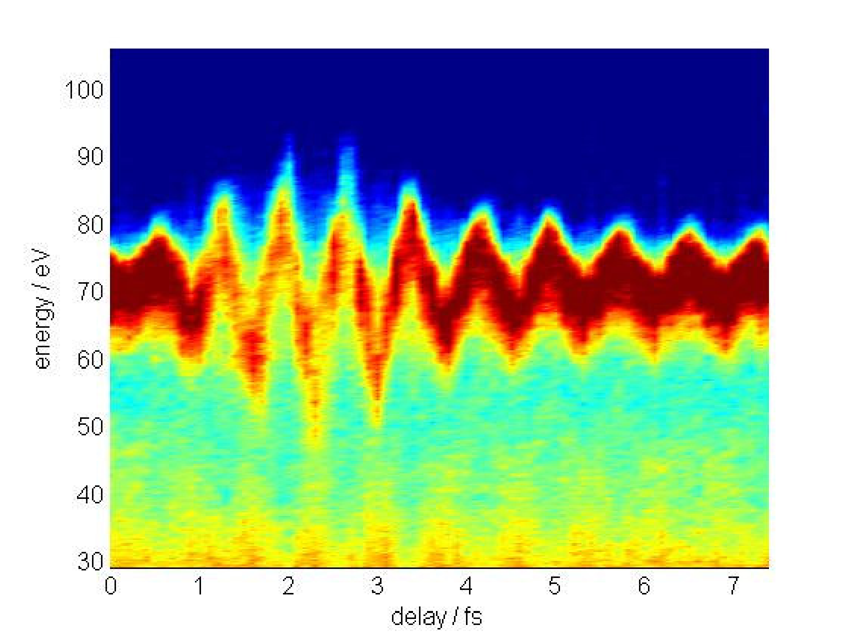
Polarization gating for isolated attosecond pulses + few-cycle UV


Terawatt many-cycle laser
Michelsen Interferometer 1
Idea of Paris Tzallas (FORTH) → put mJoules into the half-cycle that generates the attosecond pulse!!
λ/2
Harmonic Generation
Michelsen Interferometer 2
Polarization-gated many-cycle laser



XUVF
Pulsevalve
220306IC
Electrical
feedthr.
Setscrew
for gap
Thread
Seating
(polished)
Capillary
Di=0.05mm
Silicone or
Viton valve
D=3mm, d=1mm
Fine
Thread
Vespel
Coil
Springs
Replaceable
Nozzle
Contactleafsprings
Piezo



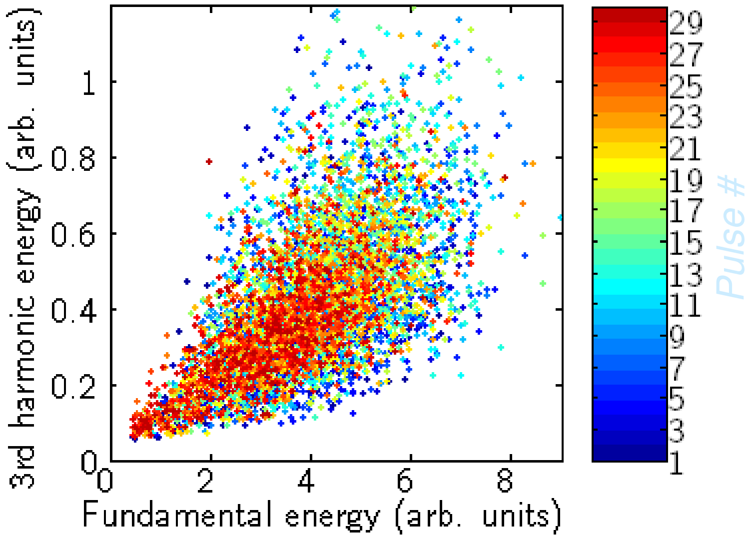


Streaking of electrons produced by ionization of Neon by a 90 eV isolated attosecond pulse (recently measured in Garching, using velocity map imaging)

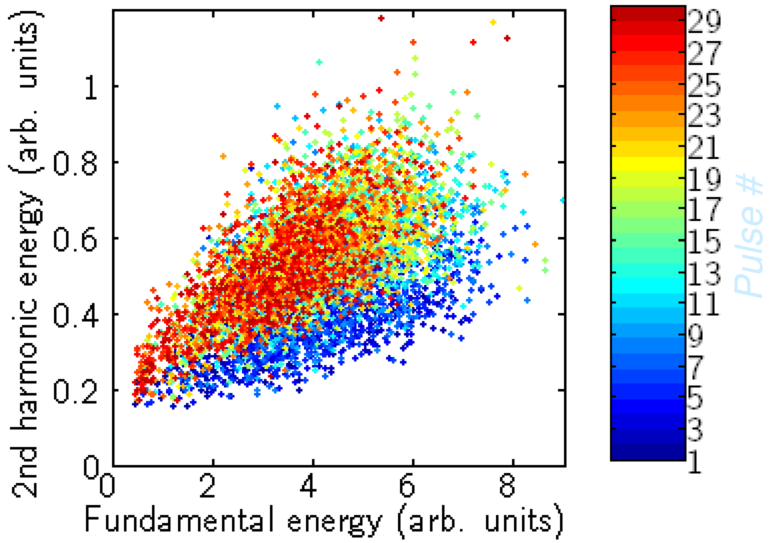
1) Exploiting the complementarity between harmonics-based and FEL-based experiments

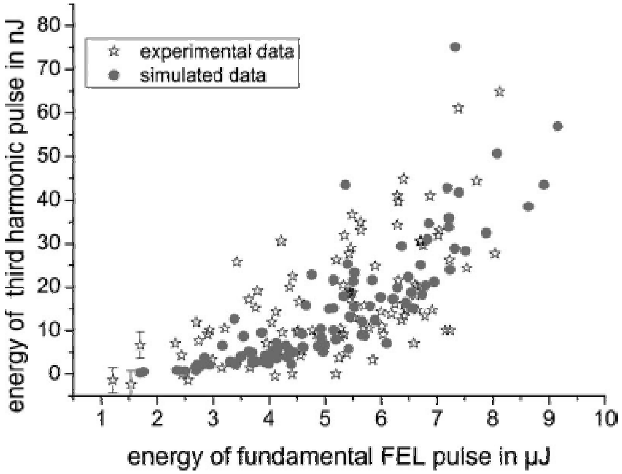
Experimental results: magnetic bottle

Third harmonic



Second harmonic

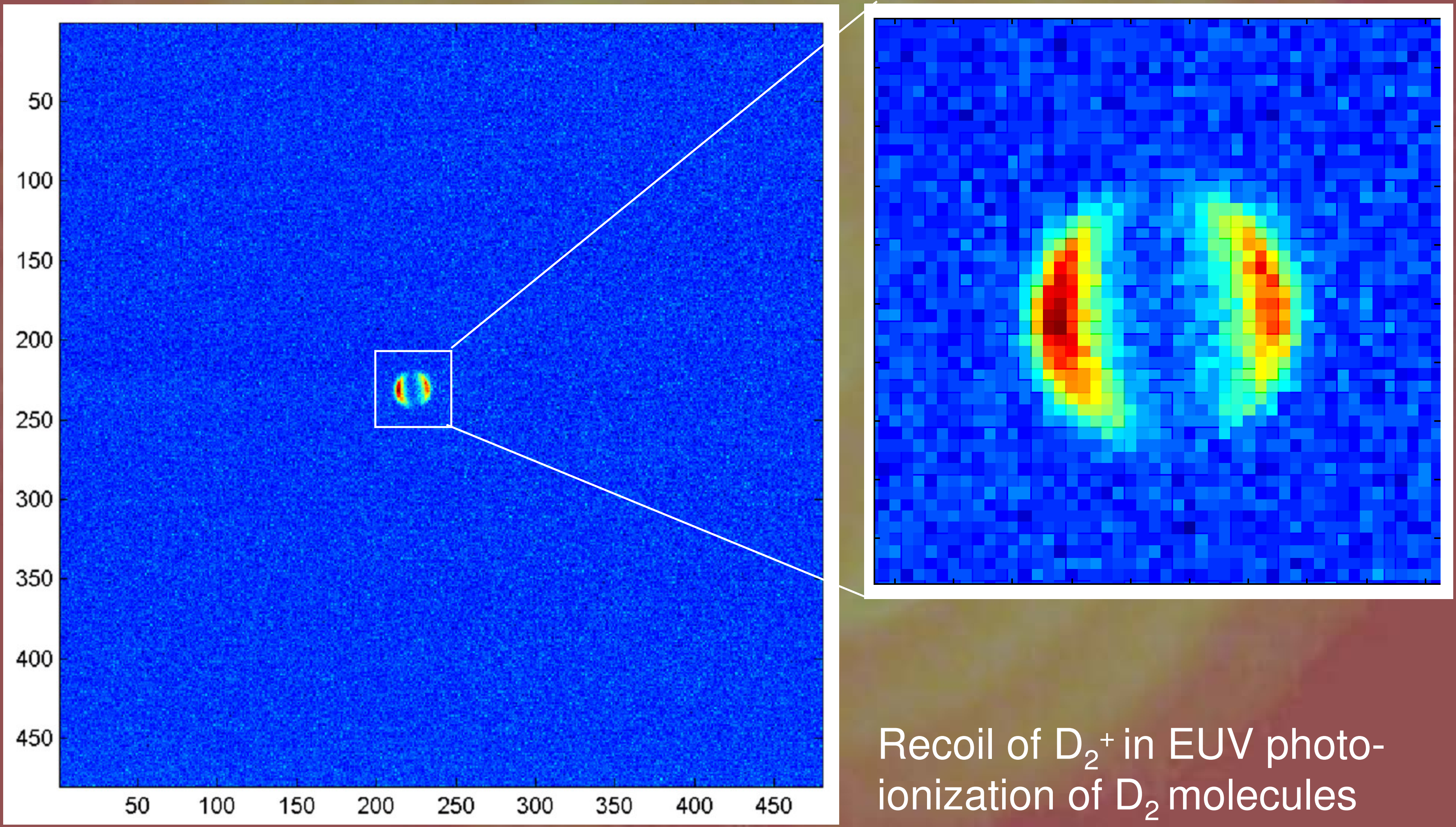




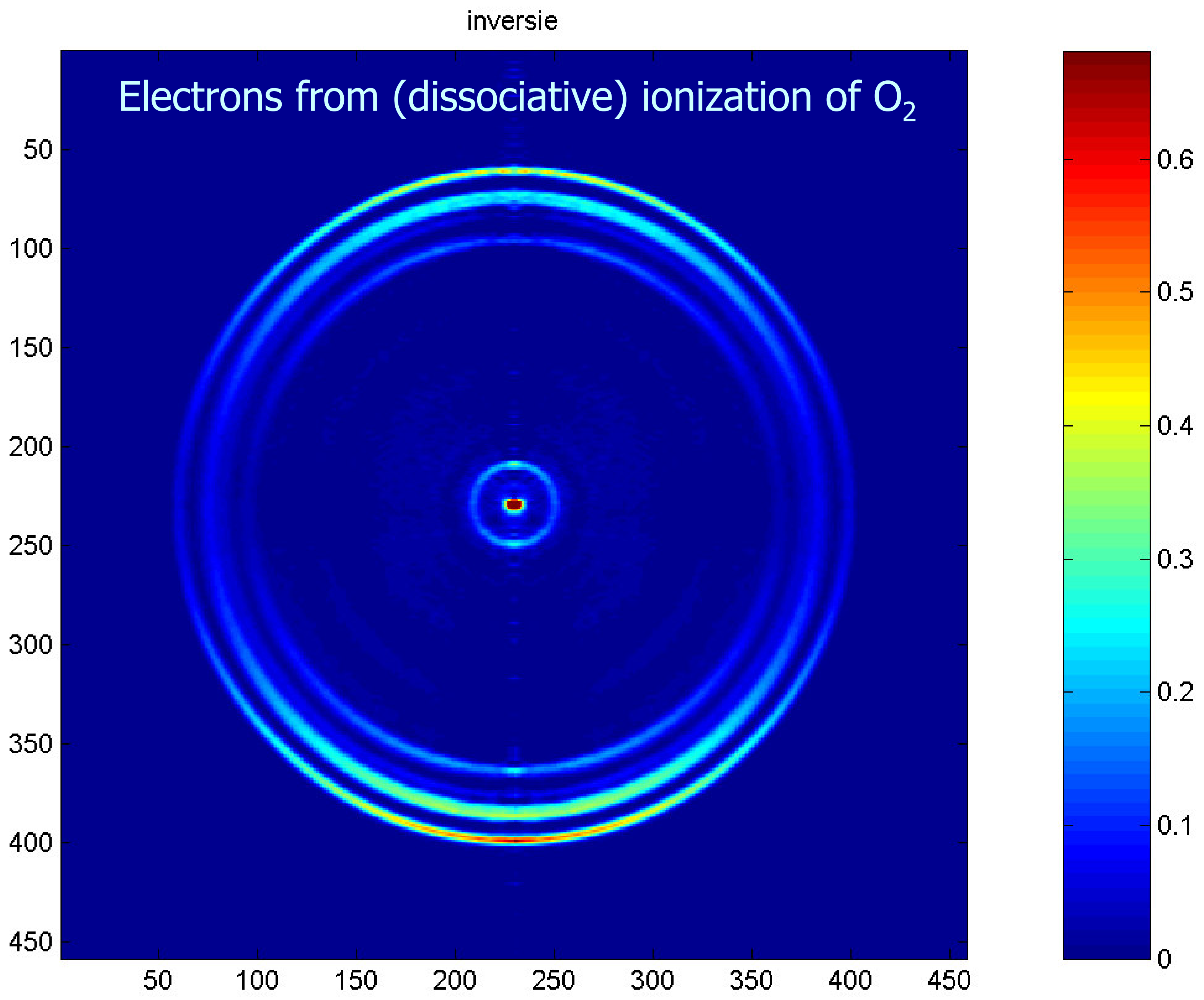
The ratio between the second harmonic and the fundamental increases towards the end of the bunches

S. Düsterer, et al., *Opt. Lett.*, **31,** 1750 (2006)

Experimental results: velocity map imaging

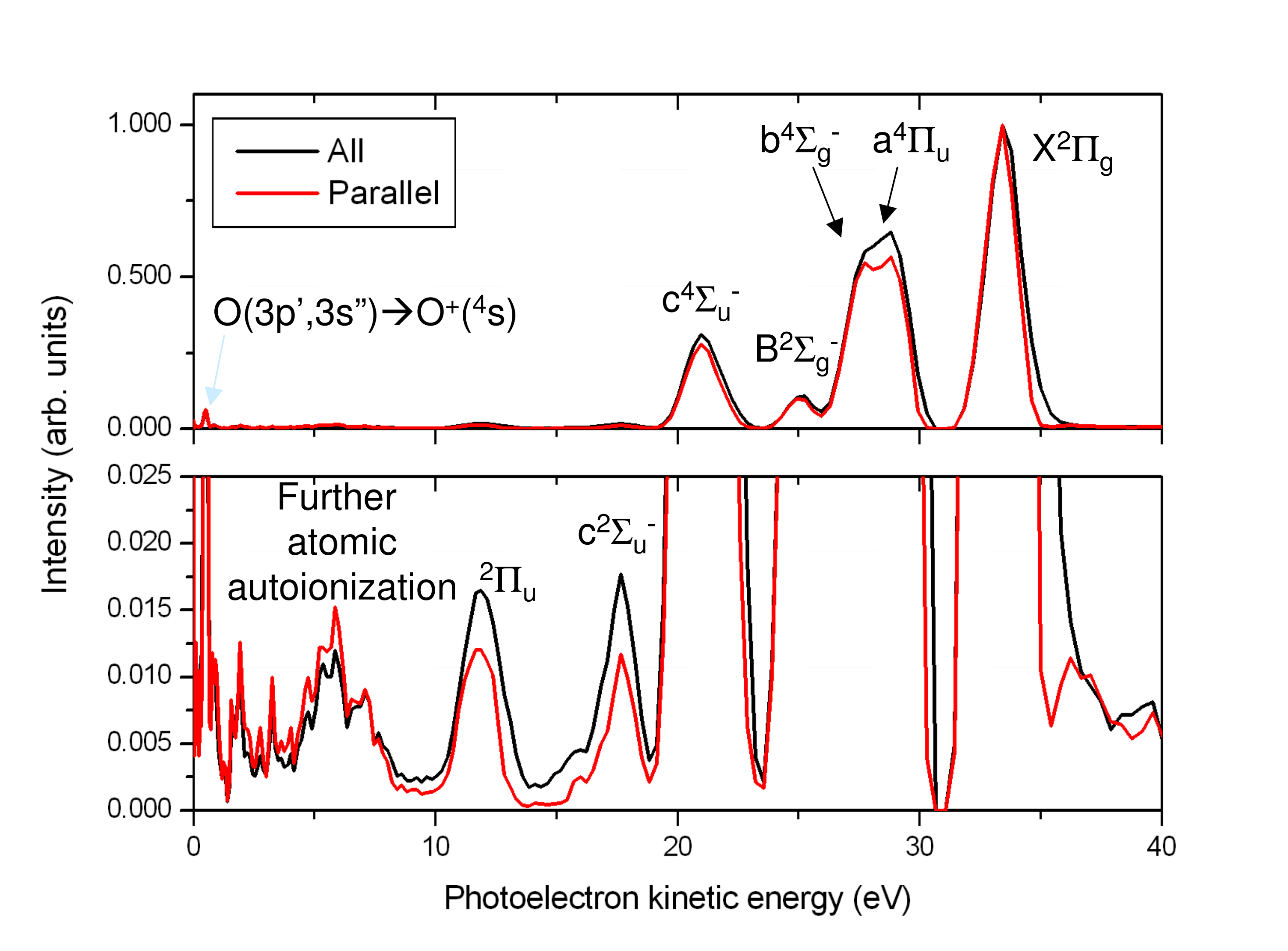


Recoil of D_2^+ in EUV photo-ionization of D_2 molecules



inversie
Electrons from (dissociative) ionization of O_2
50
100
150
200
250
300
350
400
450
0.6
0.5
0.4
0.3
0.2
0.1
0

Explore the utility of velocity map imaging at the FEL : O_2



Conclusions and Outlook

❖ Attosecond science is beginning to answer questions!

❖ Isolated attosecond pulses:
- monitoring of electron dynamics in pump-probe experiments
(first step: CEP control of electron dynamics in D_2) and
- monitoring of shake-up & Auger processes in Ne and Xe

❖ Attosecond pulse trains:
- used in attosecond electron wave packet interferometry

❖ Eventual goal: insight into the elementory electronic processes that occur during photo-absorption and that accompany chemical rearrangements

❖ Synergie between harmonics-based attosecond sources and XUV FELs will aid both fields