Attosecond laser driven electron dynamics in molecules

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Outline

Attosecond intramolecular laser driven electron dynamics

- Sub-laser-cycle dynamics in H_2^+
- Impact on ionization and high harmonic generation

Generation of zeptosecond waveforms at long HHG driver wavelengths

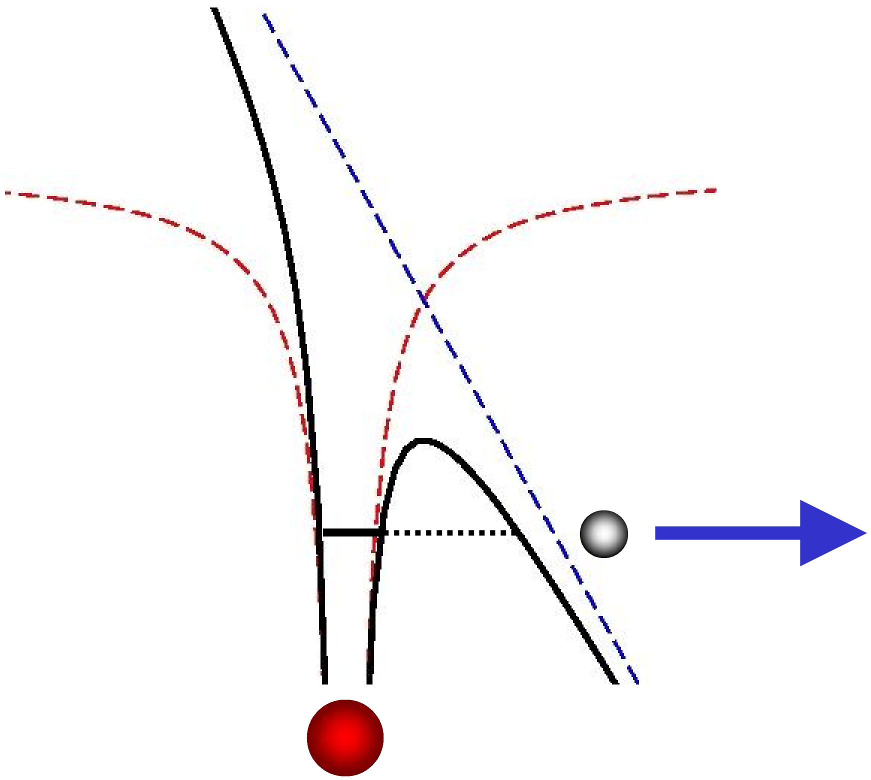
Contributions by

Michelle Miller
Carlos Hernandez Garcia
Agnieszka Jaron-Becker

Feng He (Shanghai Jiao Tong University, China)
Norio Takemoto (MPI Korea / MPI-PKS Dresden, Germany)

Reinhard Dörner (Frankfurt, Germany)
Uwe Thumm (Kansas State University, USA)

Tunnel ionization picture (Atom)

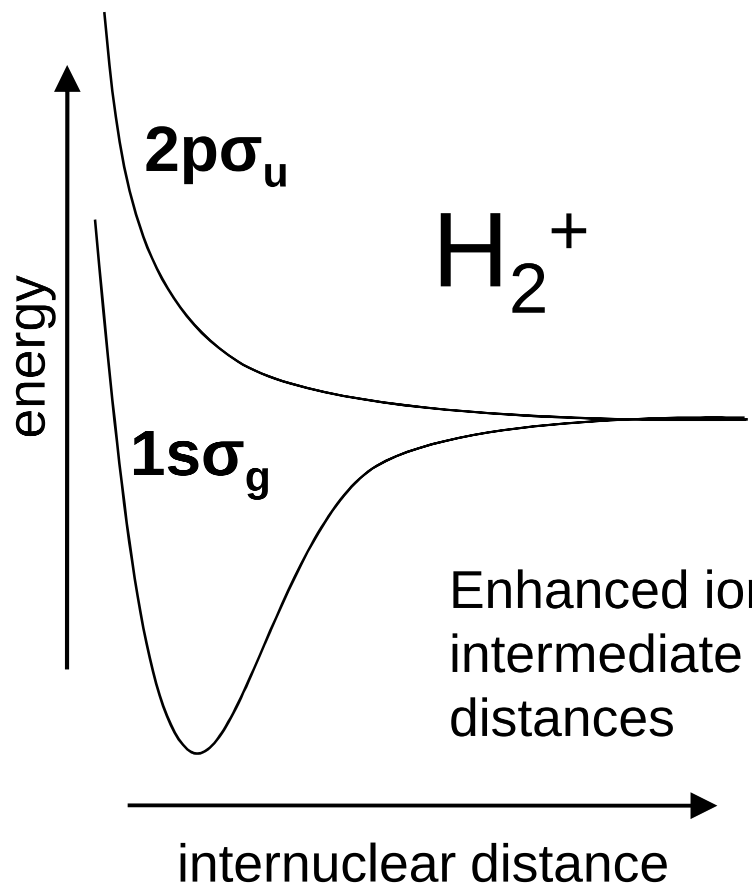


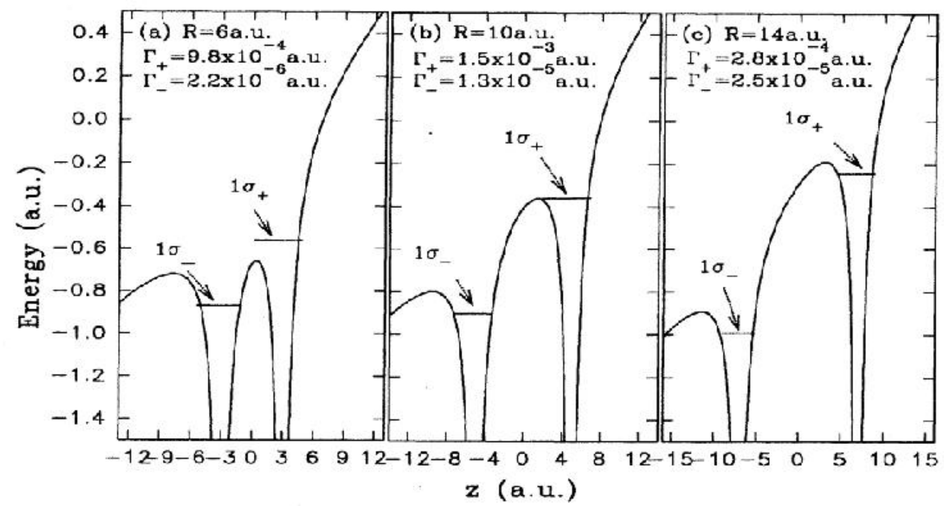
In a pulse:
- ionization strongest at peak

Over a cycle of an oscillating field:
- ionization strongest at field maxima
  (Exp.: M.Uiberacker et al., Nature **446**, 627 (2007))

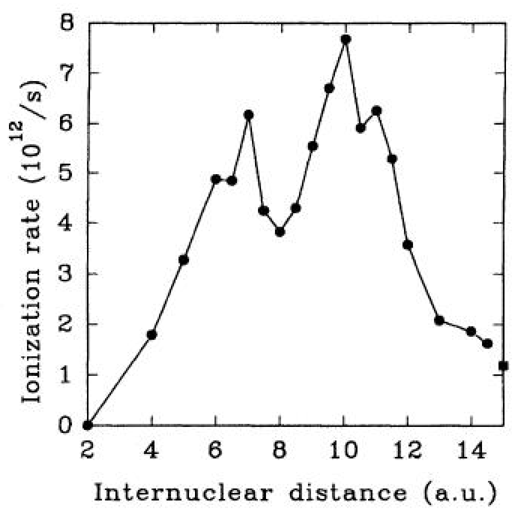
Time-resolved tunnel ionization: e.g., G.L. Yudin and M.Yu. Ivanov, Phys. Rev. A 64, 013409 (2001)

Ionization of hydrogen molecular ion





Enhanced ionization at intermediate internuclear distances

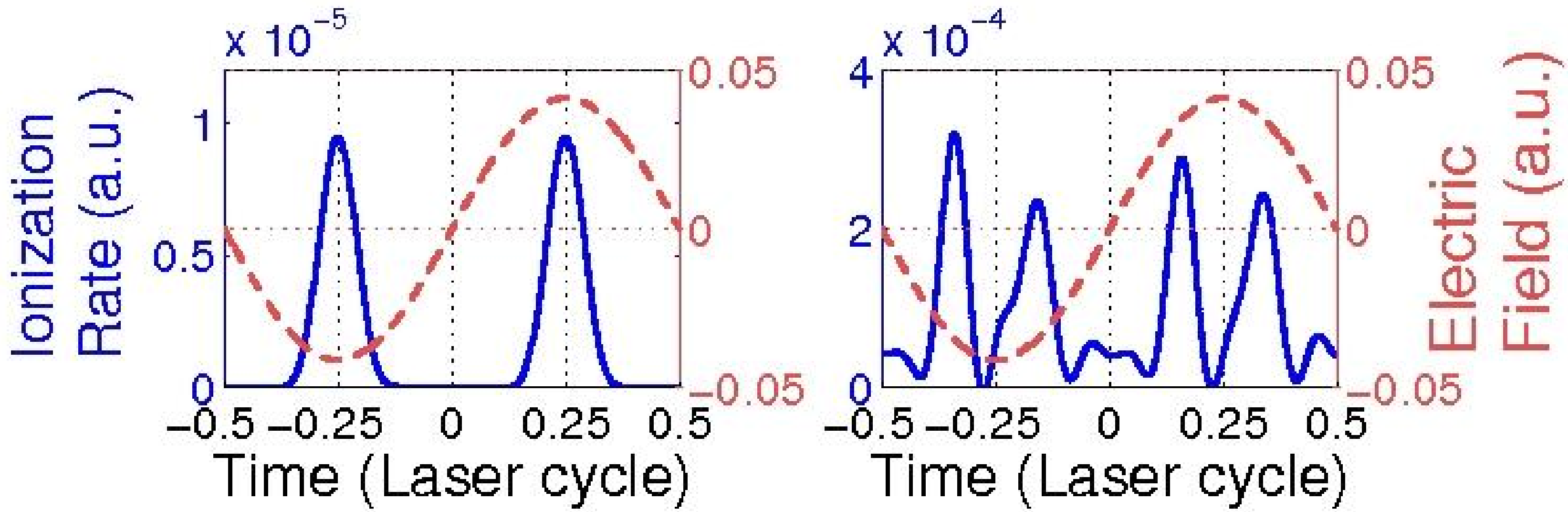


T. Zuo and A.D. Bandrauk, Phys. Rev. A 52, R2511 (1993); also T. Seideman et al., Phys. Rev. Lett. 75, 2819 (1995)

Ionization dynamics

Hydrogen atom

Hydrogen molecular ion
(6.75 a.u. < R < 7.25 a.u.)

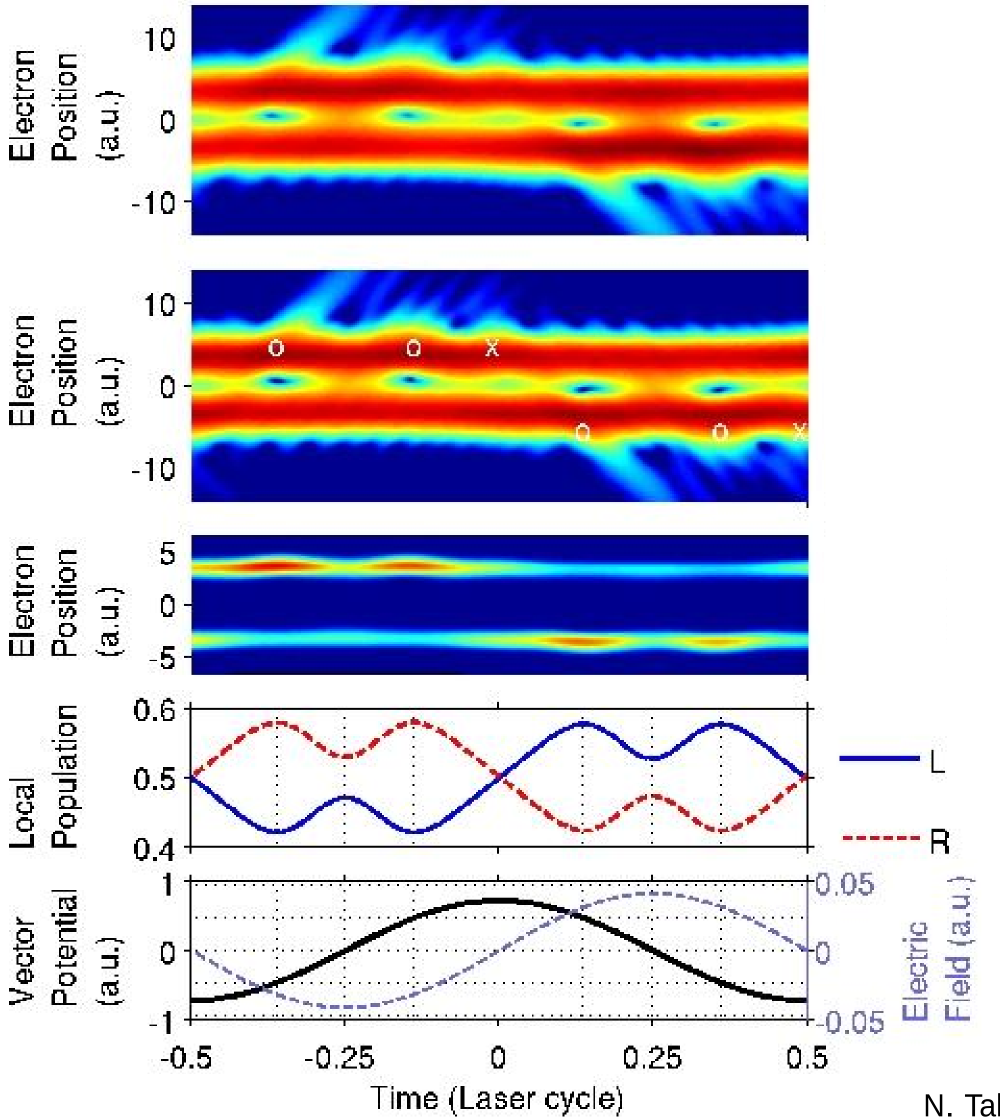


Results of numerical simulations for H_2^+ do not agree with expectation from ionization picture.

800 nm, 6 x 10^{13} W/cm^2, sin^2-pulse, 26.7 fs (FWHM), central field cycle

N. Takemoto and A. Becker, Phys. Rev. Lett. 105, 203004 (2010)

Analysis



Dimensions of electron dynamics

No influence

Electron-nuclear dynamics

No influence

Rescattering effects

No effect on timing

Intramolecular dynamics

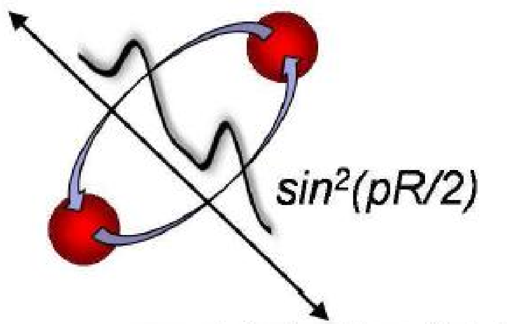
Transient localization

Two-state model

Captures the dynamics

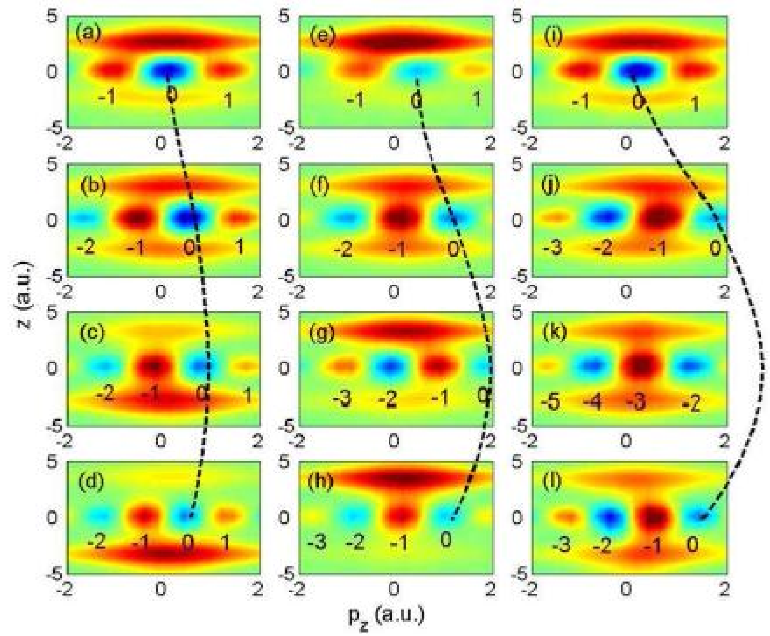
N. Takemoto and A. Becker, Phys. Rev. Lett. 105, 203004 (2010)

Momentum gates



modulated by field
p → p – A(t)/c

3x10^{12} W/cm^2 2x10^{13} W/cm^2 10^{14} W/cm^2



Wigner distribution:

$$W(z, p_z; t) = \frac{1}{\pi}\int \rho d\rho dR \int_{-\infty}^{\infty} dy \Phi^*(R, z+y, \rho; t)\Phi(R, z-y, \rho; t)\exp(2ip_z)$$

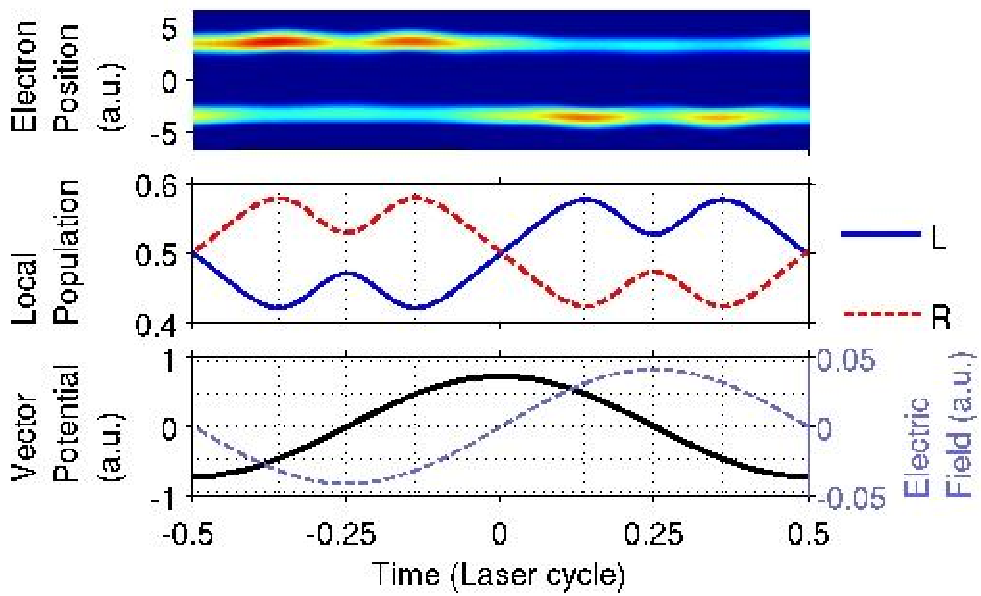
At higher intensities electron gets driven back and forth several times over one field cycle and may localize more than once at the same proton.

F. He et al., Phys. Rev. Lett. 101, 213002 (2008)

Localization condition

Two state model provides condition for electron localization

$$A_0 \sin(\mu t_{loc}) = m \frac{®}{2 < g \,|\, z \,|\, u >} \;, m = 0, \pm 1, \pm 2, \ldots$$



N. Takemoto and A. Becker, Phys. Rev. A 84, 023401 (2011)

Intermediate distance:

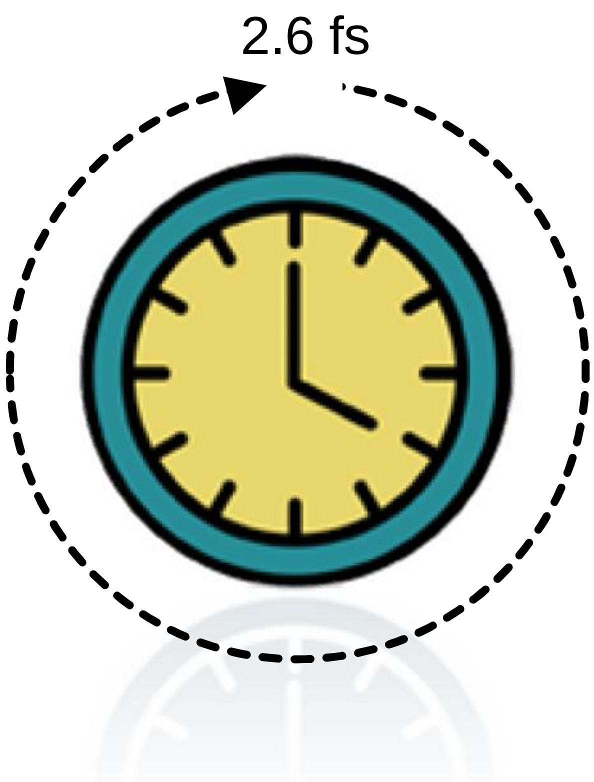
$$< g \,|\, z \,|\, u > \text{large}$$

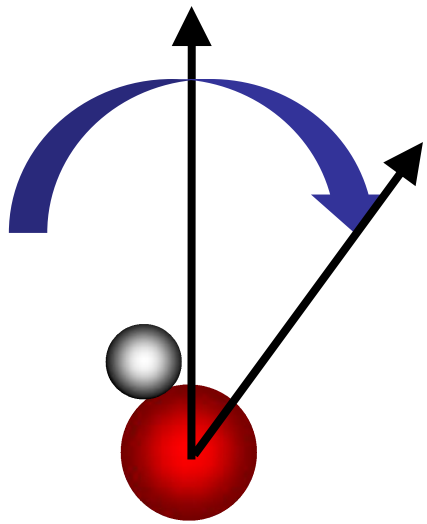
$$A_0 > \frac{®}{2 < g \,|\, z \,|\, u >}$$

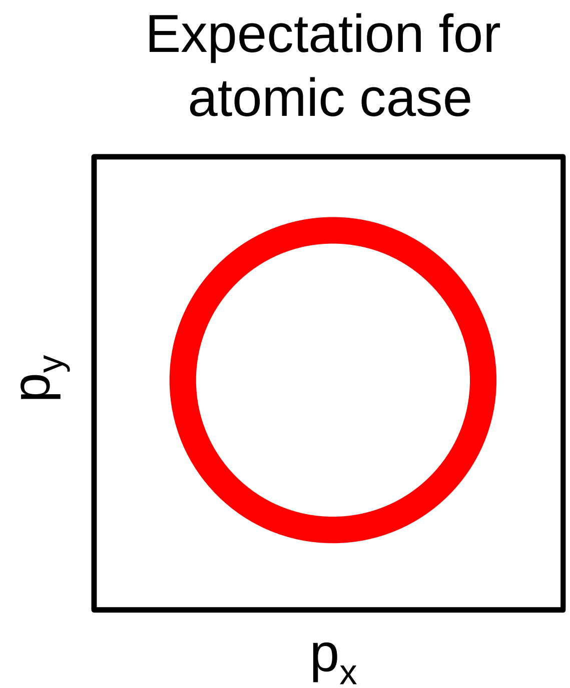
Condition fulfilled for m≠0

Localization (Ionization) more than once over one half cycle

Probing electron dynamics with attoclock



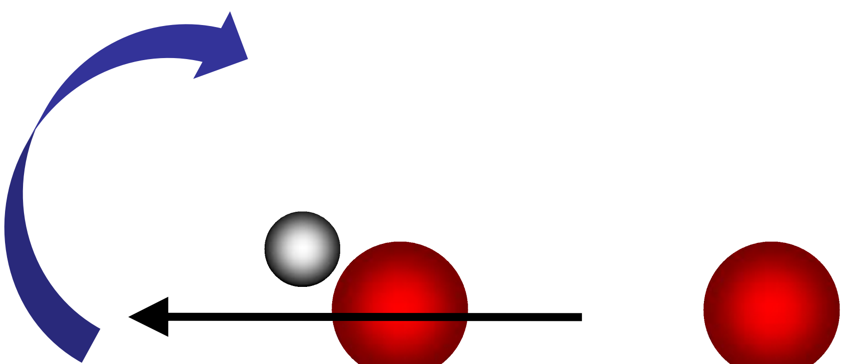


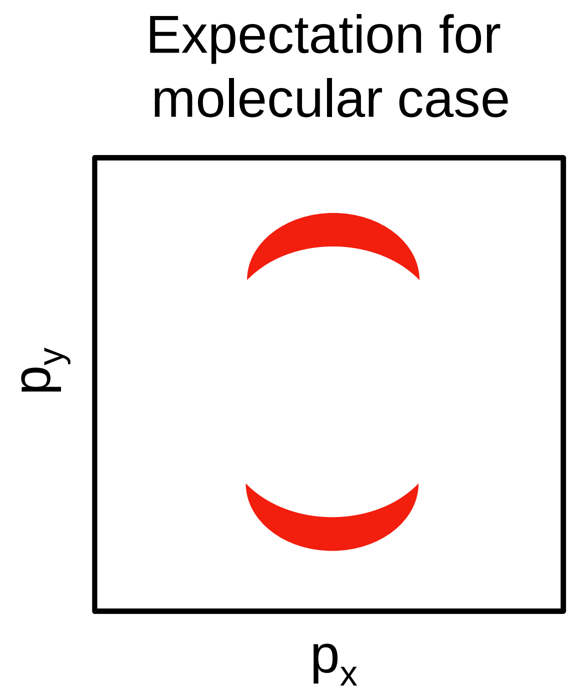


Use circularly polarized light, polarization direction acts as clock hand and determines at which time (during one field cycle of 2.6 fs) the electron is emitted.

idea first demonstrated for atoms by U.Keller (ETH Zűrich) and R.Dörner (U Frankfurt)

Probing electron dynamics with attoclock





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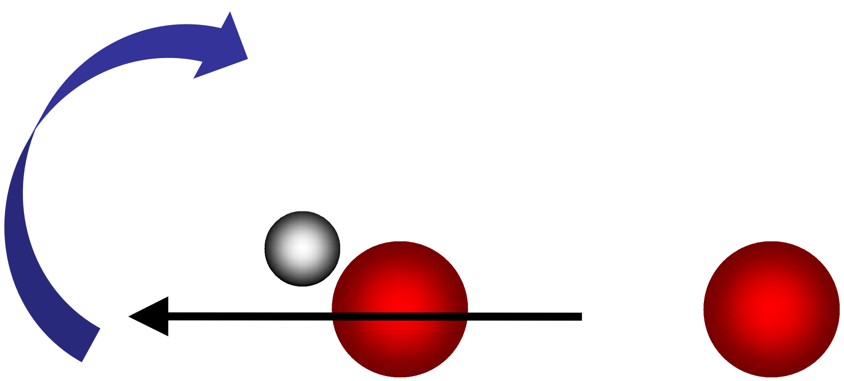
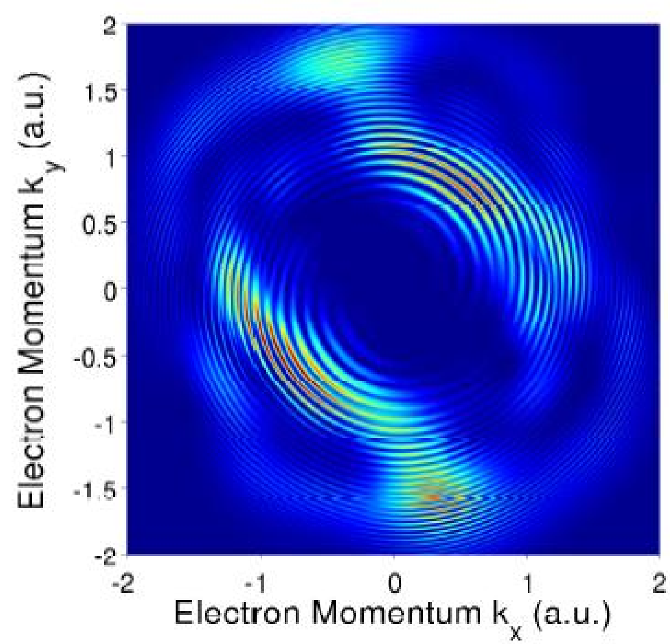
Probing electron dynamics with attoclock



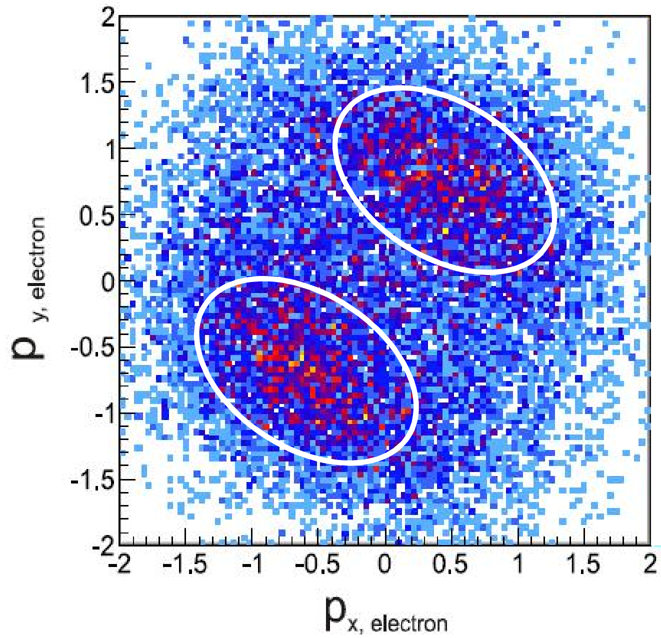
Tilted distribution for H_2^+



800 nm, 6×10^{14} W/cm^2
4-cycle (FWHM), R=7 a.u.

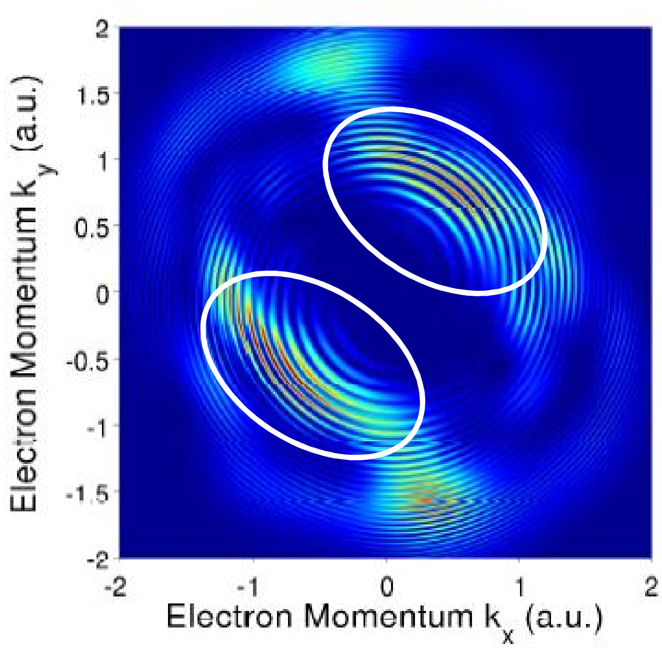
Comparison with experiment

Experiment (R. Dörner)



780 nm, $6x10^{14}$ W/cm^2
40 fs (FWHM)

Theory

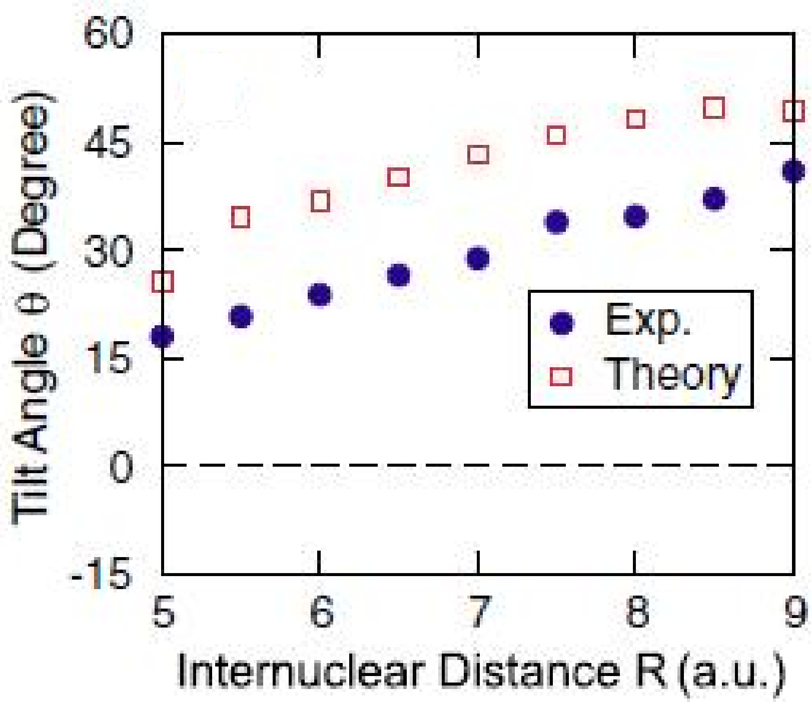


800 nm, $6x10^{14}$ W/cm^2
4-cycle (FWHM)

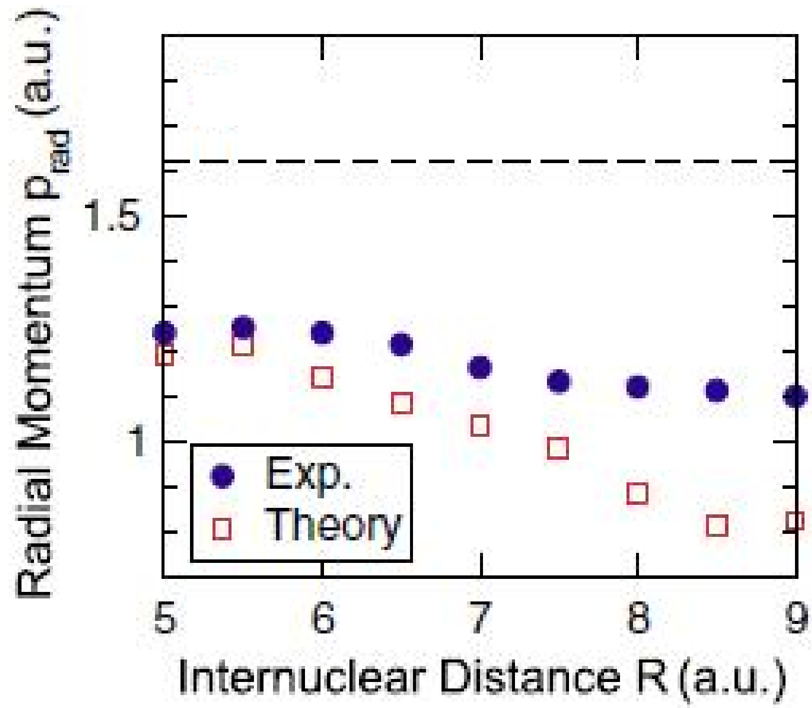
M. Odenweller et al., Phys. Rev. Lett. 107, 143004 (2011)

Comparison with experiment

Tilt angle

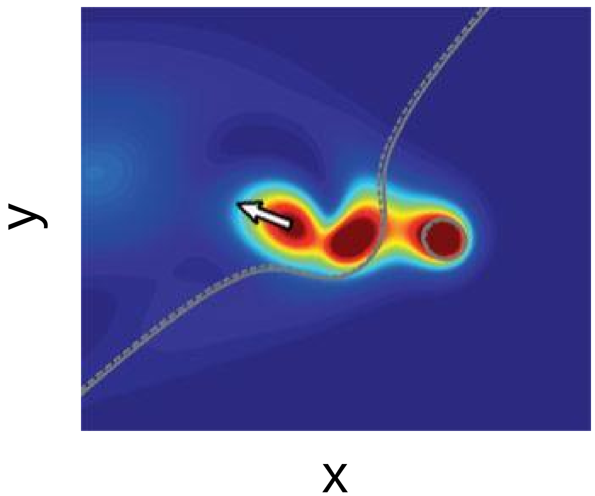


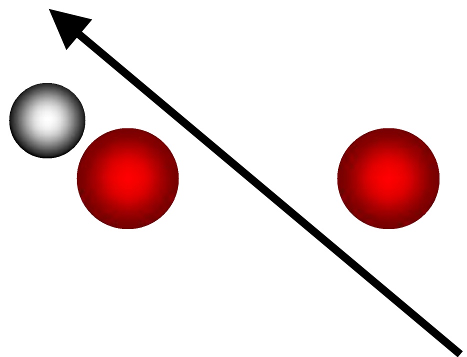
Radial momentum

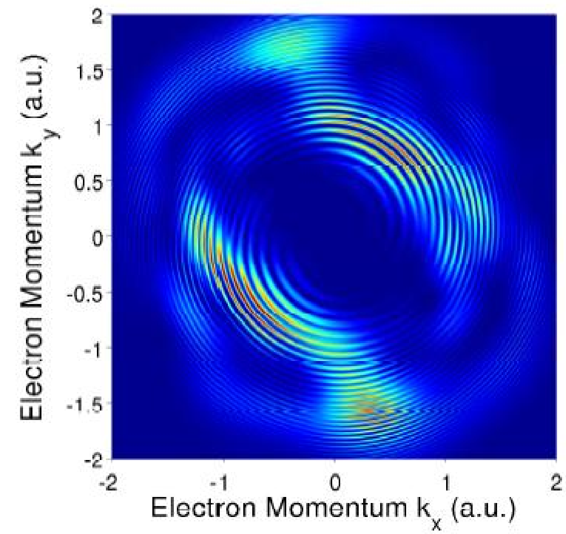


M. Odenweller et al., Phys. Rev. Lett. 107, 143004 (2011)

Unexpected ionization event







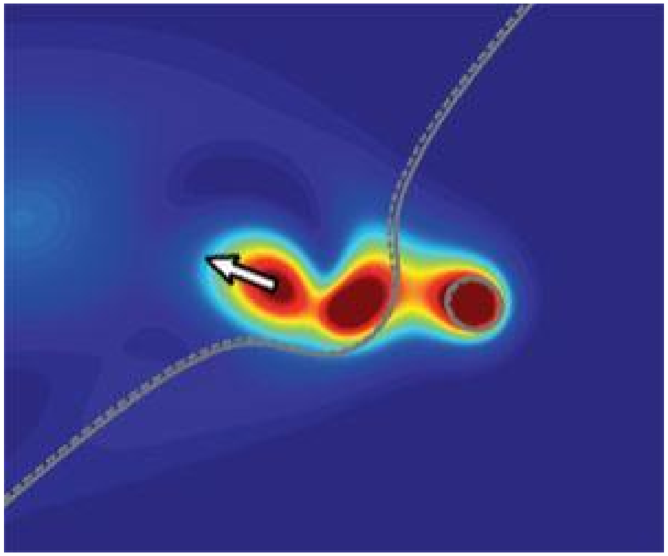
Delayed (by 356 as) unexpected ionization event

similar to prediction for linear polarization

M. Odenweller et al., Phys. Rev. Lett. 107, 143004 (2011)

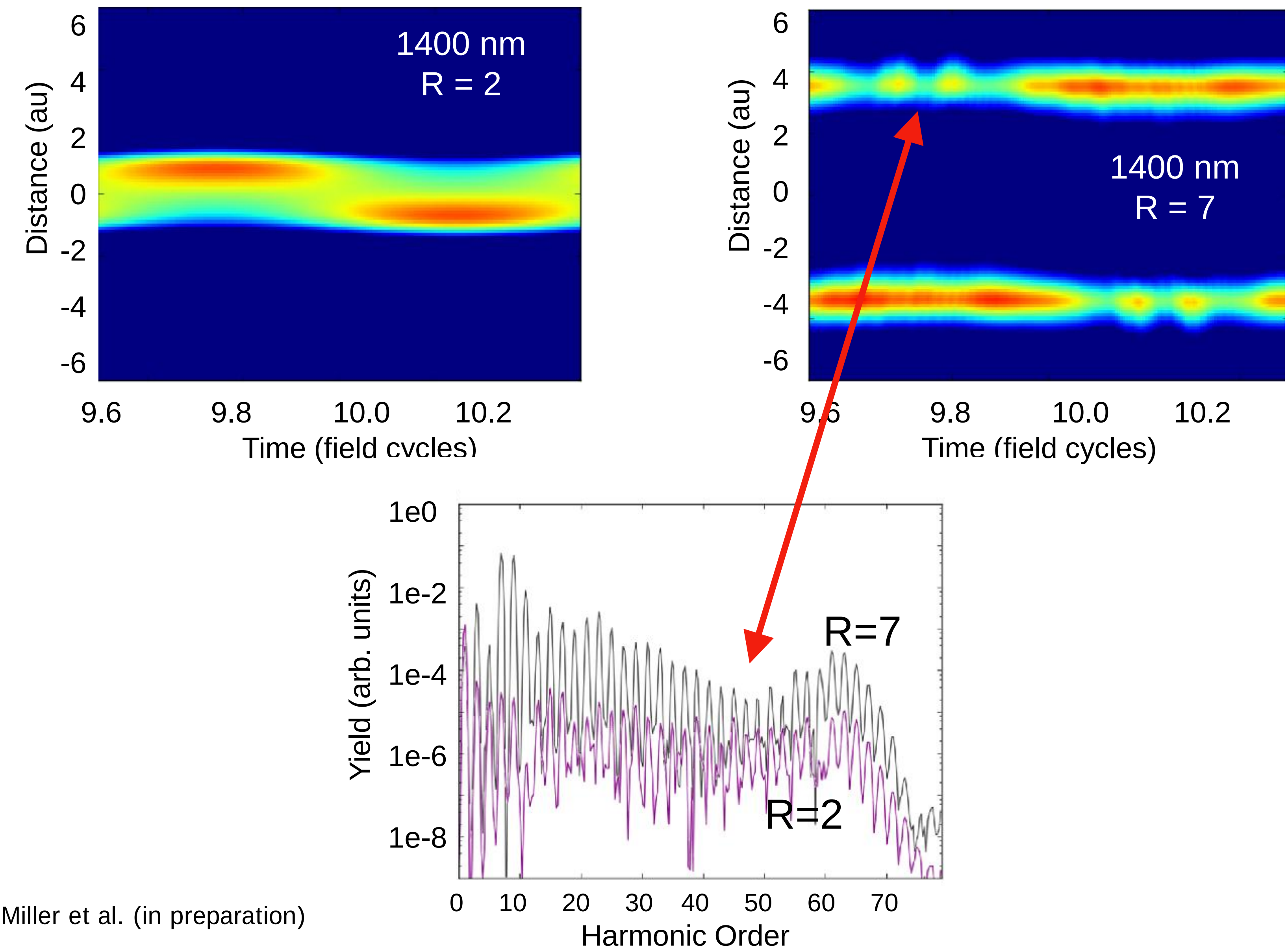
A molecular movie

“Attoclock turns electrons into movie stars”
(Lisa Grossman, New Scientist)



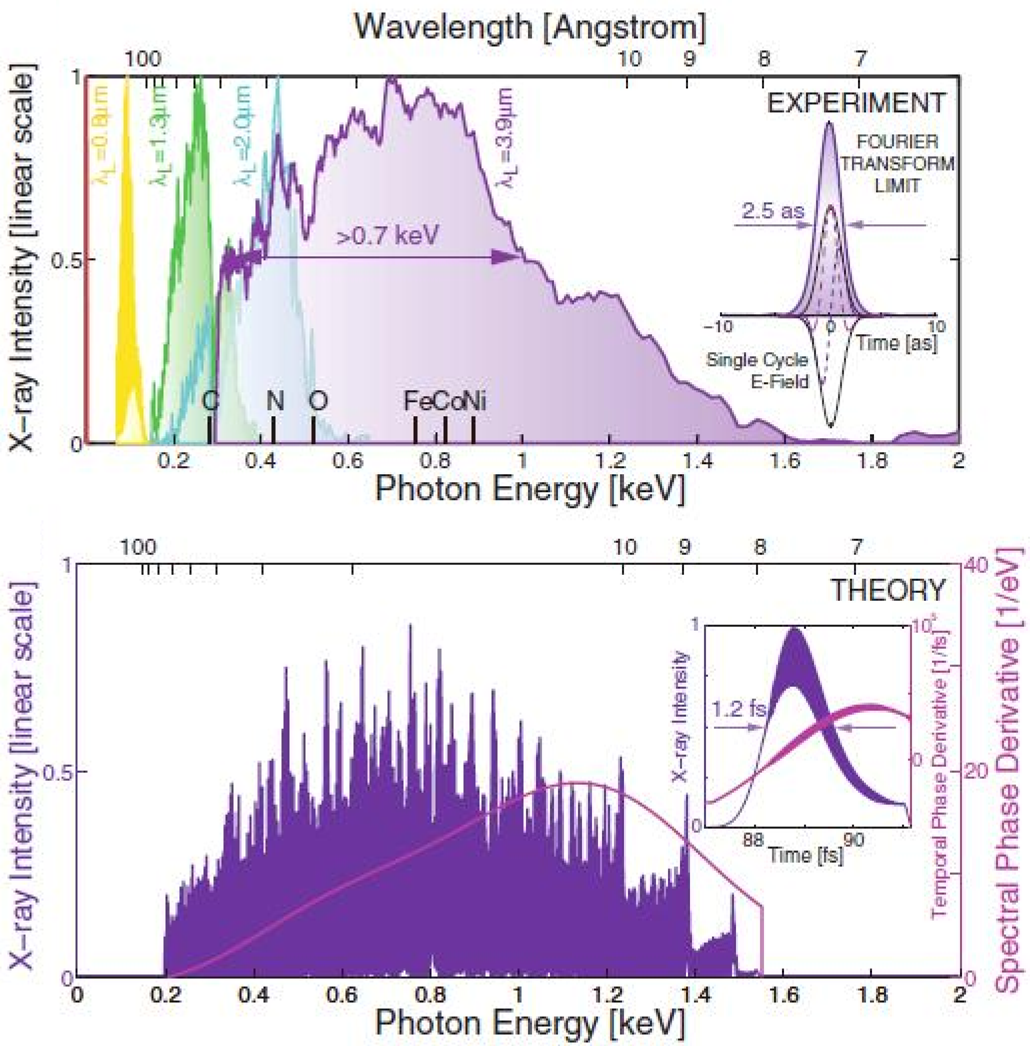
“The Steve McQueen of the subatomic world”

Attosecond dynamics and HHG



M. Miller et al. (in preparation)

keV X-ray generation via HHG



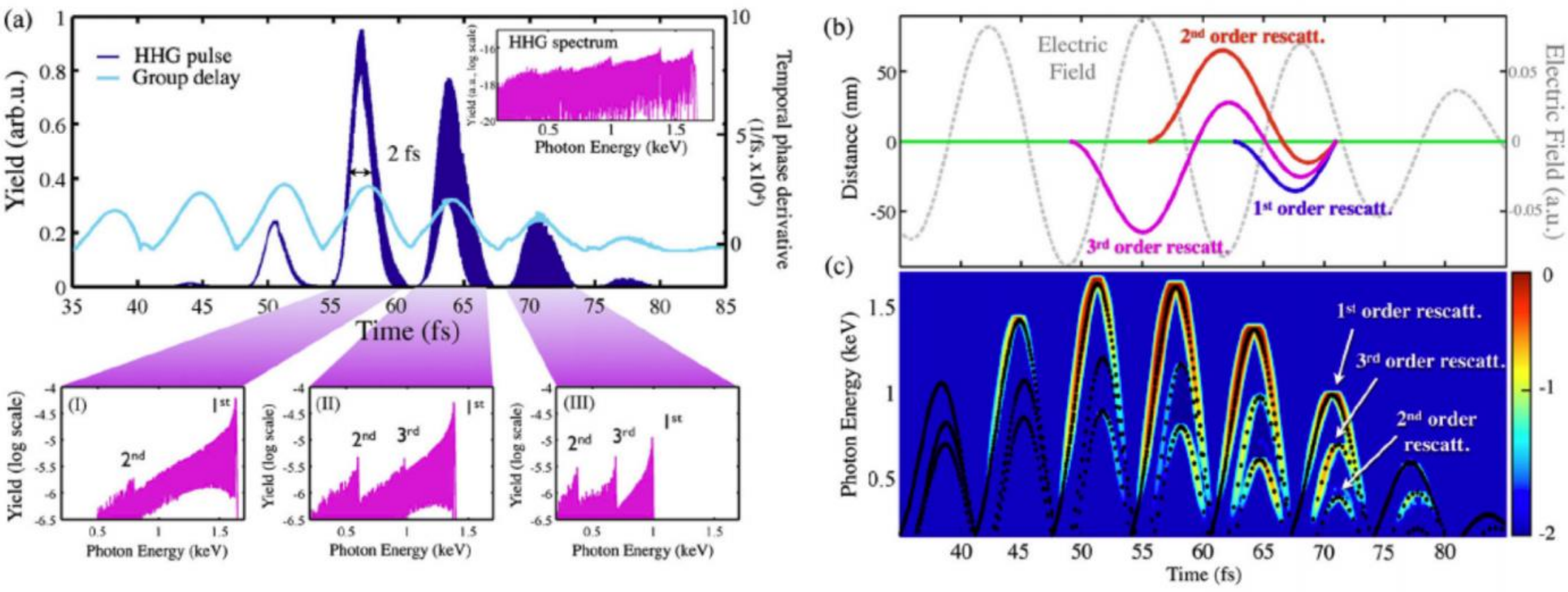
T. Popmintchev et al., Science 336, 1286 (2012)

Experimental spectrum: should support a Fourier-transform limited 2.5 as pulse

Calculations:
without compensation
of chirp it is a 1.2 fs pulse

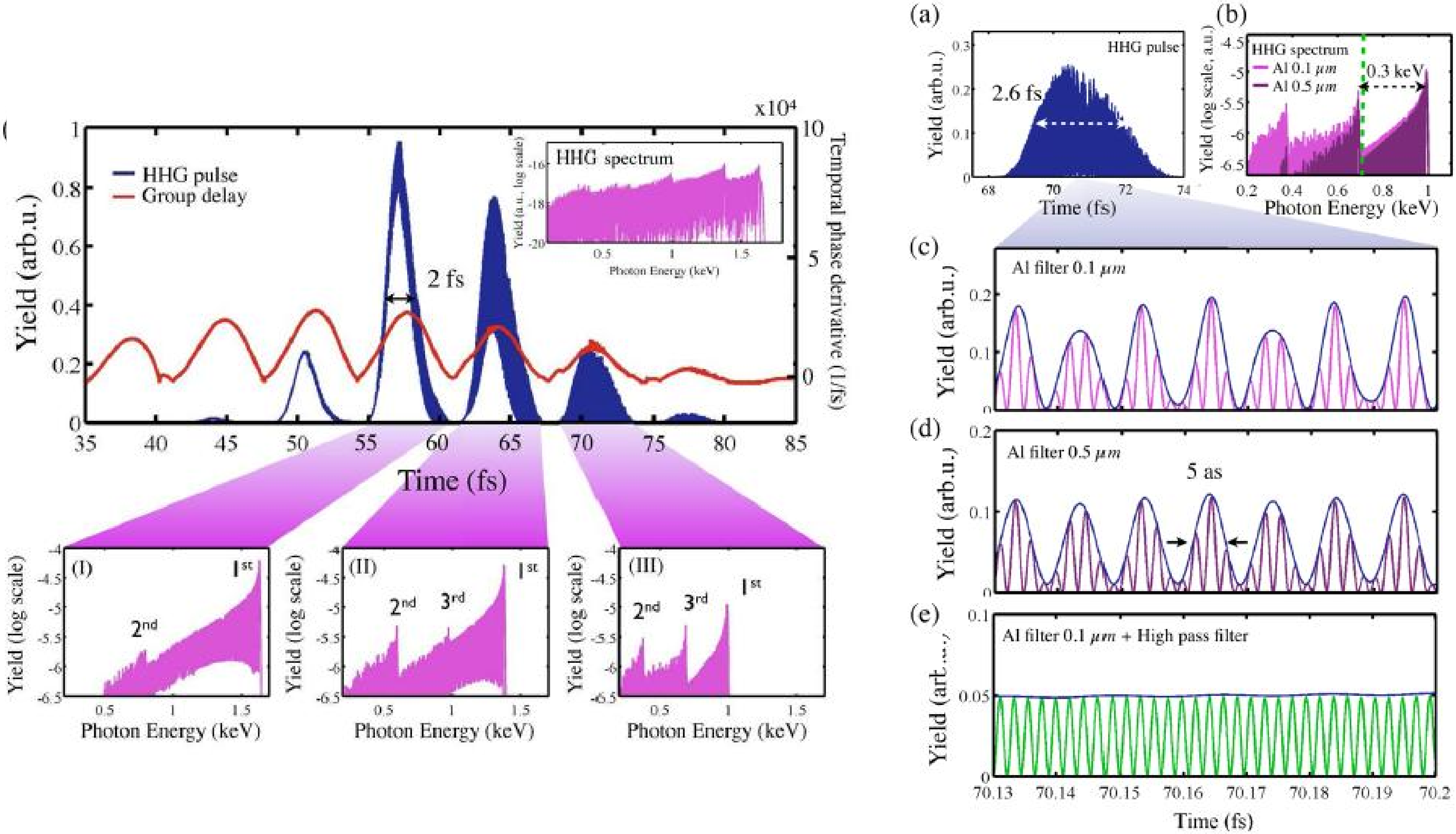
Driving laser pulse:
3.9 μm, 3.3x10[14] W/cm[2]
He gas

Multiple rescatterings



C. Hernandez-Garcia et al., Phys. Rev. Lett. 111, 033002 (2013)

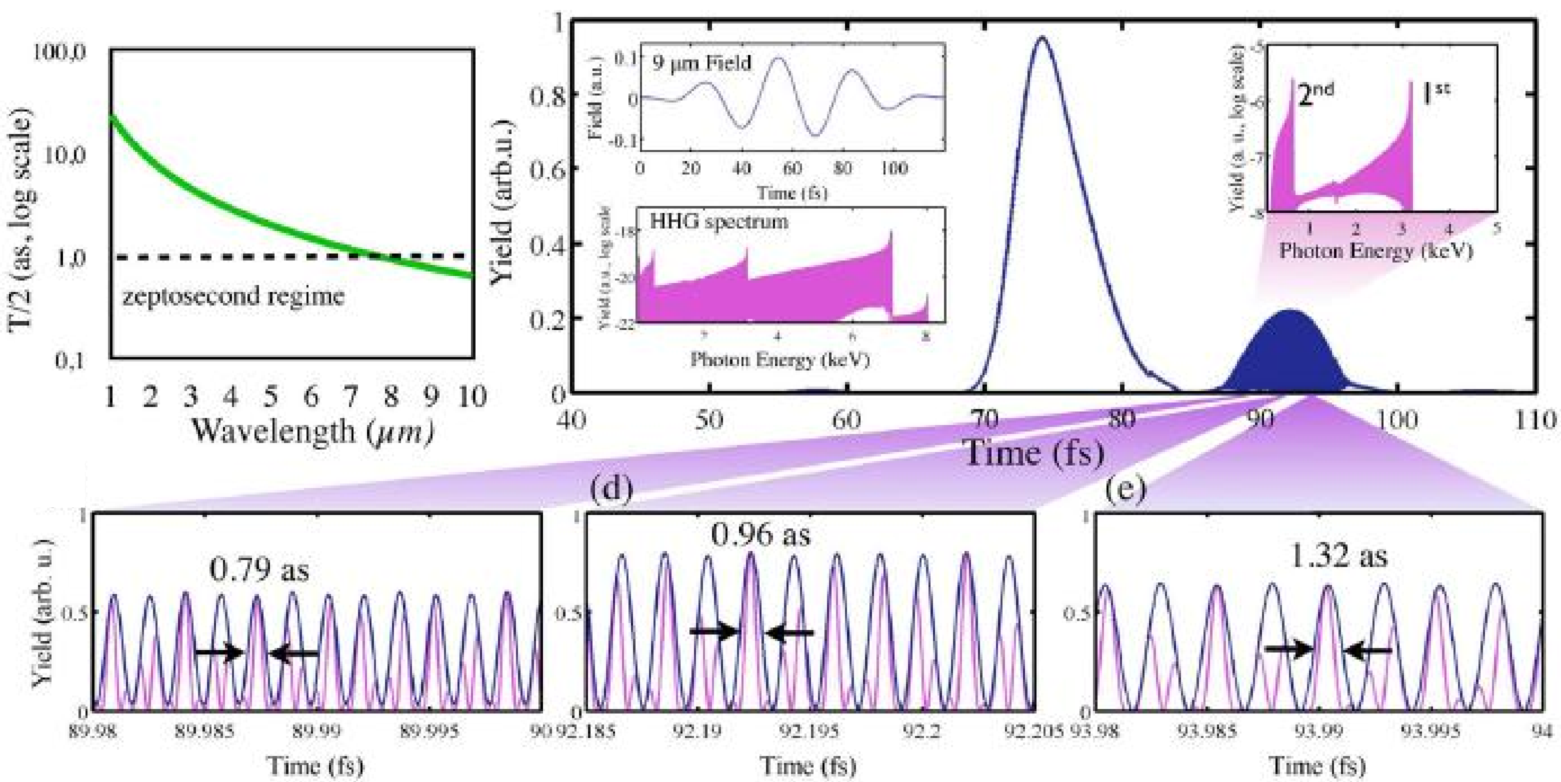
Ultrashort waveforms



HHG using midinfrared driver laser leads to fs-pulse train consisting of ultrashort waveforms due to interference from rescatterings

C. Hernandez-Garcia et al., Phys. Rev. Lett. 111, 033002 (2013)

Towards zeptosecond waveforms



Theoretical prediction: Alternative route for breaking the attosecond barrier

C. Hernandez-Garcia et al., Phys. Rev. Lett. 111, 033002 (2013)