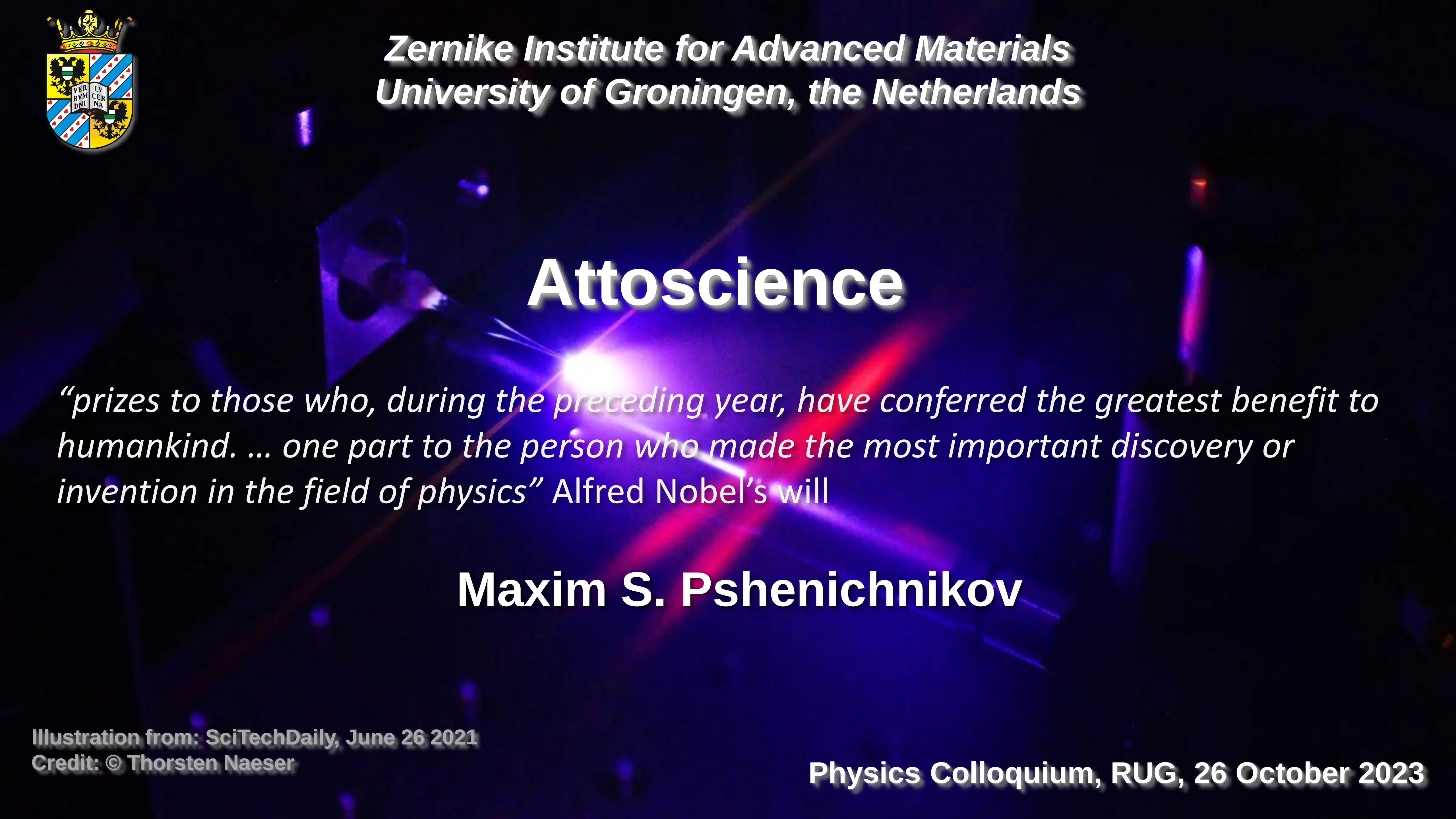
Zernike Institute for Advanced Materials
University of Groningen, the Netherlands



Attoscience

“prizes to those who, during the preceding year, have conferred the greatest benefit to humankind. … one part to the person who made the most important discovery or invention in the field of physics” Alfred Nobel’s will

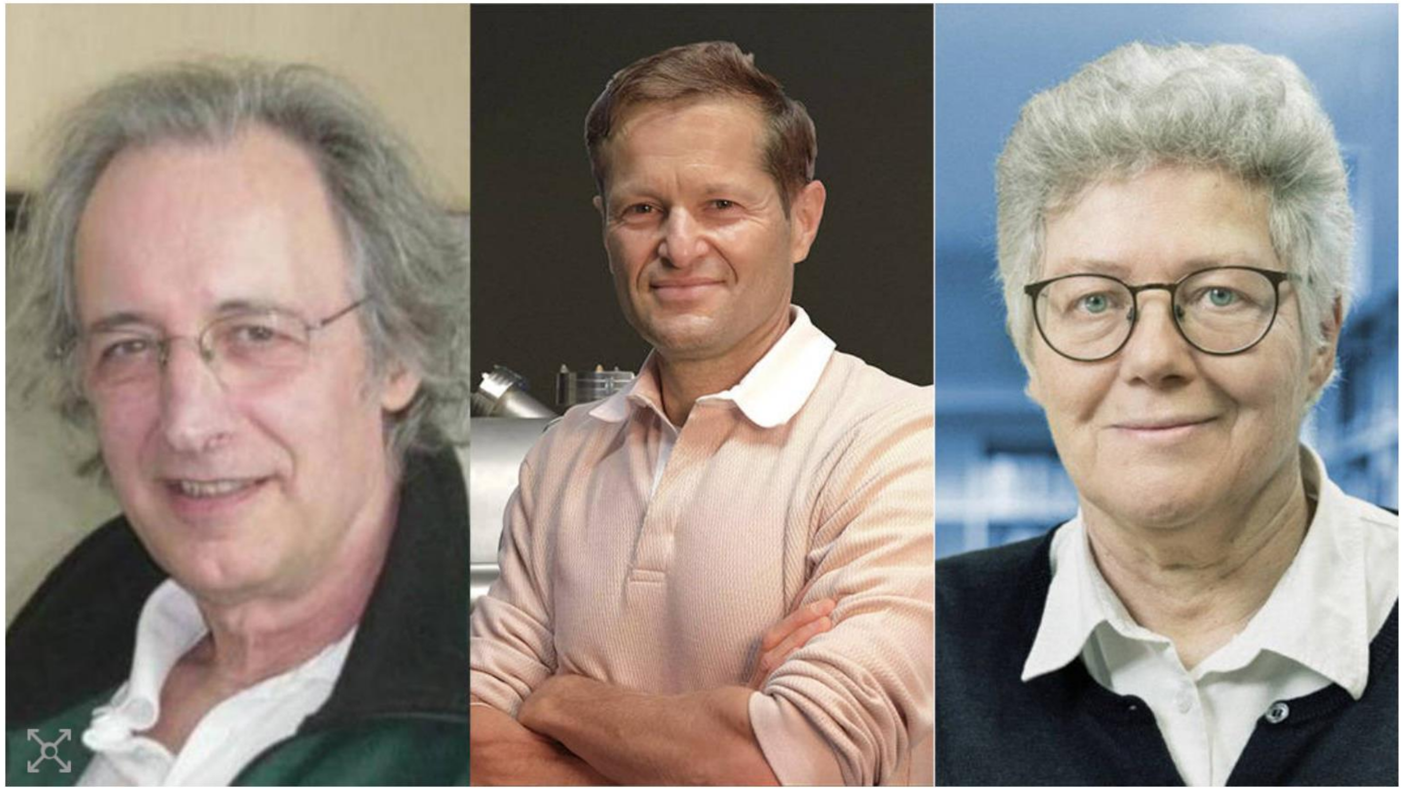
Maxim S. Pshenichnikov

Illustration from: SciTechDaily, June 26 2021
Credit: © Thorsten Naeser

Physics Colloquium, RUG, 26 October 2023

Nobel Prize in Physics 2023

For “experimental methods that generate attosecond pulses of light for the study of electron dynamics in matter”



Pierre Agostini

Ohio State University
USA

Ferenc Krausz

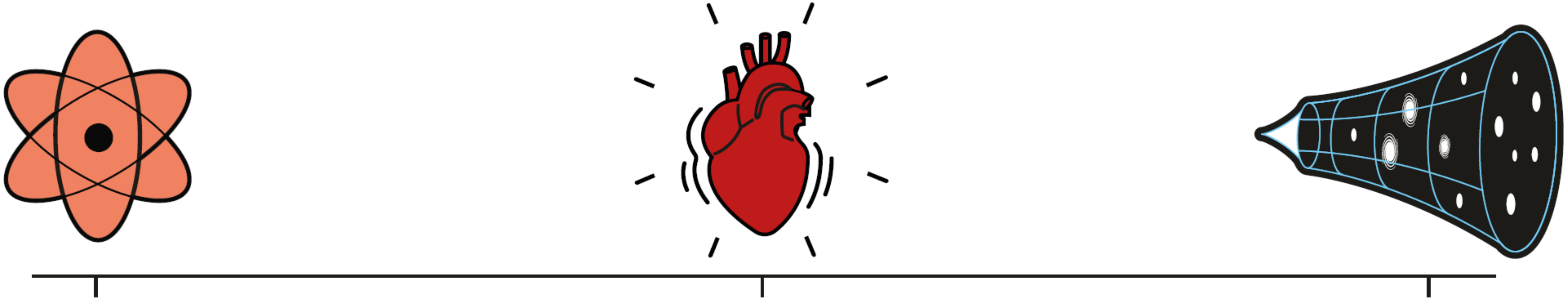
Max Planck Institute of Quantum Optics University of Munich, Germany

Anne L'Huillier

Lund University, Sweeden

What is “Attosecond” ?

1 as = 0. 000 000 000 000 000 001 s = $10^{-18}\,s$



ATTOSECOND

1/1,000,000,000,000,000,000
SECOND

HEARTBEAT

1 SECOND

AGE OF THE UNIVERSE

1,000,000,000,000,000,000
SECONDS

1 as in free space = $10^{-18}\,s \cdot 3 \times 10^{8}\,m\,s^{-1} = 0.1\text{ nm}$

Energy 1000 eV -> wavelength 1.24 nm -> opportunities for imaging

Quantum Beats

Coherent superposition of **1s** and **2p** states of H-atom (aka electronic wavepacket)

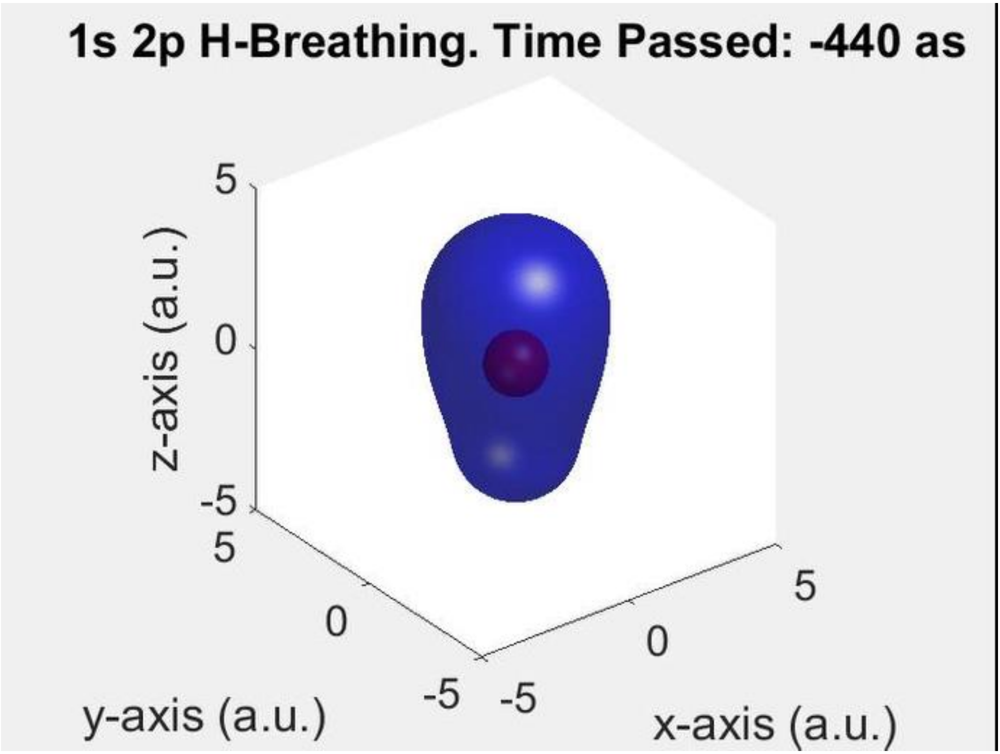
$$|\psi(\boldsymbol{r},t)\rangle = c_1 e^{-i\mathcal{E}_{1s}t/\hbar}|u_{1s}(\boldsymbol{r})\rangle + c_2 e^{-i\mathcal{E}_{2p}t/\hbar}|u_{2p}(\boldsymbol{r})\rangle$$

$$|\psi(\boldsymbol{r},t)|^2 = |c_1|^2|u_{1s}|^2 + |c_2|^2|u_{2p}|^2 + 2Re[c_1^* c_2 u_{1s}^* u_{2p}\, e^{-i(\mathcal{E}_{2p}-\mathcal{E}_{1s})t/\hbar}]$$

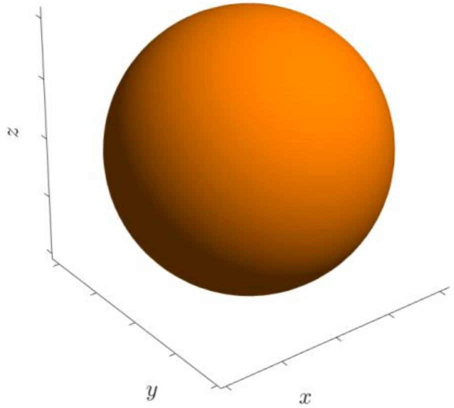
- oscillation with period $T = h/|\mathcal{E}_{2p} - \mathcal{E}_{1s}| = h/\Delta\mathcal{E}$
- the larger $\Delta\mathcal{E}$, the faster the motion in the superposition state

$\Delta\mathcal{E} = 10.2$ eV $T \approx 400$ as -> superfast!

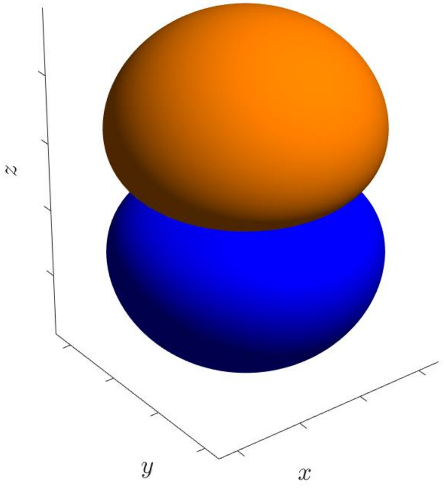
$|\psi(\boldsymbol{r},t)|^2$



1s state



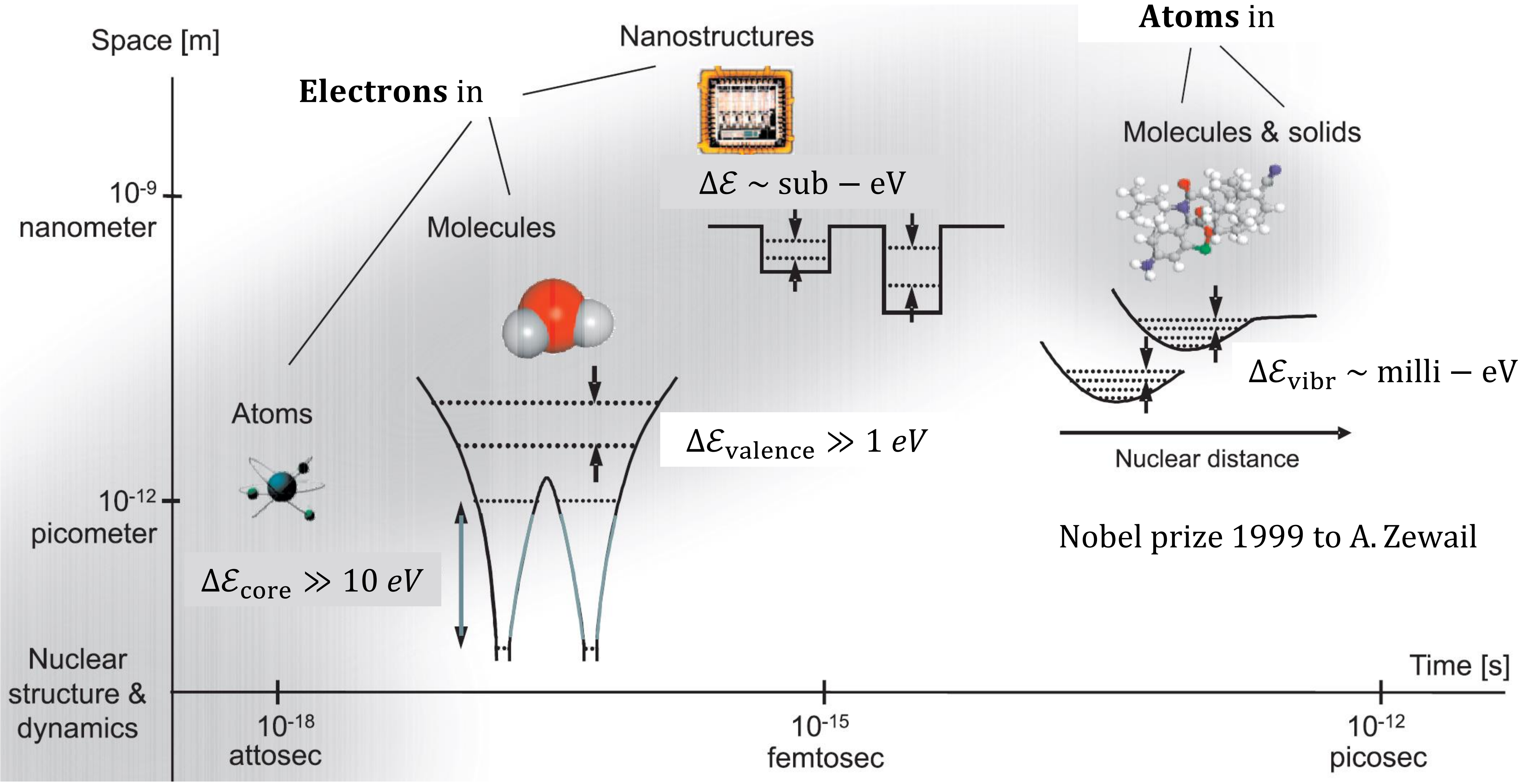
2p state



Attosecond pulses provide access to extremely fast processes at the atomic-size scale

Courtesy of Prof. Mauro Nisoli, Politecnico di Milano, Italy

Characteristic length and time scales for structure and dynamics



Electrons in

Atoms in

$\Delta\mathcal{E} \sim \mathrm{sub-eV}$

$\Delta\mathcal{E}_{\mathrm{vibr}} \sim \mathrm{milli-eV}$

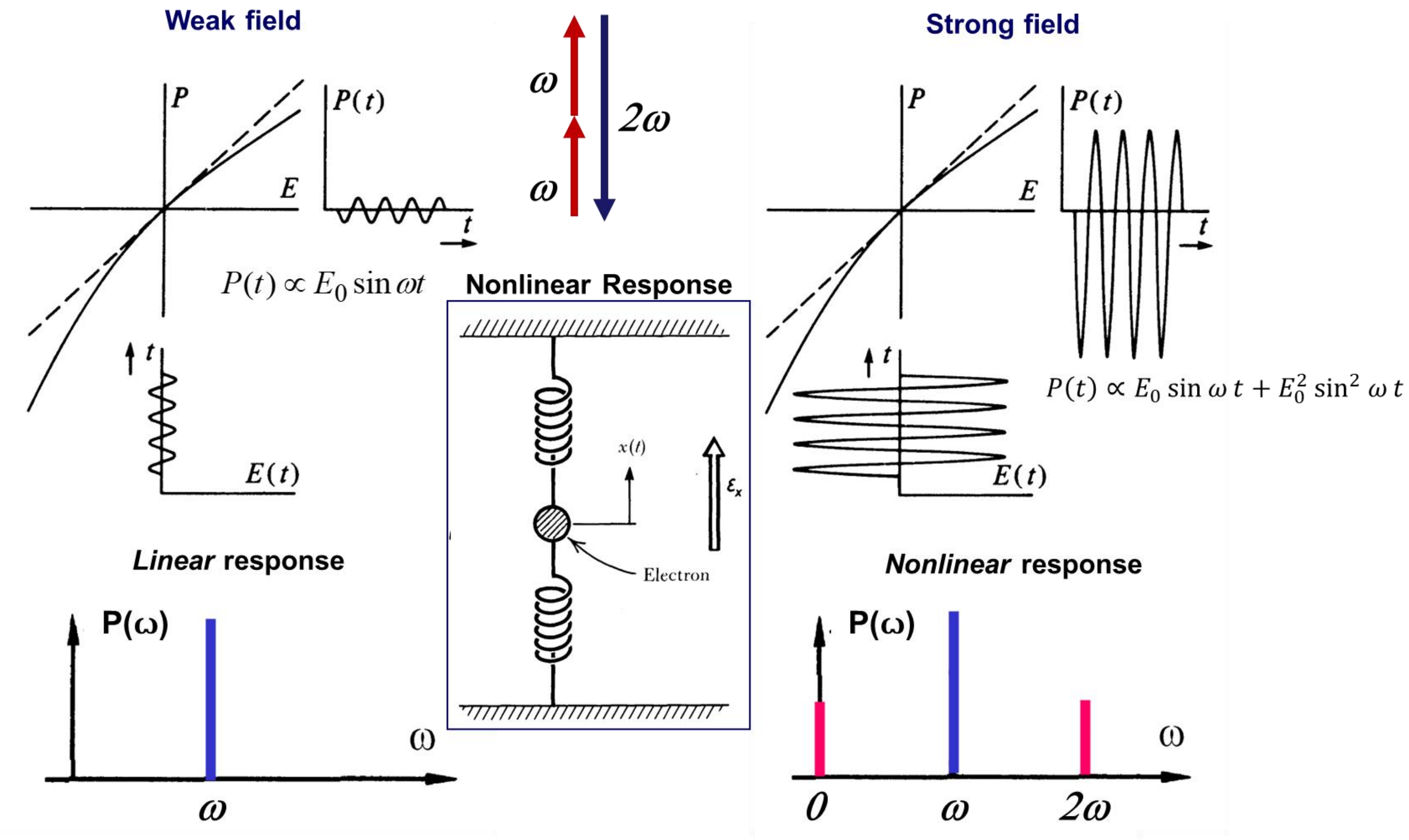
$\Delta\mathcal{E}_{\mathrm{valence}} \gg 1\ eV$

$\Delta\mathcal{E}_{\mathrm{core}} \gg 10\ eV$

Nobel prize 1999 to A. Zewail

Krausz&Ivanov. Rev. Mod. Phys. 81, 2009

Perturbative Approach to Nonlinear Frequency Conversion



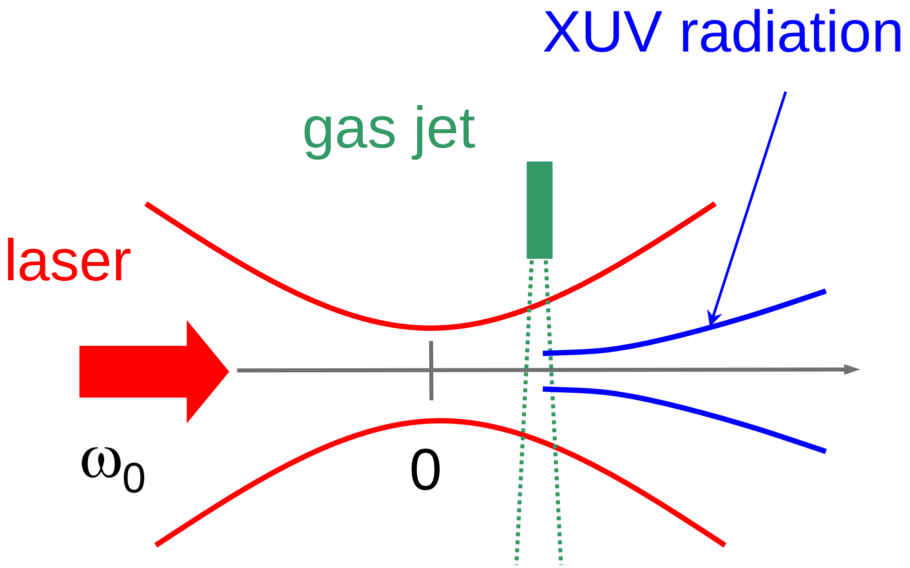
$$P = \epsilon_0\chi^{(1)}E + \epsilon_0\chi^{(2)}E^2 + \epsilon_0\chi^{(3)}E^3 + \cdots = P^{(1)} + P^{(2)} + P^{(3)} + \cdots = P + P^{NL}$$

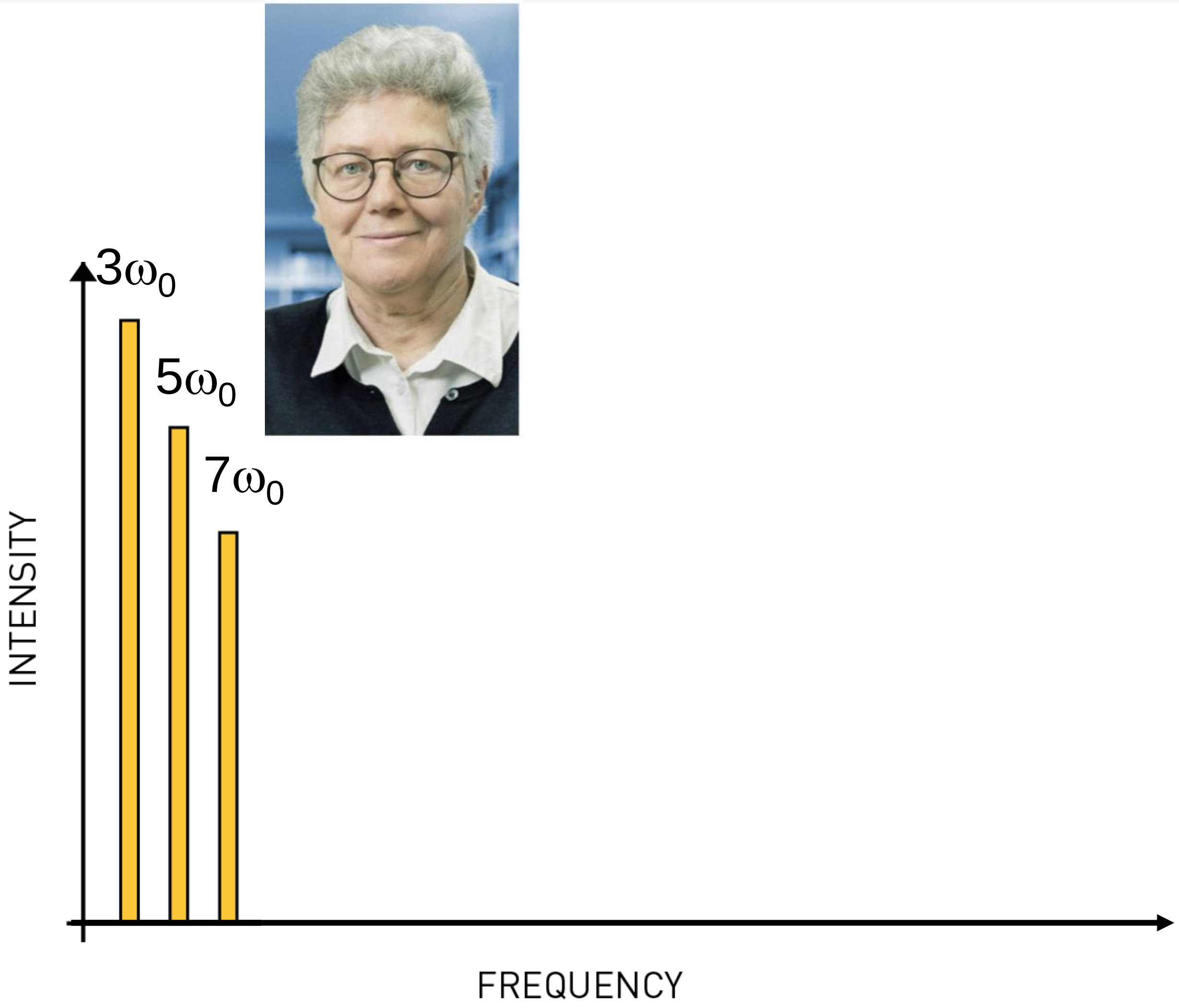
Linear polarization

Noninear polarization

High harmonic generation (HGG)

An intense $10^{13}\ \mathrm{W/cm^2}$ ultrashort light pulse focused on a gas jet of neon or argon





Ferray, L'Huillier *et al.*, J. Phys. B **21**, L31 (1988)
McPherson *et al.*, J. Opt. Soc. Am. B **4,** 595 (1987)

High harmonic generation (HGG)

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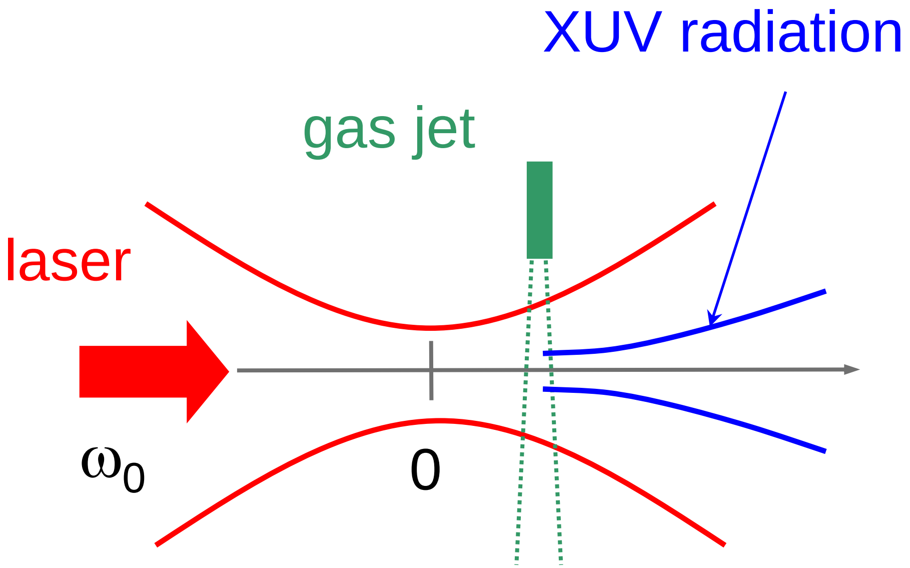
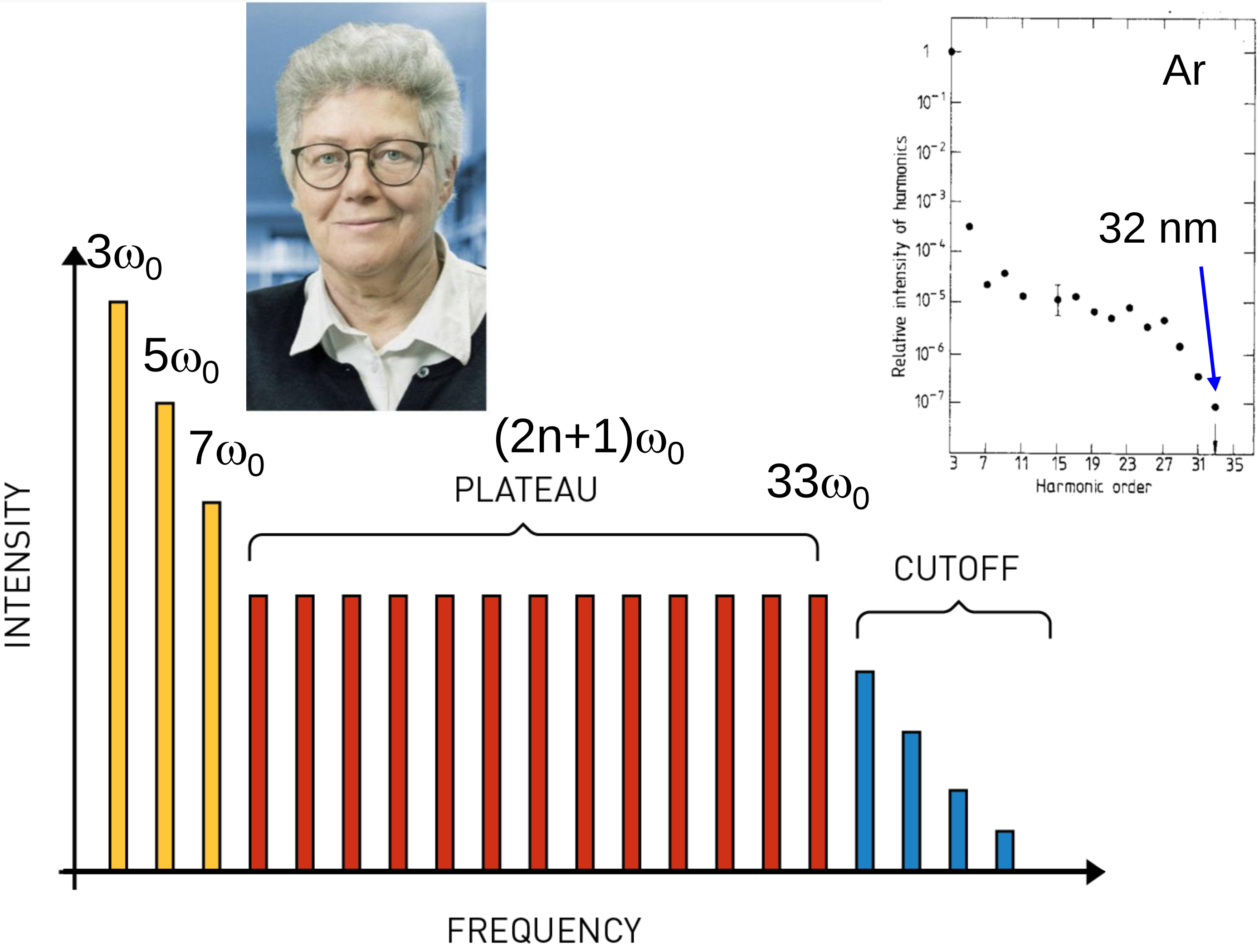
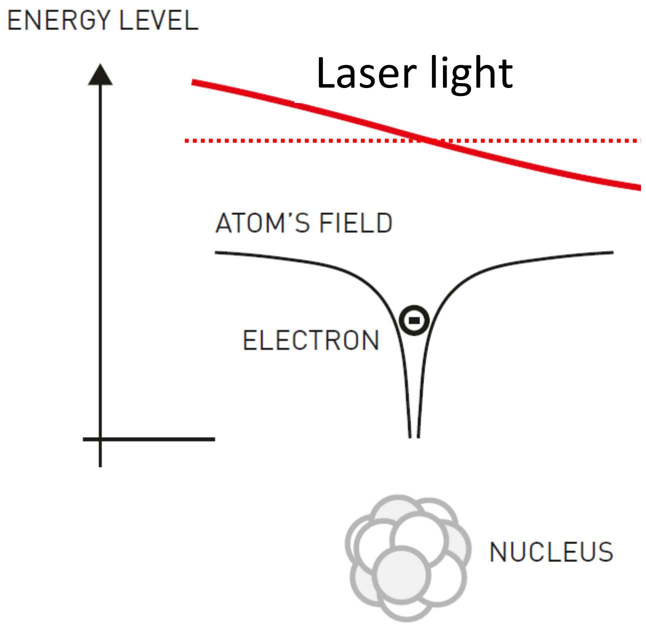




Odd harmonics of the visible light are generated up to the XUV region

Ferray, L'Huillier *et al*., J. Phys. B **21**, L31 (1988)
McPherson *et al*., J. Opt. Soc. Am. B **4,** 595 (1987)

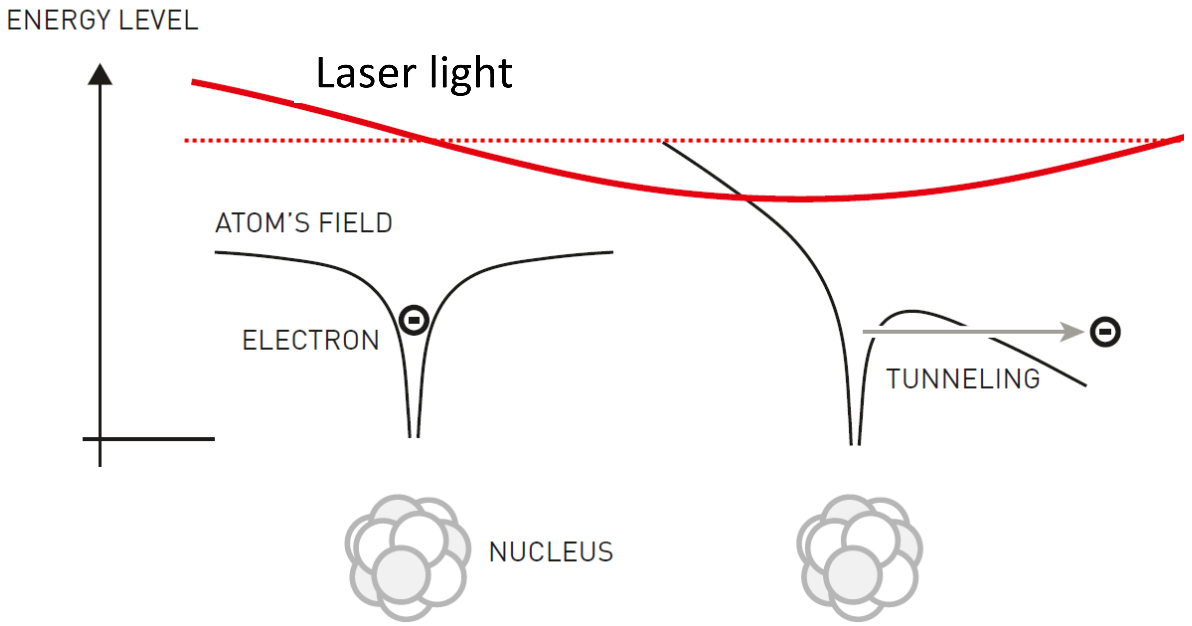
The rescattering semiclassical model of HHG (1990's)



0. The laser field is zero.
An electron is bound to
an atom's nucleus

Keldysh, Sov. Phys. JETP 20, 1307 (1965)
K.J. Schafer *et al.*, Phys. Rev. Lett. **70**, 1599 (1993)
P.B. Corkum, Phys. Rev. Lett., **71**, 1994 (1993)

The rescattering semiclassical model of HHG (1990’s)



0. The laser field is zero. An electron is bound to an atom’s nucleus

1. The nucleus field is distorted by the laser pulse. The electron **tunnels out** and **accelerates** in the laser field out off the nucleus

Keldysh, Sov. Phys. JETP 20, 1307 (1965)
K.J. Schafer *et al.*, Phys. Rev. Lett. **70**, 1599 (1993)
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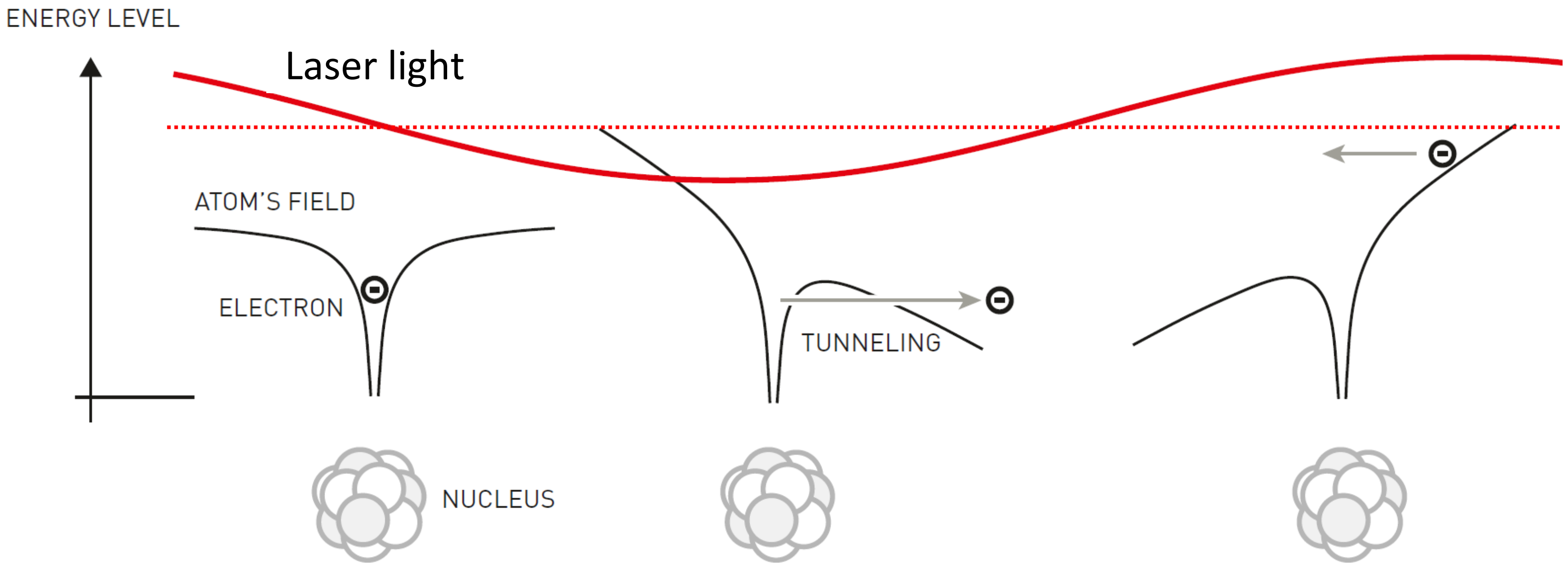

0. The laser field is zero. An electron is bound to an atom’s nucleus

1. The nucleus field is distorted by the laser pulse. The electron **tunnels out** and **accelerates** in the laser field out off the nucleus

2. The laser field changes direction; the electron is **pulled back** to the direction it came from

Keldysh, Sov. Phys. JETP 20, 1307 (1965)
K.J. Schafer *et al.*, Phys. Rev. Lett. **70**, 1599 (1993)
P.B. Corkum, Phys. Rev. Lett., **71**, 1994 (1993)

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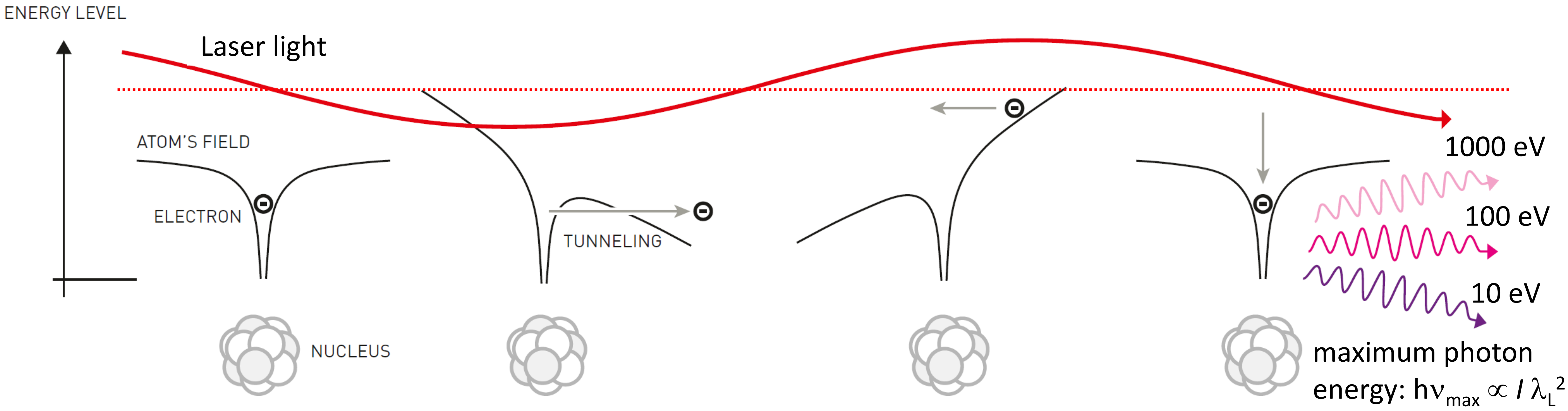


maximum photon energy: $h\nu_{max} \propto I\,\lambda_L^2$

0. The laser field is zero. An electron is bound to an atom’s nucleus

1. The nucleus field is distorted by the laser pulse. The electron **tunnels out** and **accelerates** in the laser field out off the nucleus

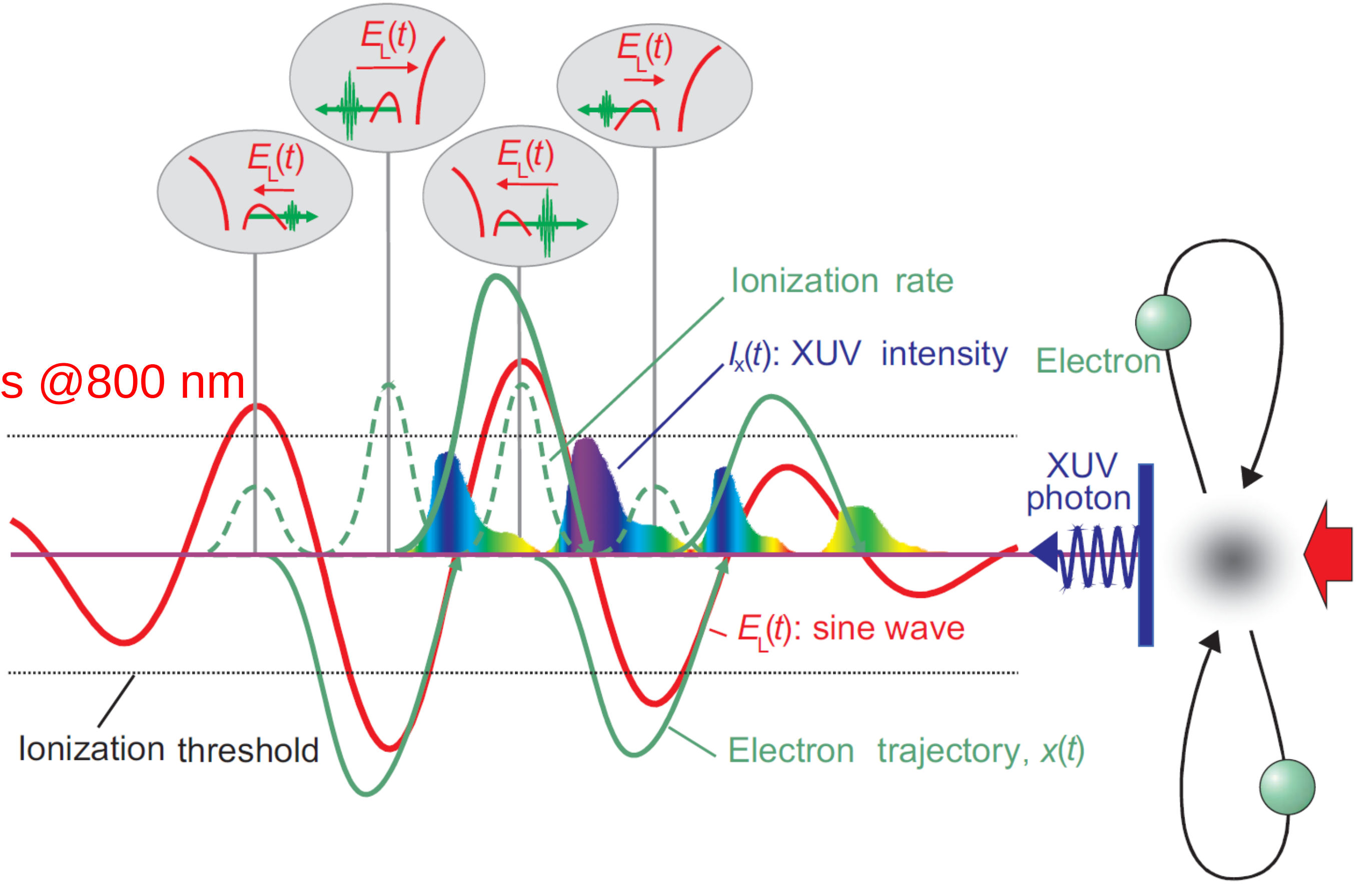
2. The laser field changes direction; the electron is **pulled back** to the direction it came from

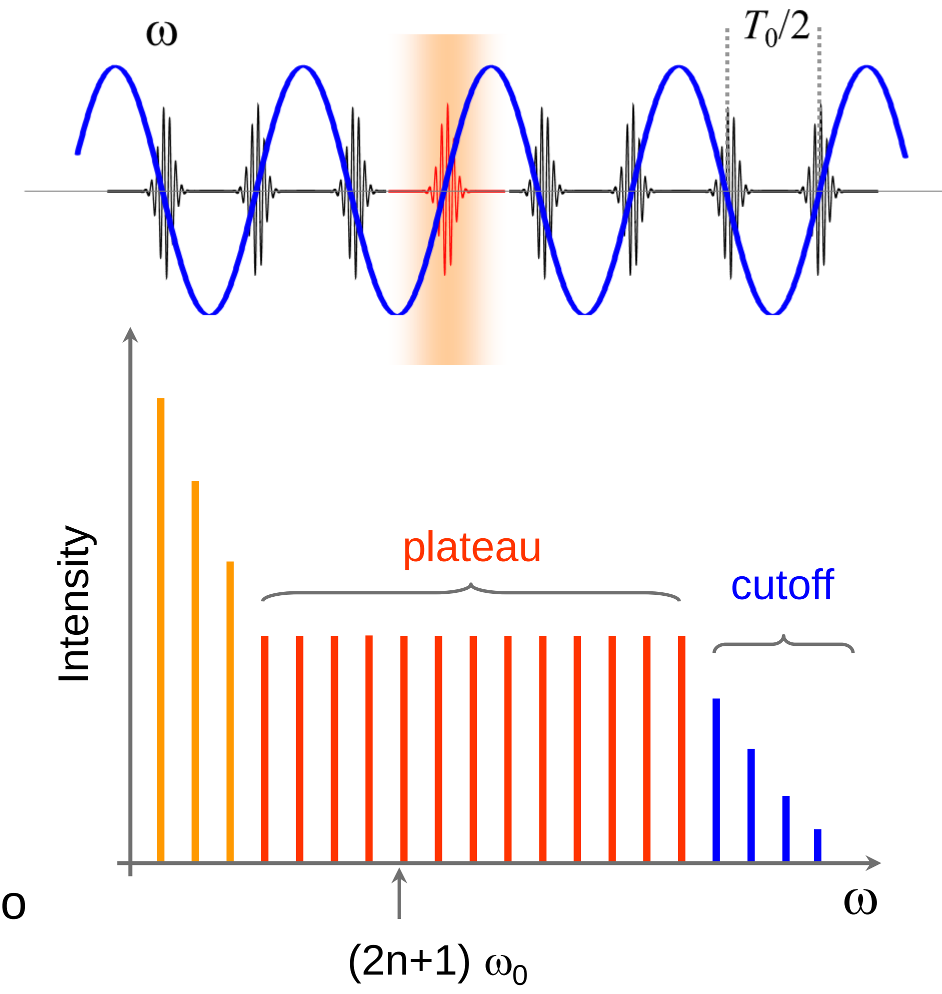
3. To reattach to the atom’s nucleus, the electron must **rid itself of the extra energy**. This is emitted **as an UV flash** which wavelength depends on how far the electron moved

Keldysh, Sov. Phys. JETP 20, 1307 (1965)
K.J. Schafer *et al.*, Phys. Rev. Lett. **70**, 1599 (1993)
P.B. Corkum, Phys. Rev. Lett., **71**, 1994 (1993)

The rescattering semiclassical model of HHG (1990’s)

5 fs @800 nm





$E_{max} = I_p + 3.17\ U_p$

$U_p \propto I\ \lambda_L^2$

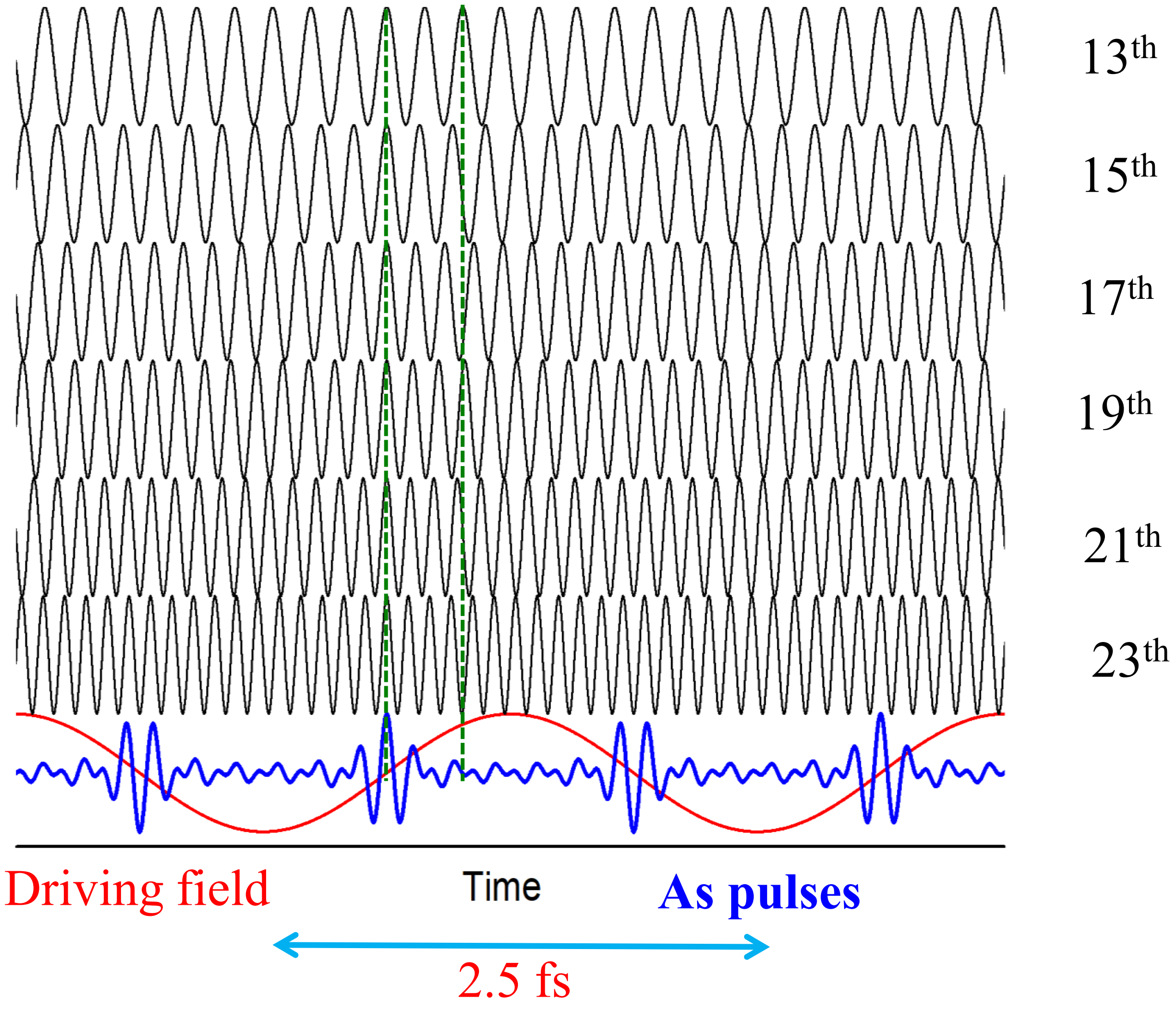
Recollisions occur two times per period T of the driving field which leads to

1. Appearing of a **train** of as pulses with repetition rate of T/2
2. The spectrum of the emitted field is **equidistant** with peaks at $(2n+1)\omega_0$ -> HHG
3. The XUV pulse is perfectly **synchronized** with the driving pùlse

Krausz&Ivanov. Rev. Mod. Phys. 81, 2009

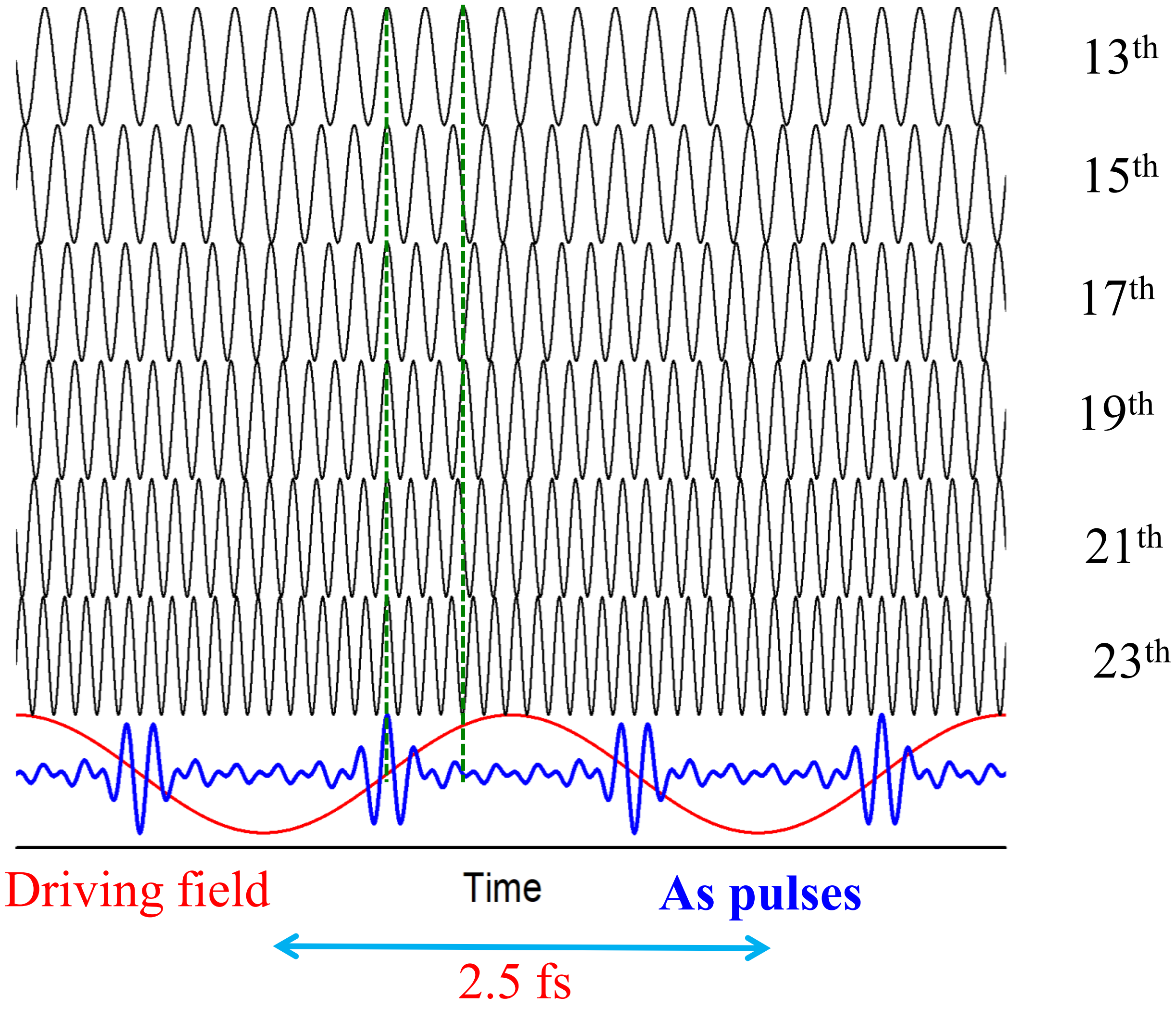
Synthesis of Attosecond Pulses from High Harmonics

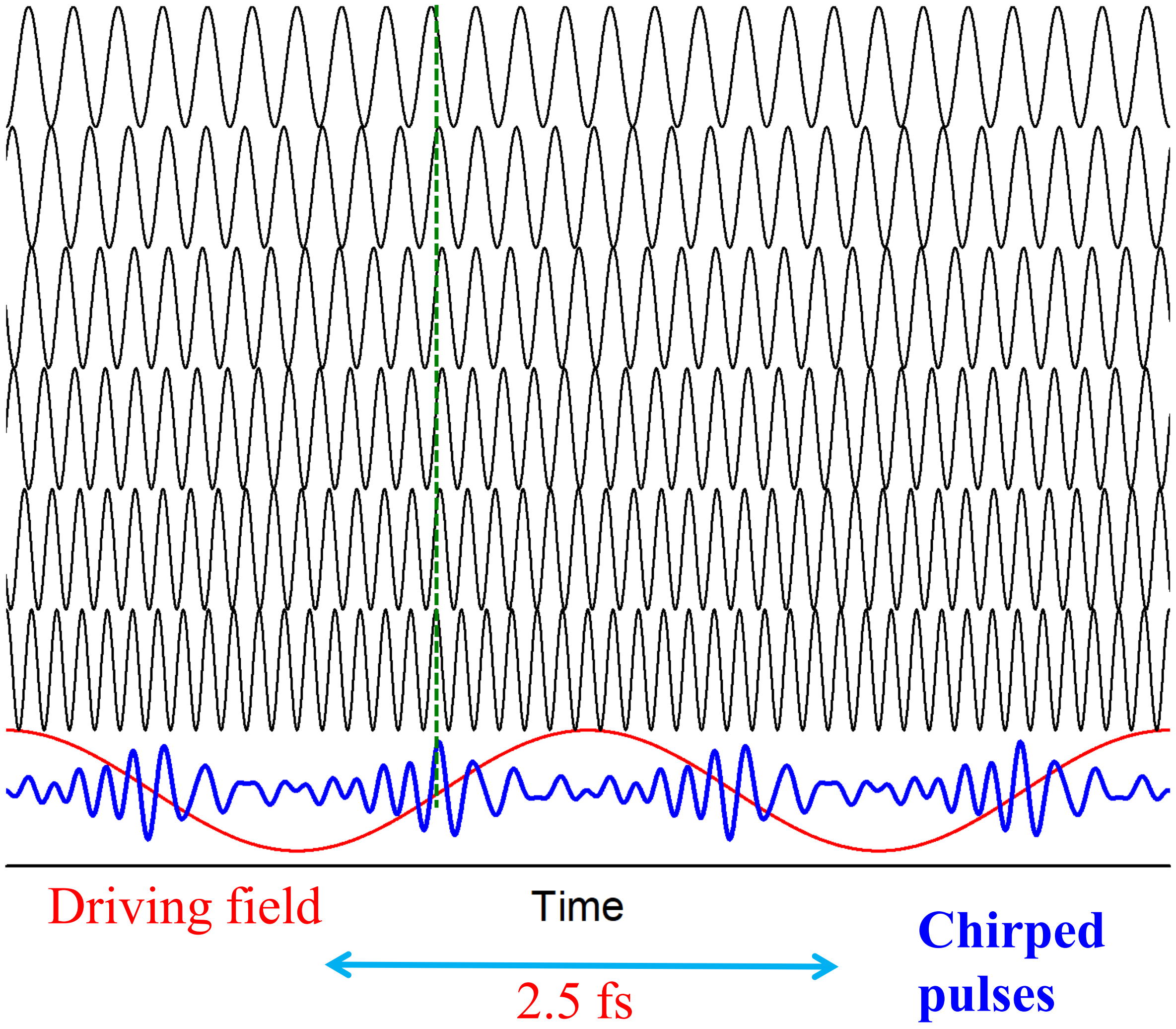
Phases of all harmonics are the same



Gy. Farkas and Cs. Tóth, Physics Letters A168, 447 (1992)

Synthesis of Attosecond Pulses from High Harmonics


Phases of all harmonics are the same

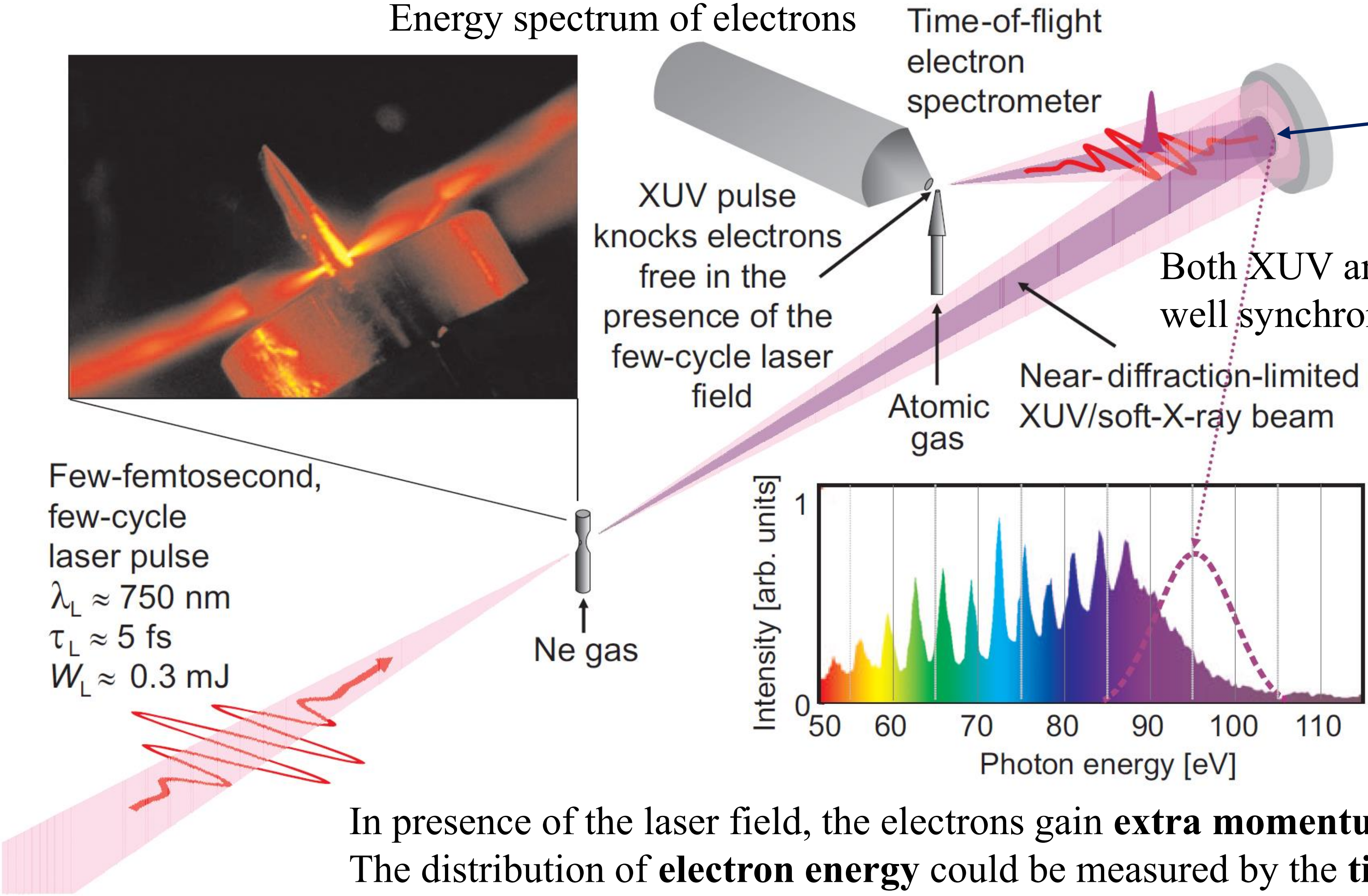

Phases of all harmonics are NOT the same

Bandwidth is not enough; **phase-locking** is needed. But how to prove the pulses are attosecond?

Gy. Farkas and Cs. Tóth, Physics Letters A168, 447 (1992)

How to find out if XUV pulses are really attosecond?

Two-photon, Two-color Photoionization Approach



Movable mirror for XUV radiation only

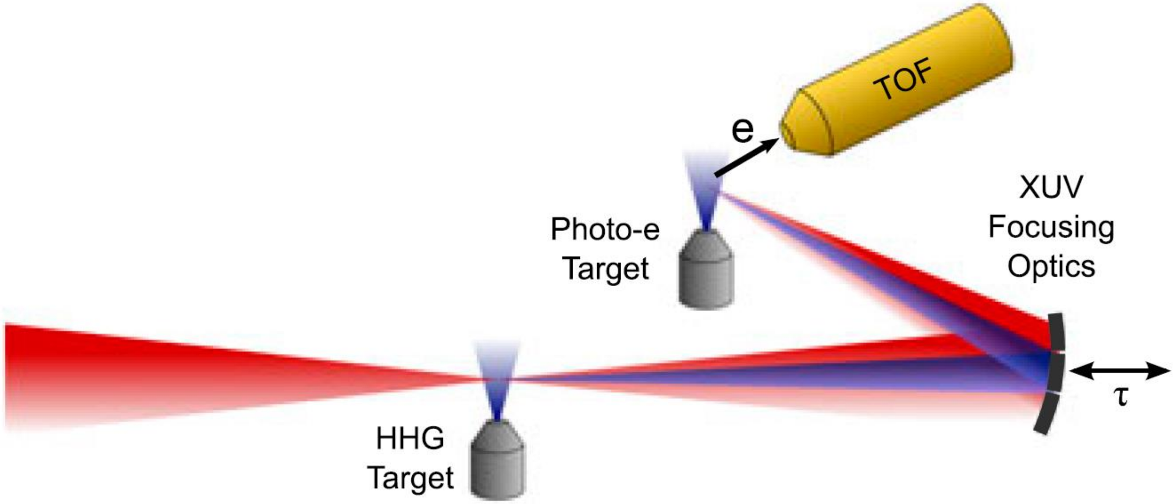
Both XUV and laser pulses are well synchronized in time!

In presence of the laser field, the electrons gain **extra momentum and energy**
The distribution of **electron energy** could be measured by the **time-of-flight technique**

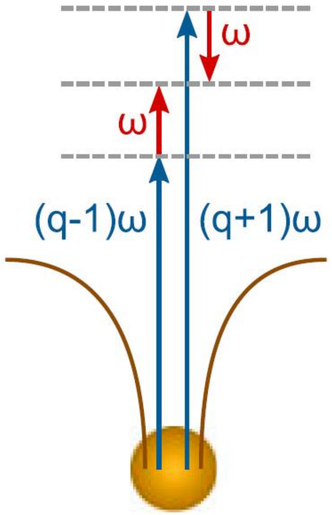
Krausz&Ivanov. Rev. Mod. Phys. 81, 2009

RABBIT Technique: Train of 250-as Pulses

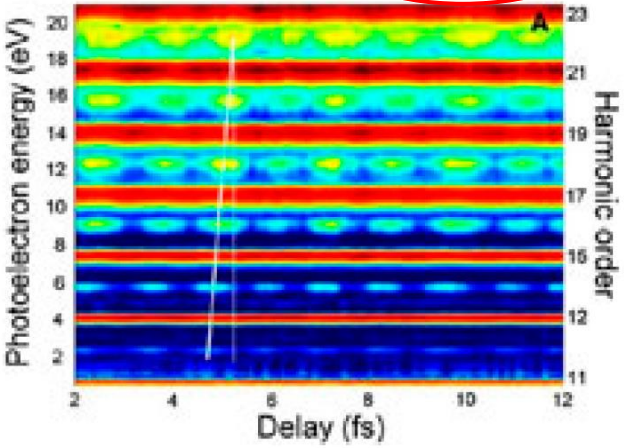
The reconstruction of attosecond beating by interference of two-photon transitions

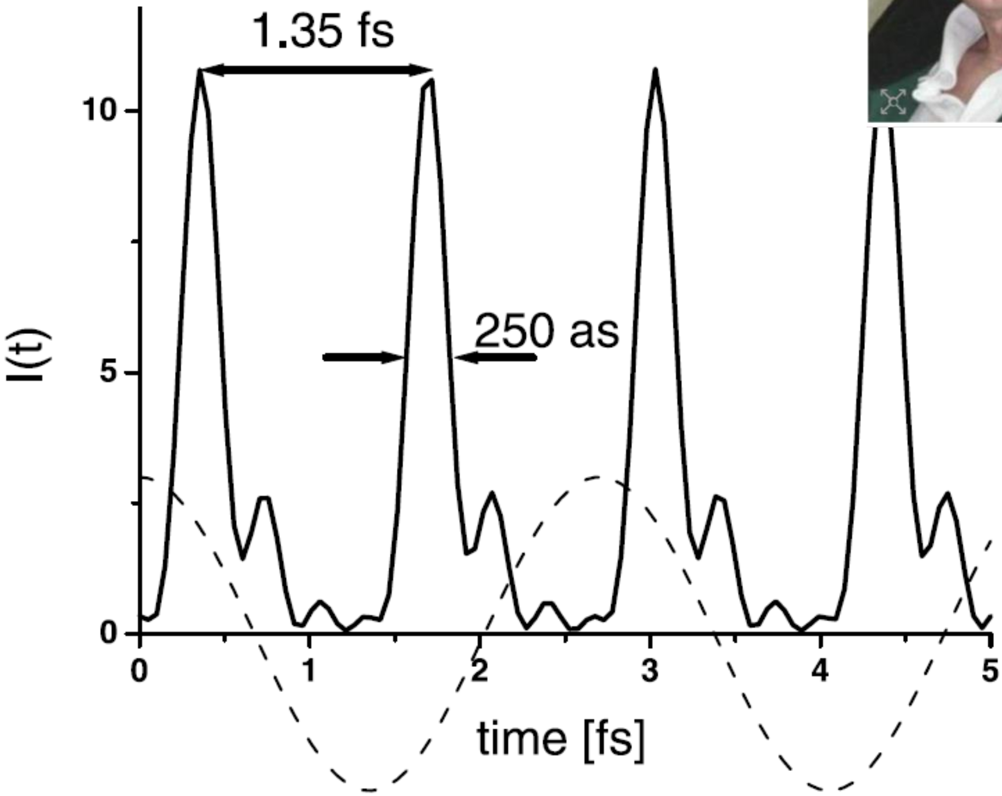


$$I(t) = \left| \sum_q A_q e^{-i\omega_q t + i\phi_q} \right|^2$$



$$S_n(\tau) \propto \cos\left(2\omega_0\tau + \boxed{\phi_{n-1} - \phi_{n+1}} + \Delta\phi_{\mathrm{atomic}}\right)$$

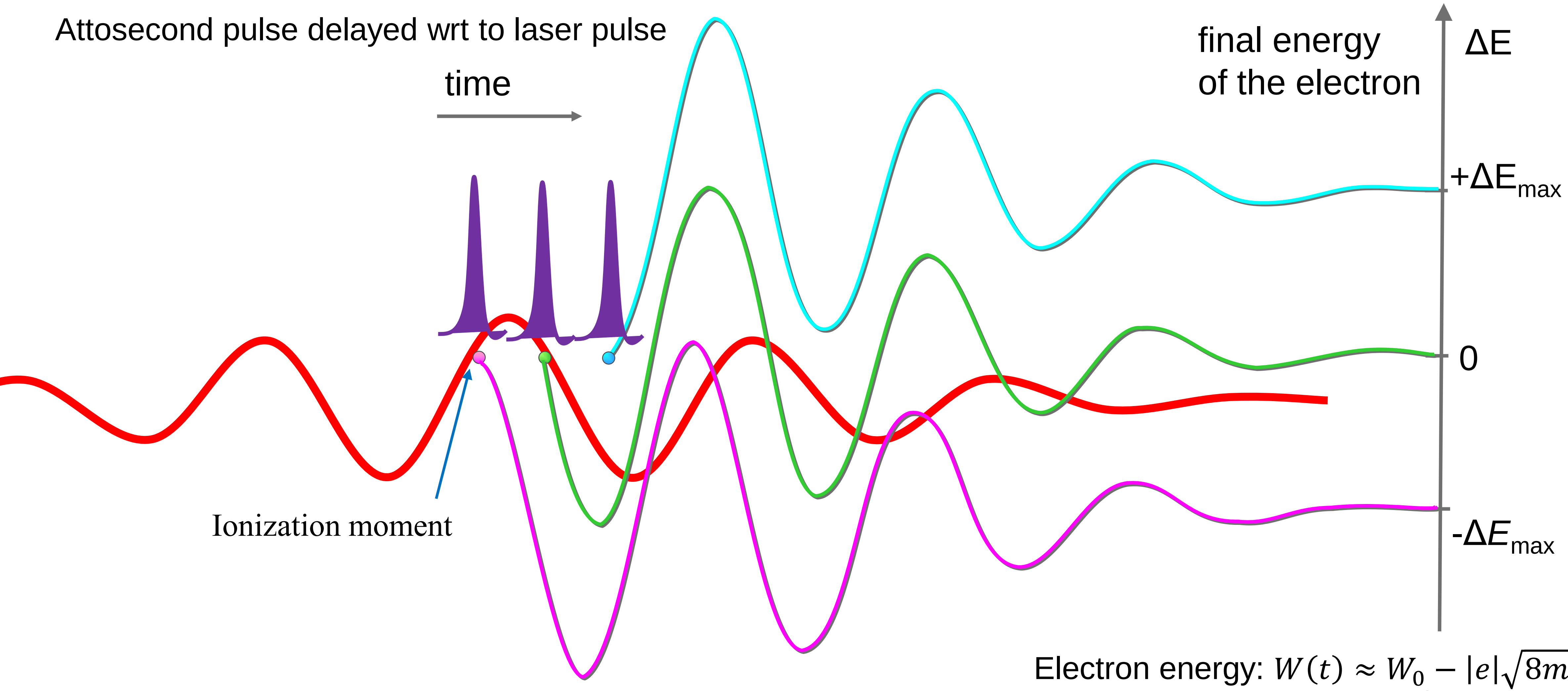




P. Agostini and Co, Science 292, 1689 (2001)

From: Villeneuve, Contemporary Physics 59, 47-61 (2018)

Attosecond Streaking



Attosecond pulse delayed wrt to laser pulse

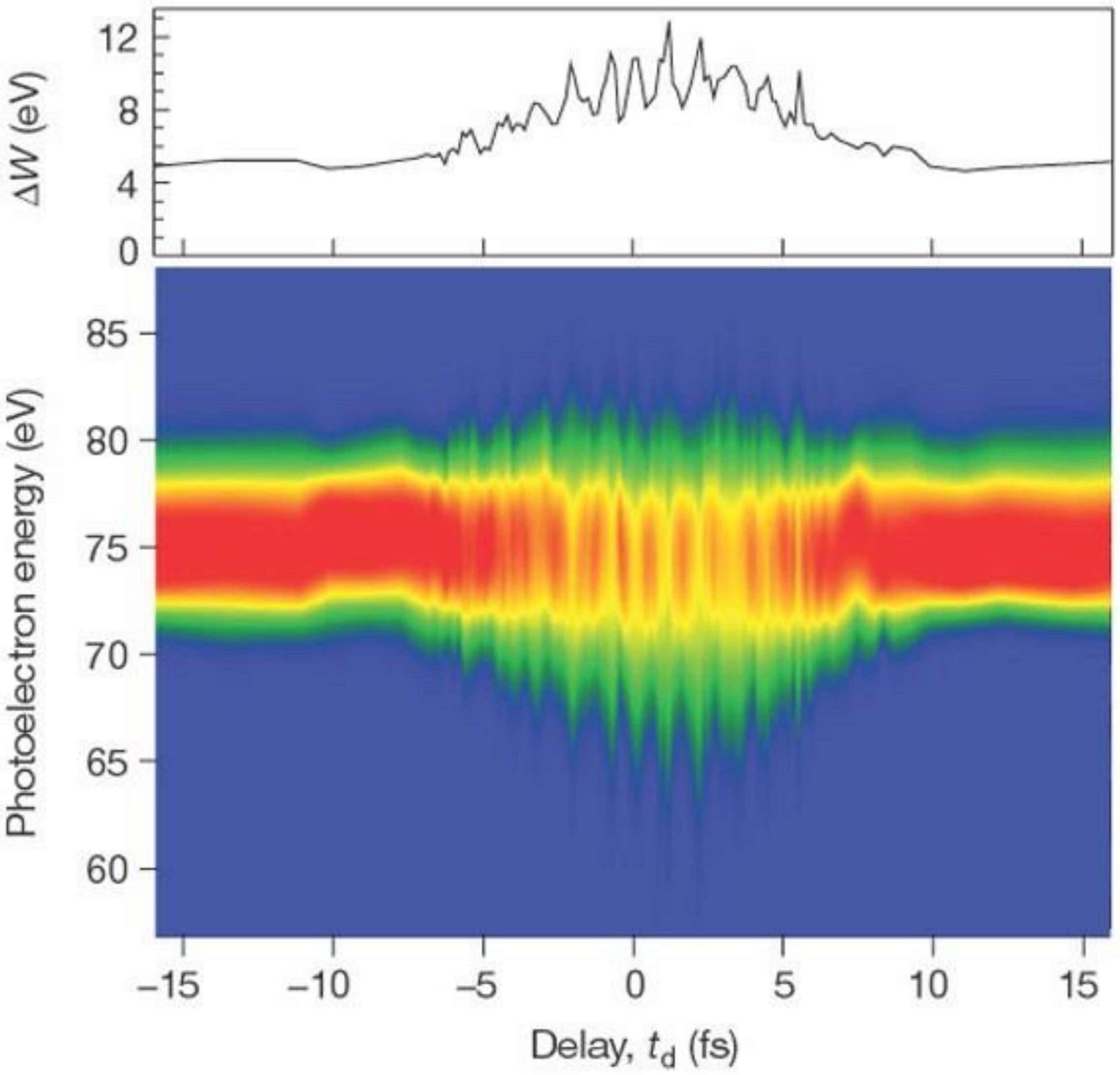
Electron energy: $W(t) \approx W_0 - |e|\sqrt{8mW_0}\,\boldsymbol{A}(t)$

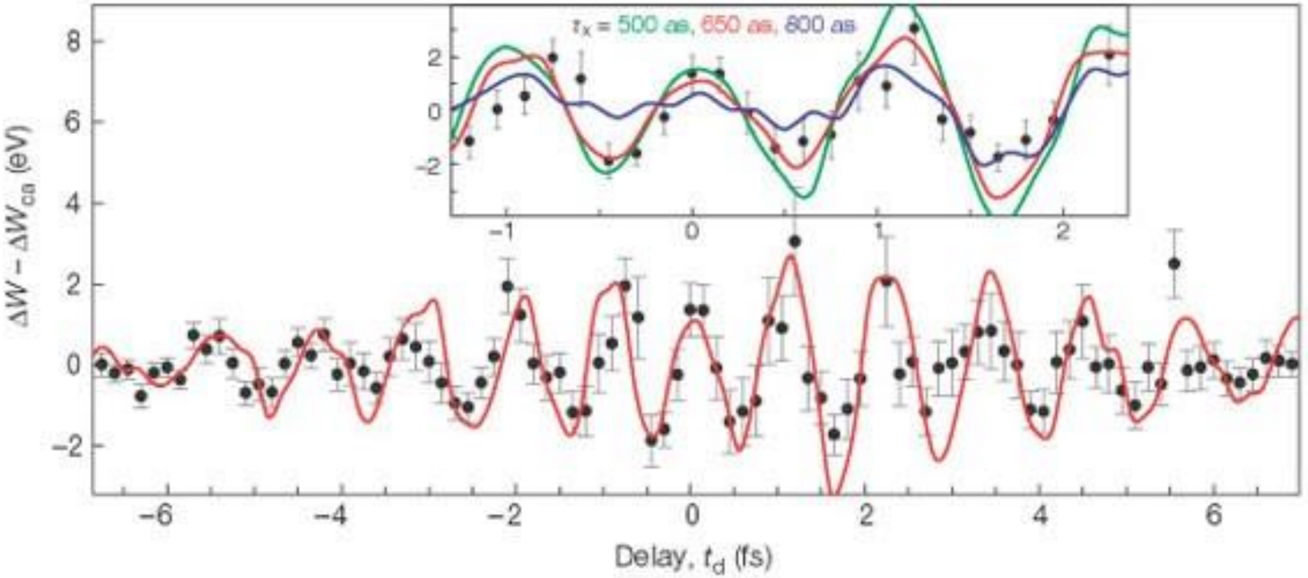
$$W_0 = \hbar\omega_X - I_p; \quad \boldsymbol{A}(t) = \int_t^{+\infty} \boldsymbol{E}_{IR}(t')dt'$$

The energy of photoelectrons depends on the moment they’re produced wrt to the laser field: streaking

courtesy of R. Kienberger, TUM Munich (Germany) and M. Nisoli (Milano)

Single 650 Attosecond Pulse Generated !

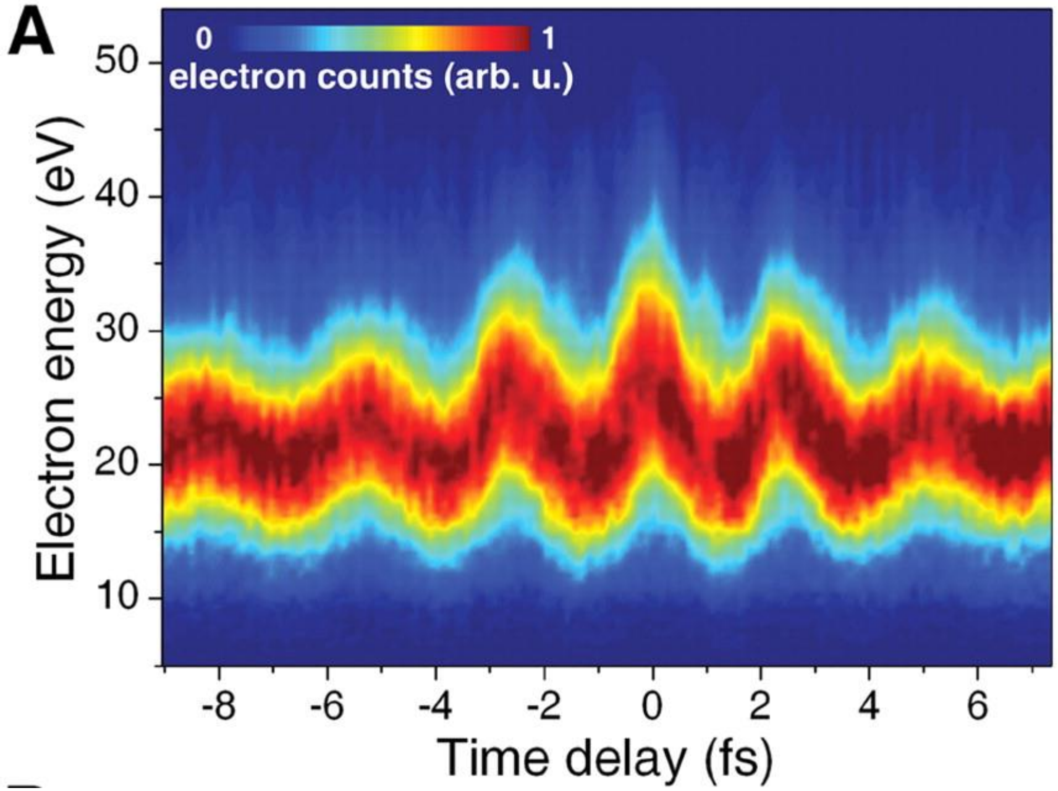




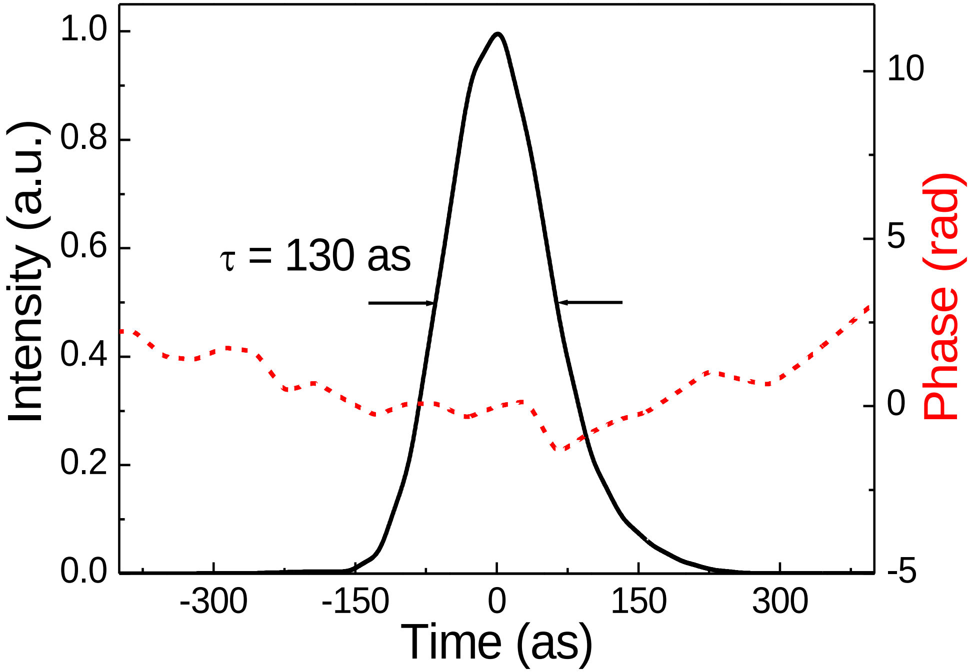
M. Hentschel *et al.*, Nature **414**, 509 (2001)

130 Attosecond Pulse Fully Characterized

Frequency-resolved optical gating for complete reconstruction of attosecond burst



Retrieved Intensity profile and phase

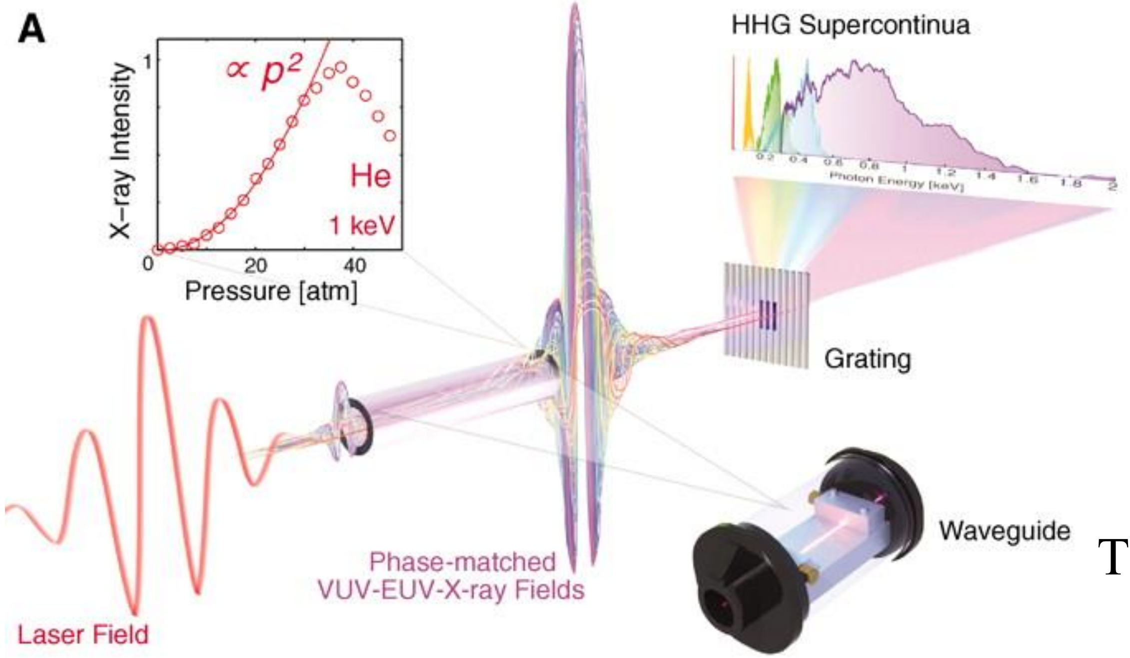


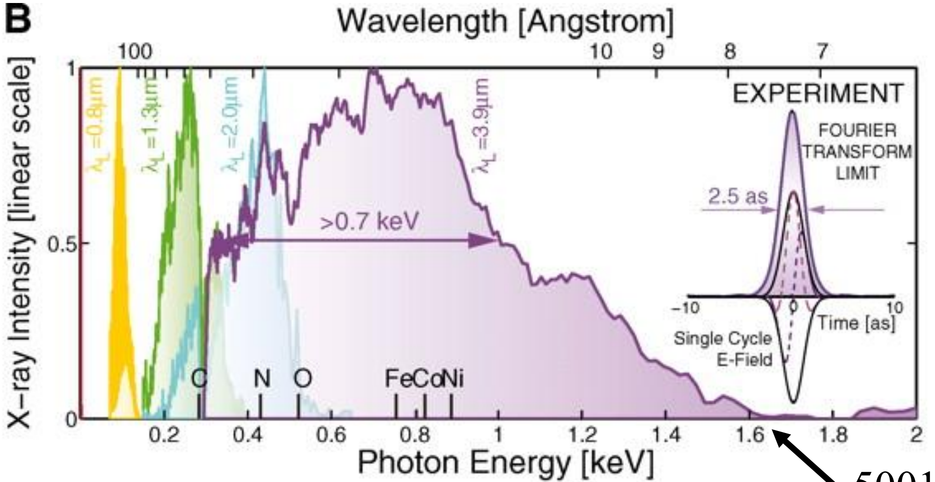
Aluminum foil for dispersion compensation of the XUV pulse

Near-single cycle pulse !

De Silvestri, Nisoli *et al.*, Science **314**, 443 (2006)

5001st Harmonic and 53 as Pulses Generated

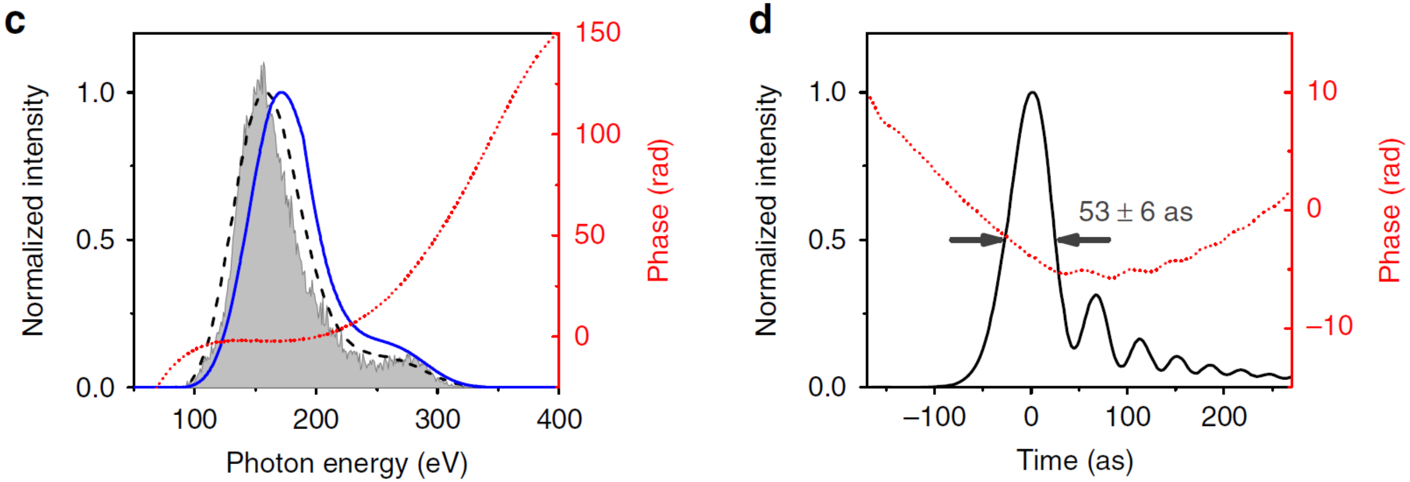




5001st harmonic!

The HHG spectrum extends below 1 nm –> x-ray range

Murnane&Kapteyn group,
SCIENCE 336, 1287 (2012)



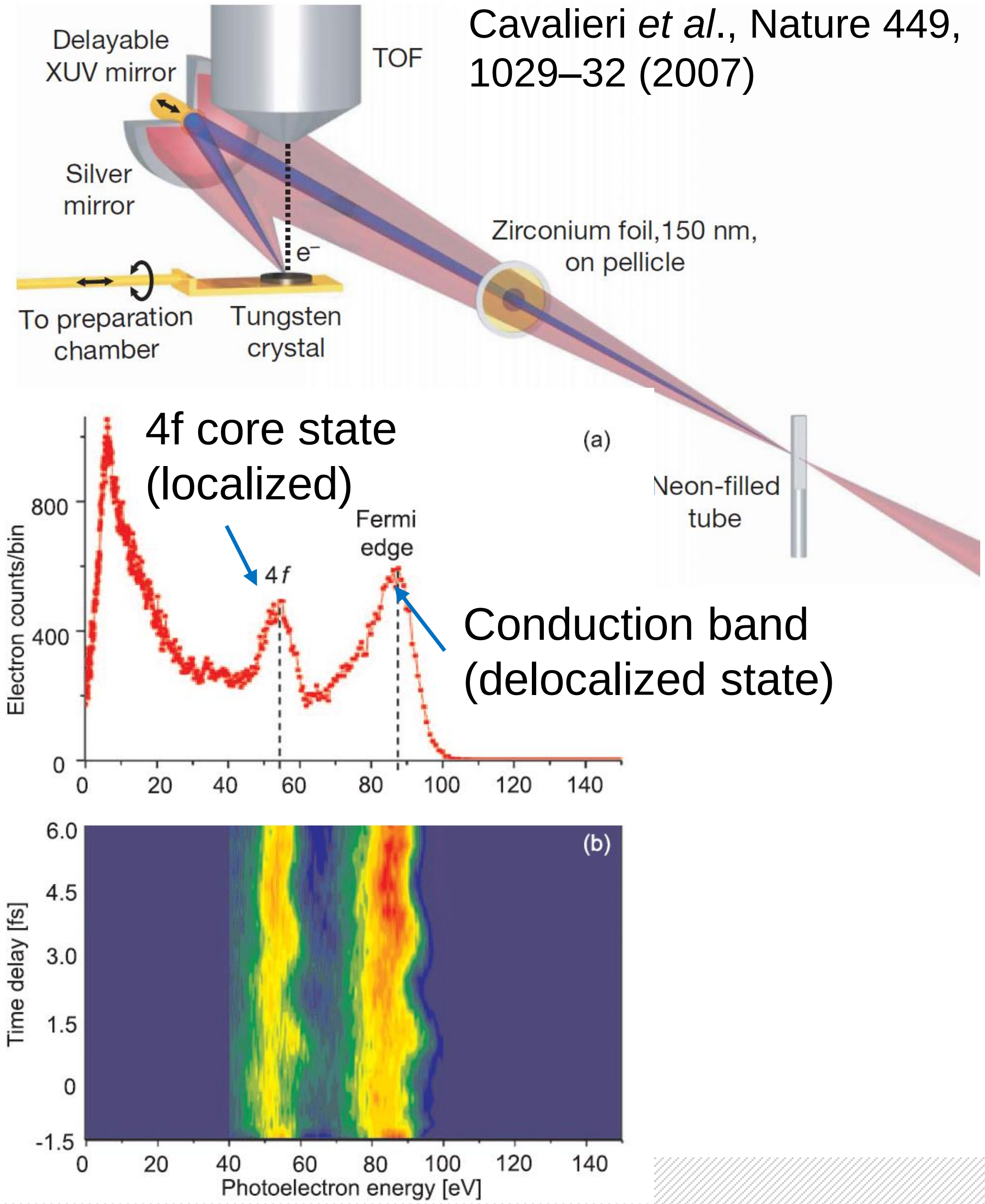
Zenghu Chang group,
Nat.Comm. 8, 186, 2017

Attosecond laboratory

Courtesy of Prof. Mauro Nisoli
Politecnico di Milano, Italy

ATTOSECOND RESEARCH CENTER

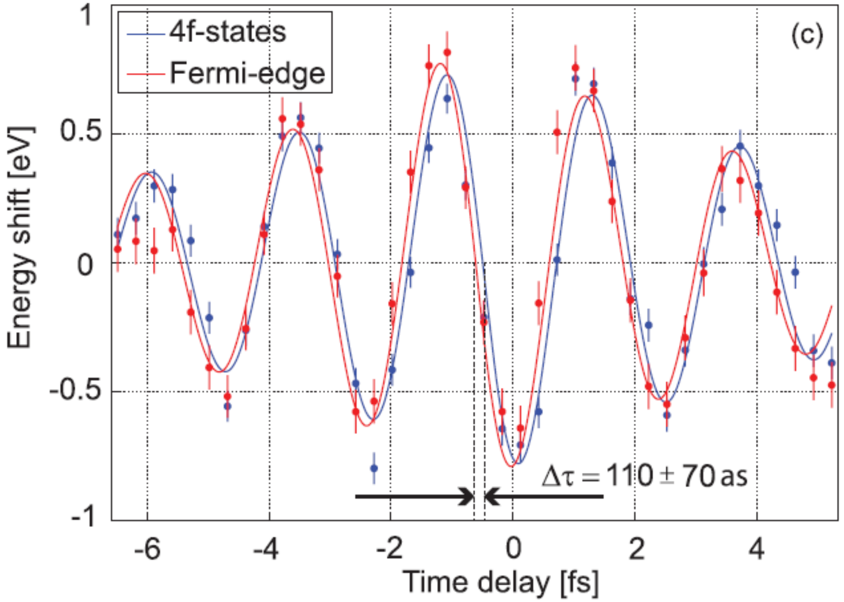
Photoelectron emission from single-crystal tungsten



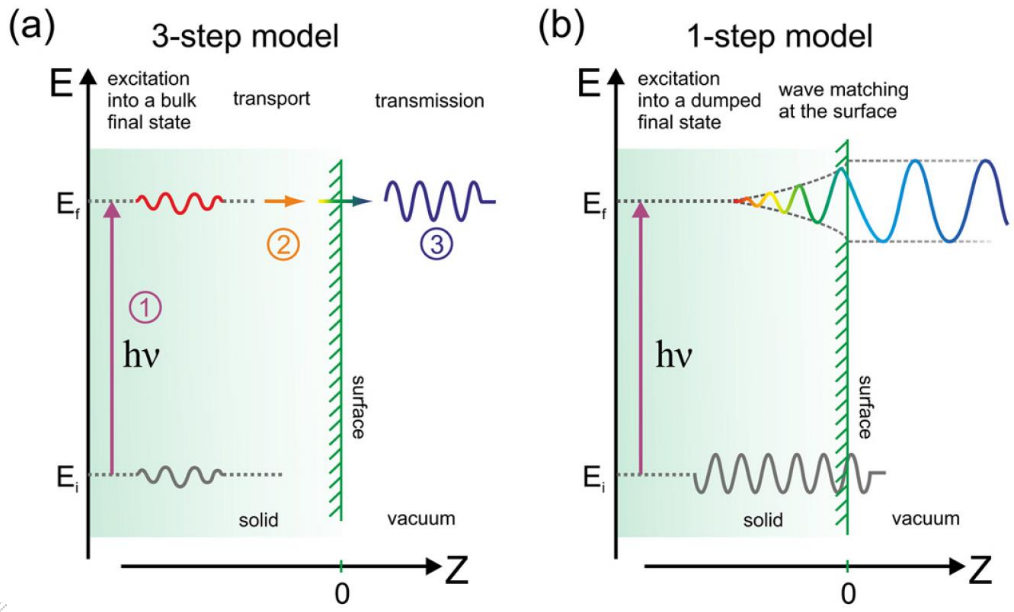
Cavalieri *et al*., Nature 449, 1029–32 (2007)

4f core state (localized)

Conduction band (delocalized state)

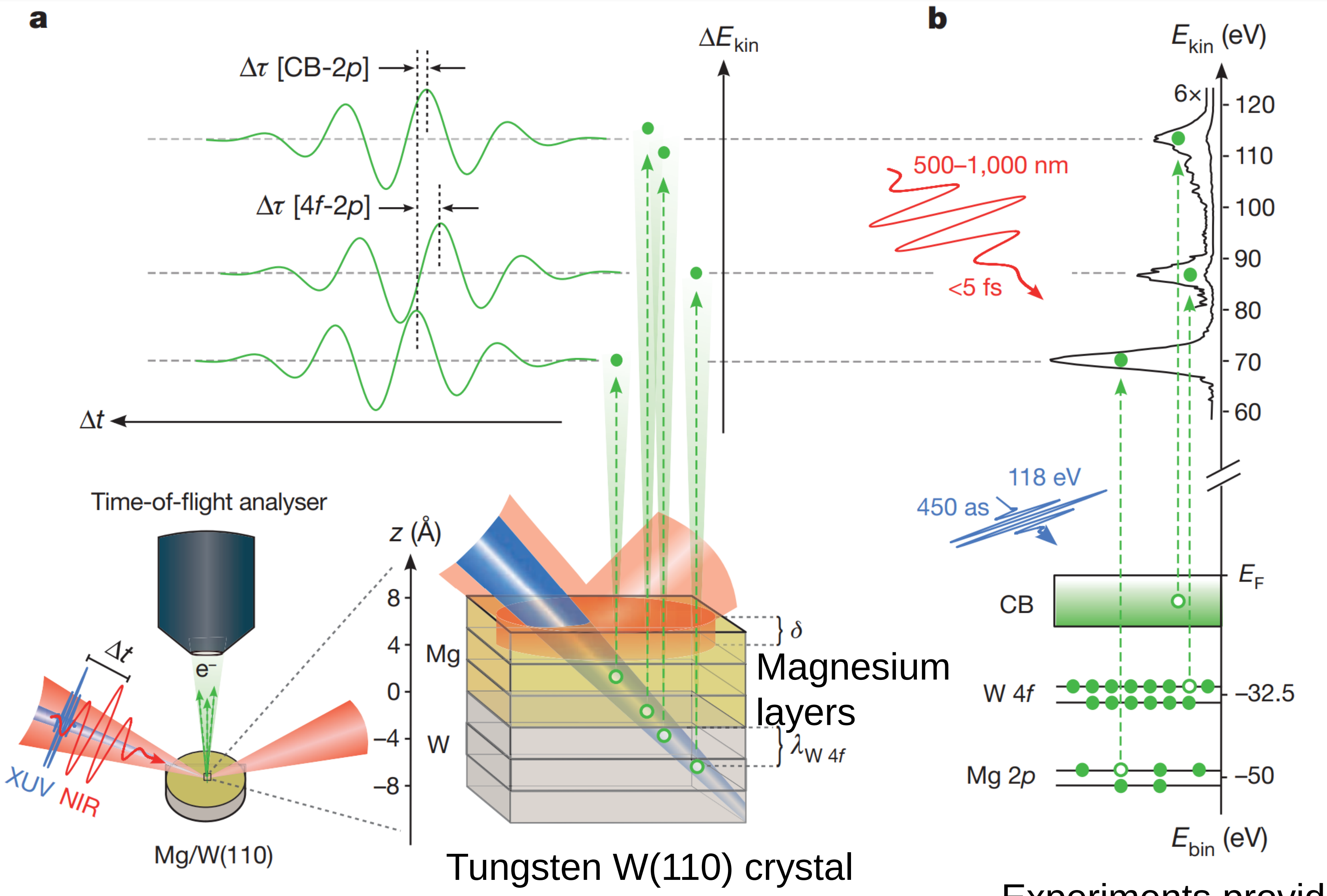


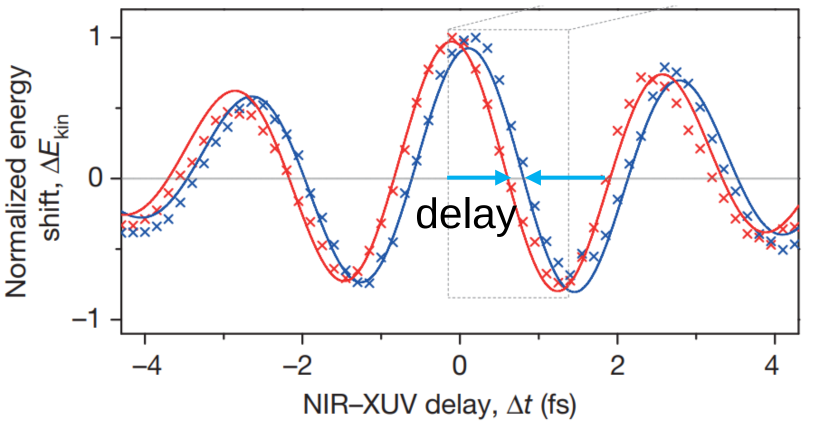
A 110 as delay was observed in emission of core electrons with respect to the emission of conduction band electrons

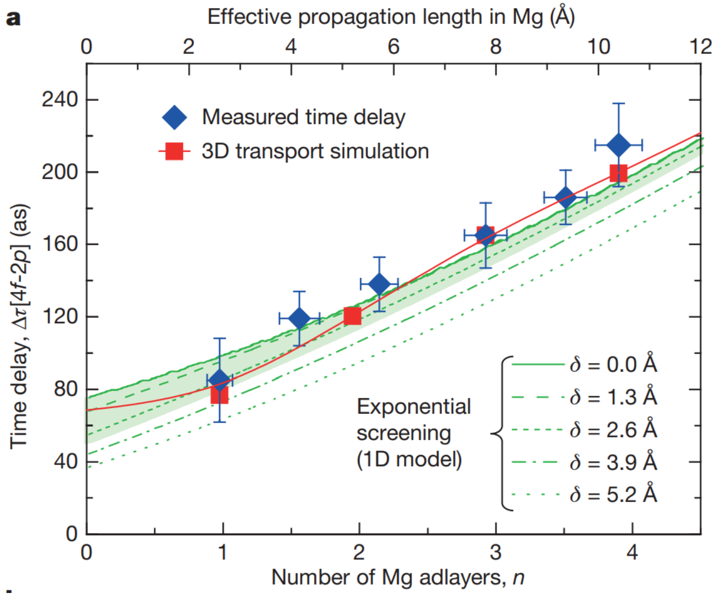


Controversy: How does the transport occur?

Electron propagation and dielectric screening





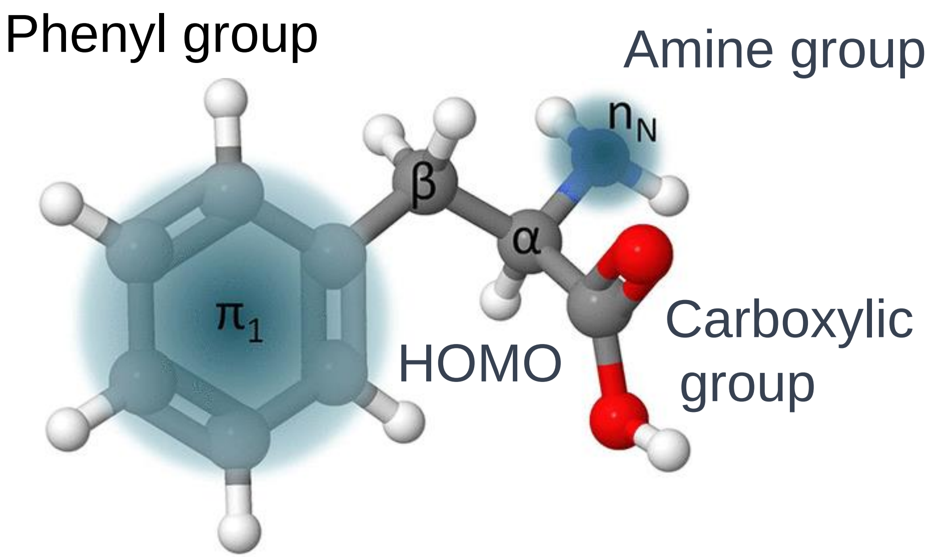


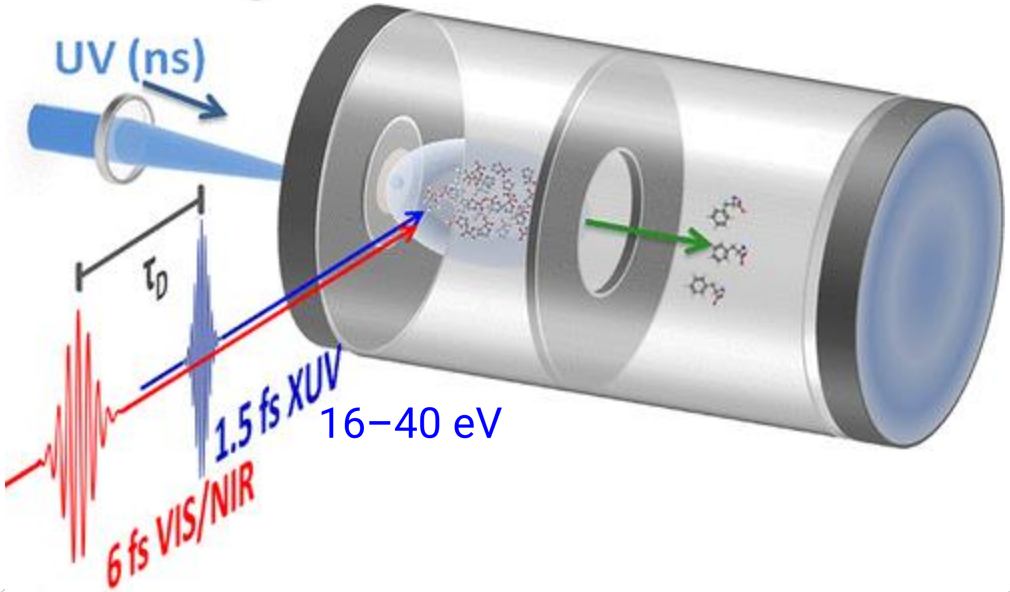
Neppl,… Krausz, Kienberger, Nature 517, 342 (2015)

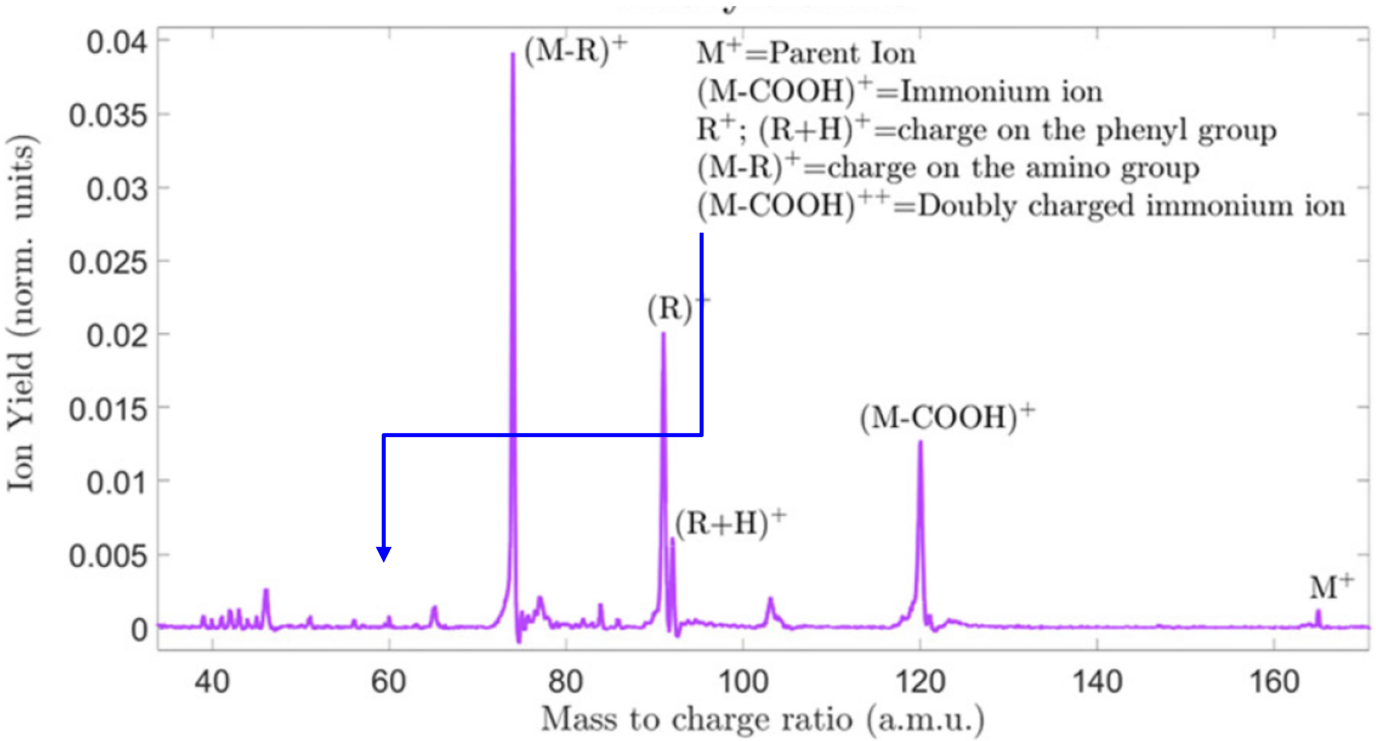
Experiments provided real-time access to atomic-scale charge transport dynamics in solids

Charge migration in aminoacids

Molecular structure of phenylalanine







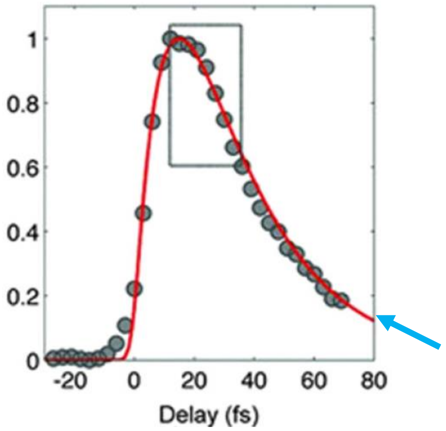
XUV pulse ionizes the molecule and induces CT states

The yield of immonium di-cations is a particularly sensitive probe of **charge location** because the local ionization potential will increase as the hole approaches the **amine group** causing ionization by the VIS/NIR pulse to be suppressed

Calegari *et al*., Science 346, 336–9 (2014)

Charge migration in aminoacids

The yield of immonium di-cations (M-COOH)$^{++}$

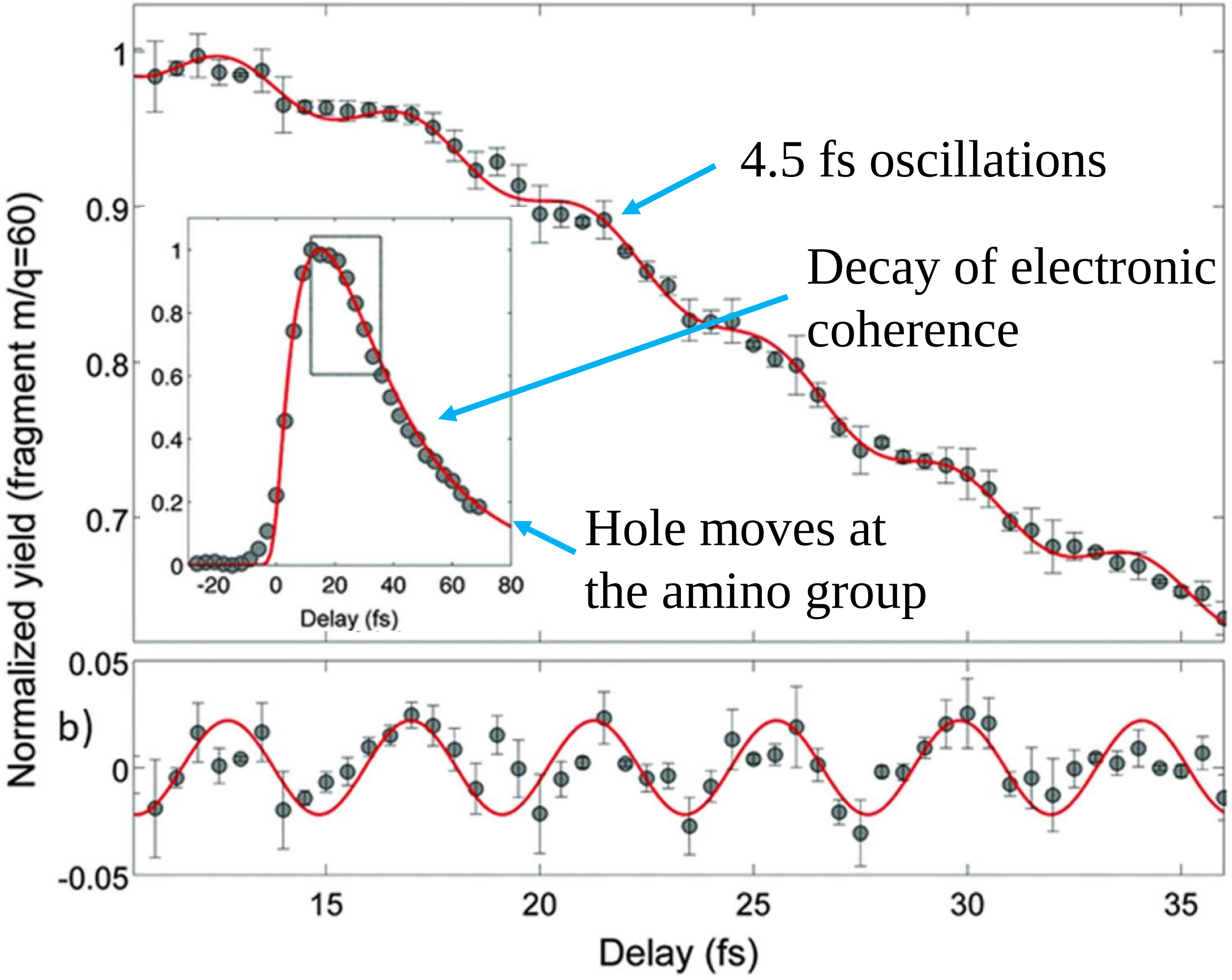


Hole moves at
the amino group

Calegari *et al.*, Science 346, 336–9 (2014)

Charge migration in aminoacids

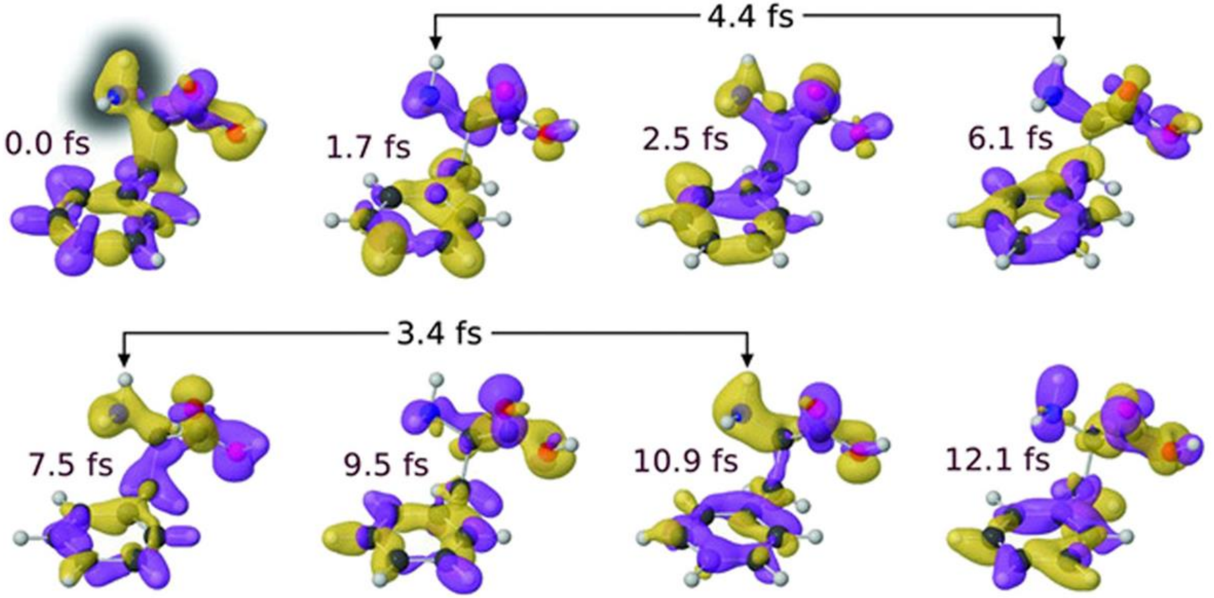
The yield of immonium di-cations (M-COOH)$^{++}$



4.5 fs oscillations

Decay of electronic coherence

Hole moves at the amino group



The periodic beating is attributed to a charge motion of **purely electronic origin** between amino and phenyl group

Calegari *et al.*, Science 346, 336–9 (2014)

Conclusions, or What's Next?

1. **Advancements in technology**: attosecond laser sources will become more sophisticated and more readily available (53 as pulses, Nat.Comm. 8, 186, 2017)

2. **New attosecond-resolution spectroscopies** (e.g., pump-probe and two-dimensional correlation spectroscopy) are within reach (perhaps, in combination with free-electron lasers)

3. **New insights into fundamental physics**: Electron dynamics in strongly correlated (magnetic) systems and topological materials. Learning about matter from HGG it produces. This may lead to the development of new materials with unique properties (Nature 538, 359, 2016)

4. **New strong field physics:** the quantum-optical nature of high harmonic generation and HHG entanglement (Nat. Comm 11, 4598, 2020; Nat. Phys. 17, 1104, 2021; Nat. Phys. 2023). This has implications for the development of quantum technologies

5. **New disease diagnostics**: Centre for Molecular Fingerprinting (https://www.cmf.hu/en/). Krausz: "we have been able to detect eight different types of cancers with an excellent efficiency. We have also detected one type of a very severe coronary disease, pre-diabetes, diabetes, and stroke." (BMC Cancer 21, 1287, 2021; Anal. Chem. 9565, 23, 2023)

Acknowledgments & References

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Reviews and other resources on the topic:

Attosecond spectroscopy for the investigation of ultrafast dynamics in atomic, molecular and solid-state physics, R. Borrego-Varillas, M. Lucchini and M. Nisoli, Rep. Prog. Phys. 85, 066401 (2022)

Attosecond science. D. Villeneuve, Contemp. Phys. 59, 47 (2018)

Attosecond Physics, Krausz and Ivanov. Rev. Mod. Phys. 81, 163 (2009)

What is an "attosecond pulse", and what can you use it for? Emilio Pisanty, https://physics.stackexchange.com/questions/782972/what-is-an-attosecond-pulse-and-what-can-you-use-it-for

https://www.nobelprize.org/prizes/physics/2023/press-release/

P. Corkum, Generation and Measurement of Attosecond Pulses (Technion lecture)

https://www.youtube.com/watch?app=desktop&v=XHjLSIVeaZg