# The world inside atoms revealed by ultrafast light pulses: The 2023 Physics Nobel Prize

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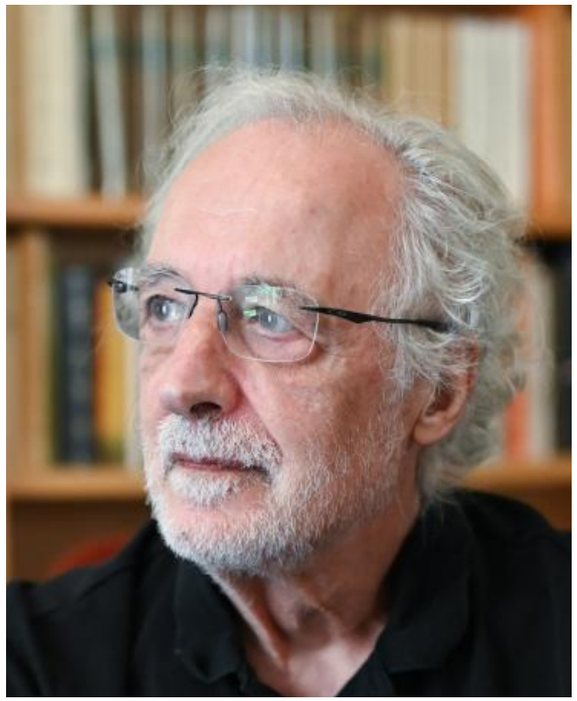


**Pierre Agostini**
The Ohio State University, USA

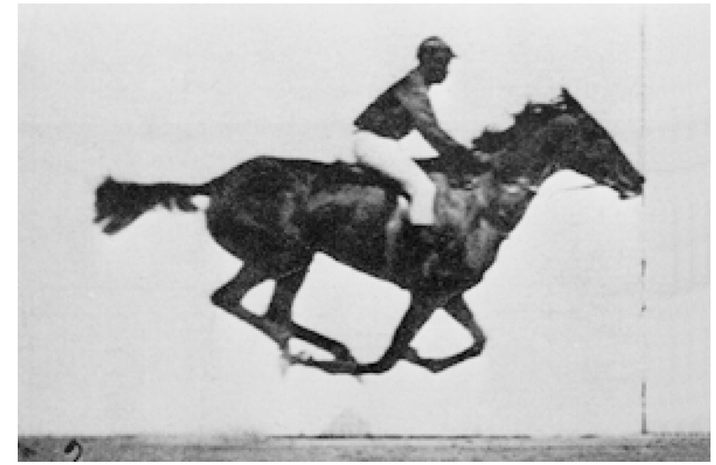
**Ferenc Krausz**
Max Planck Institute for Quantum Optics, Germany

**Anne L'Huillier**
Lund University, Sweden

*"for experimental methods that generate attosecond pulses of light for the study of electron dynamics in matter"*

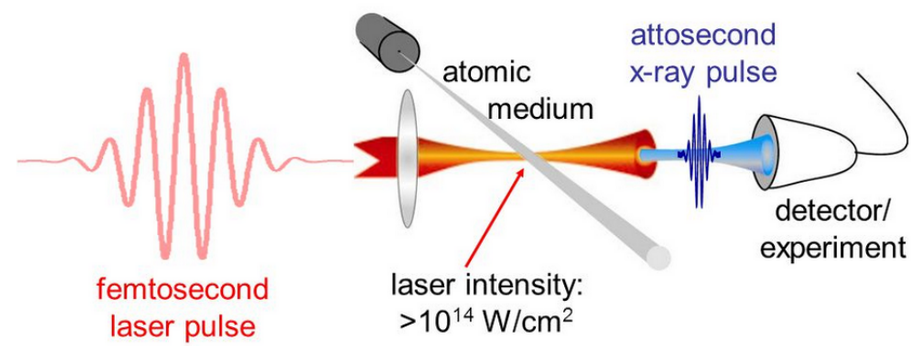
# Outline

1. **Imaging fast motion**
   - Leland Stanford's horse bet
   - Shutters and stroboscopes



2. **Attosecond light pulses**
   - How long is an attosecond?
   - Why attosecond pulses?

3. **Creating attosecond pulses**
   - Experiments by Agostini, Krausz, and L'Huillier
   - Attosecond physics



# Stanford's horse bet

**Mid 19th century:**

Leland Stanford bet $25,000 that all four of a horse's feet **come off the ground** at a fast trot

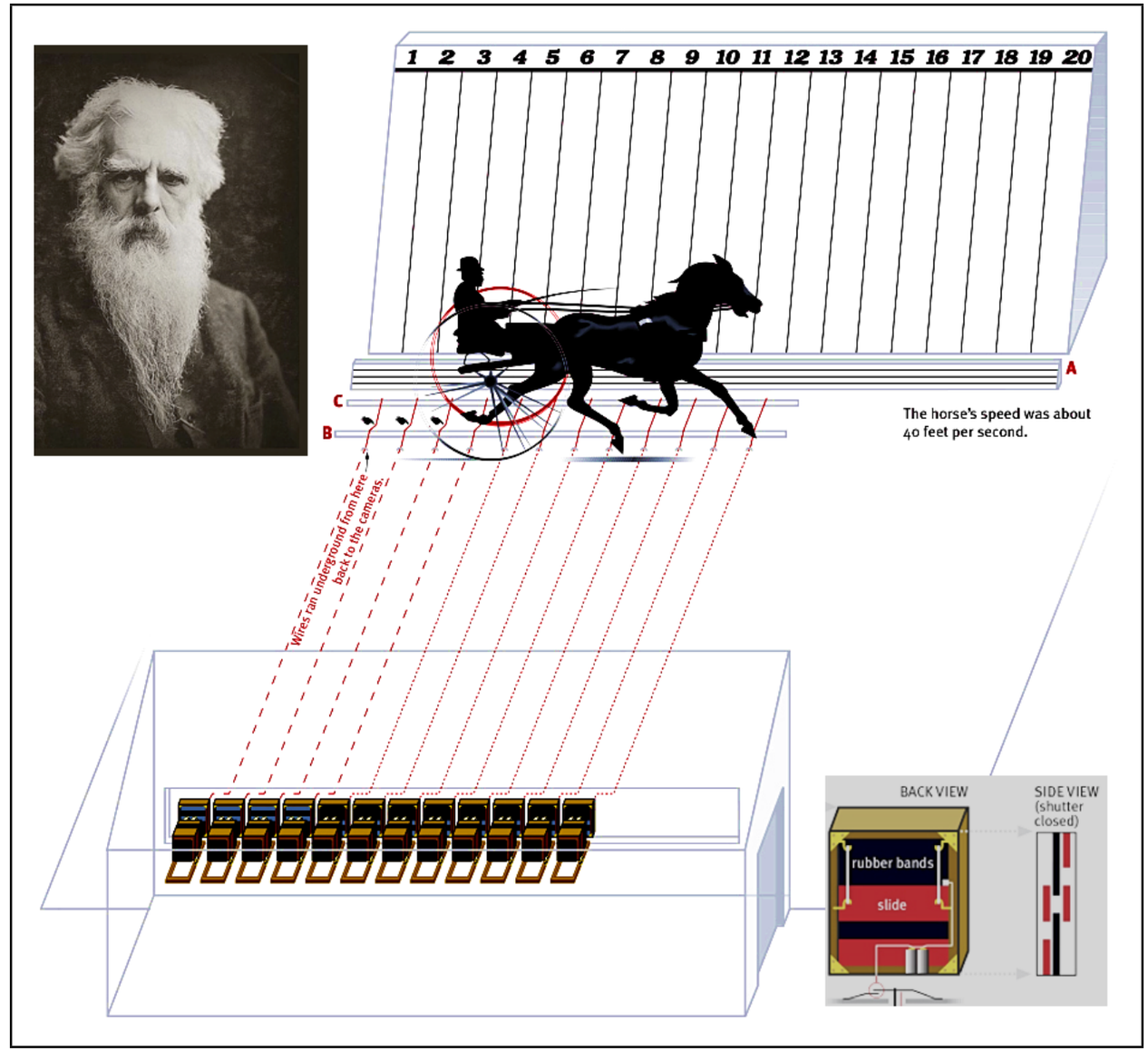

# Eadweard Muybridge: The man who stopped time

## Imaging a moving horse leg:

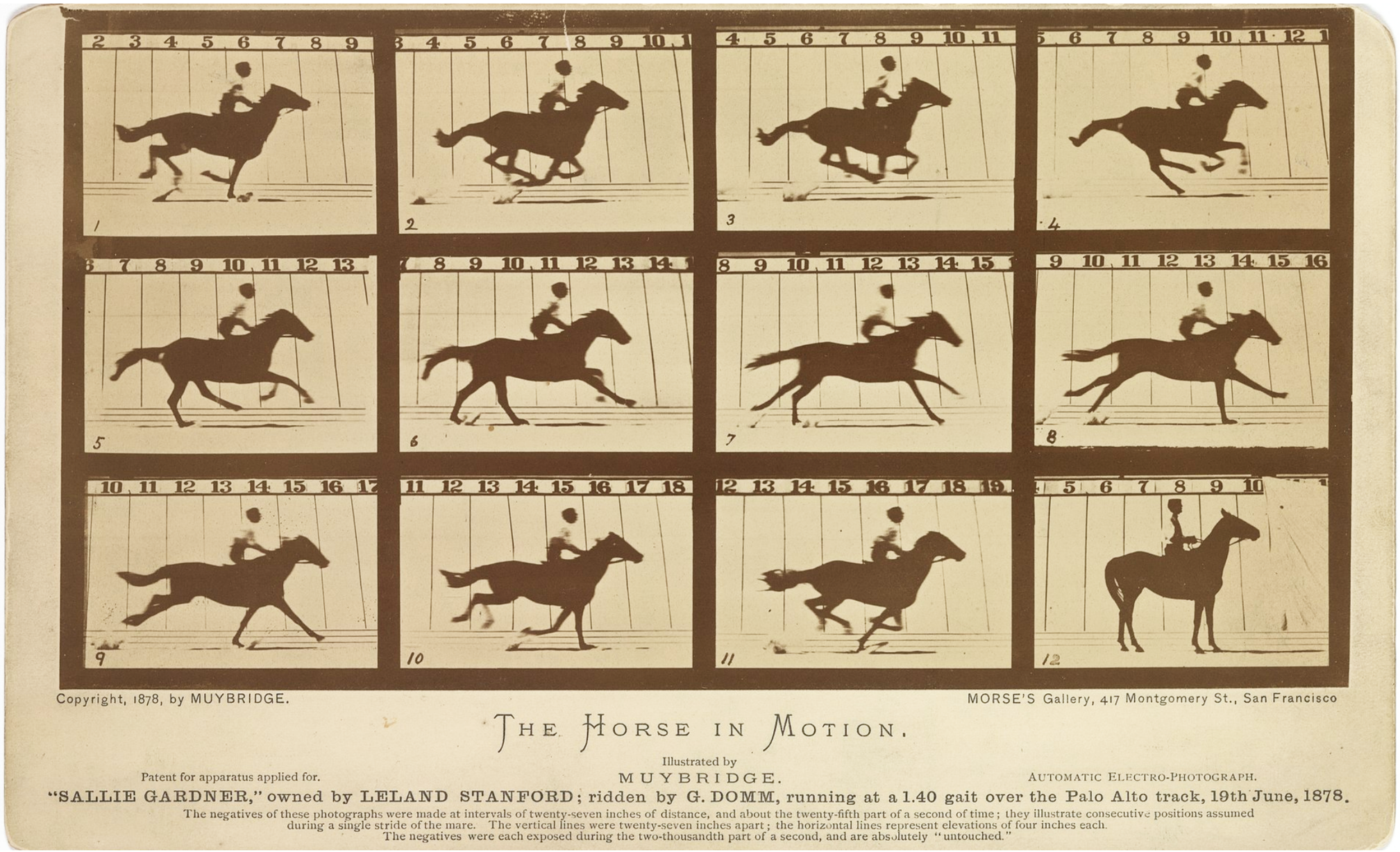
- required spatial resolution $\Delta x \approx 0.01\,\mathrm{m}$
- horse's speed $v \approx 10\,\mathrm{m/s}$
- **exposure time** should not exceed $T = \Delta x / v \approx 1/1000\,\mathrm{s} = 1\,\mathrm{ms}$

## First high-speed camera:

- 1873: Stanford approached English photographer **Eadweard Muybridge**
- Muybridge initially believed task impossible (exposures usually took seconds in 1870's)
- developed successful technique over 5+ years
- spring-loaded wooden shutters, triggered electrically, exposure time $1/1000\,\mathrm{s}$



# The horse in motion: book and early movie



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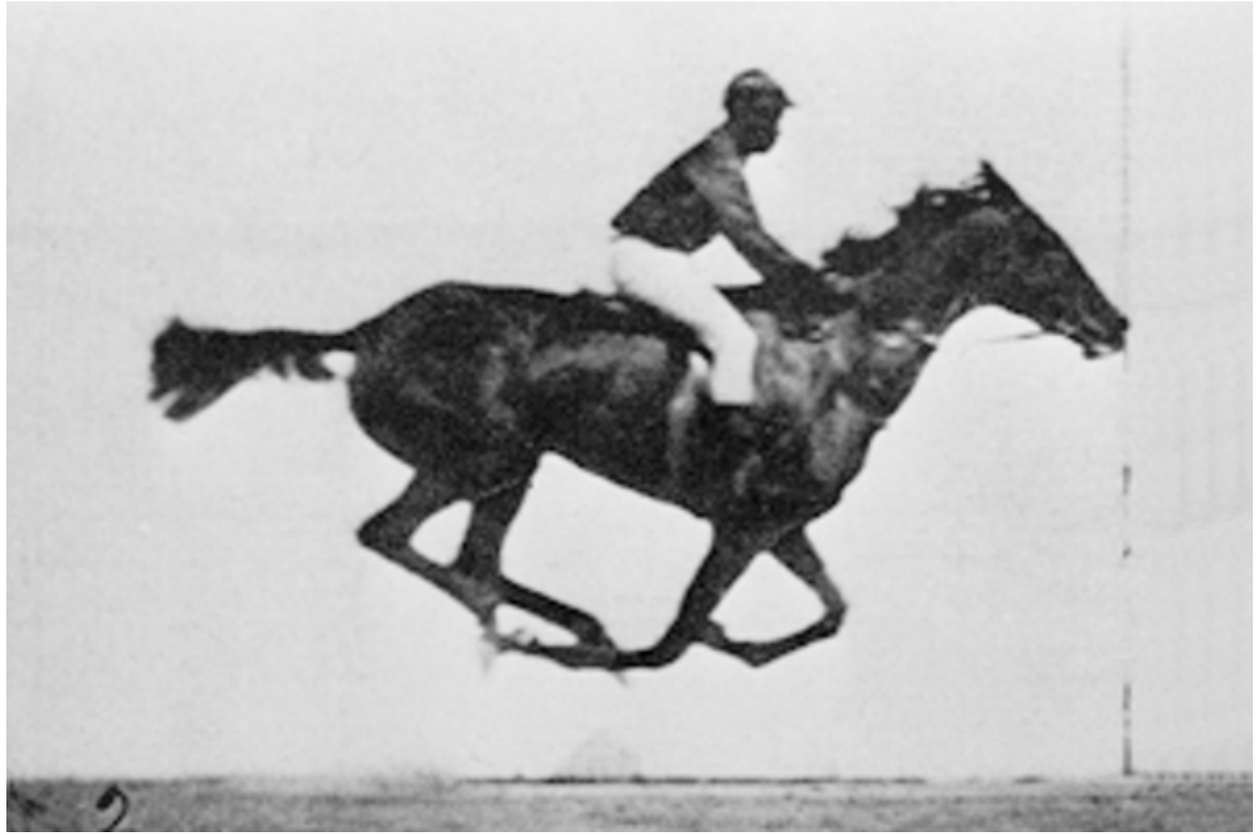


# Speed of motion and shutter speed

- hummingbird wings beat up to 100 times per second
- motion of wings "frozen" in picture when shutter speed faster than 1/100 s

Shutter speed 1/50s

1/2000s

**To image dynamic process, exposure needs to be shorter than characteristic time scale**

# Stroboscope

- instead of opening and closing a shutter, illuminate object by **light flash** or sequence of flashes
- can be achieved mechanically, for example via rotating wheel
- shorter pulses produced by electronic **flash tubes**

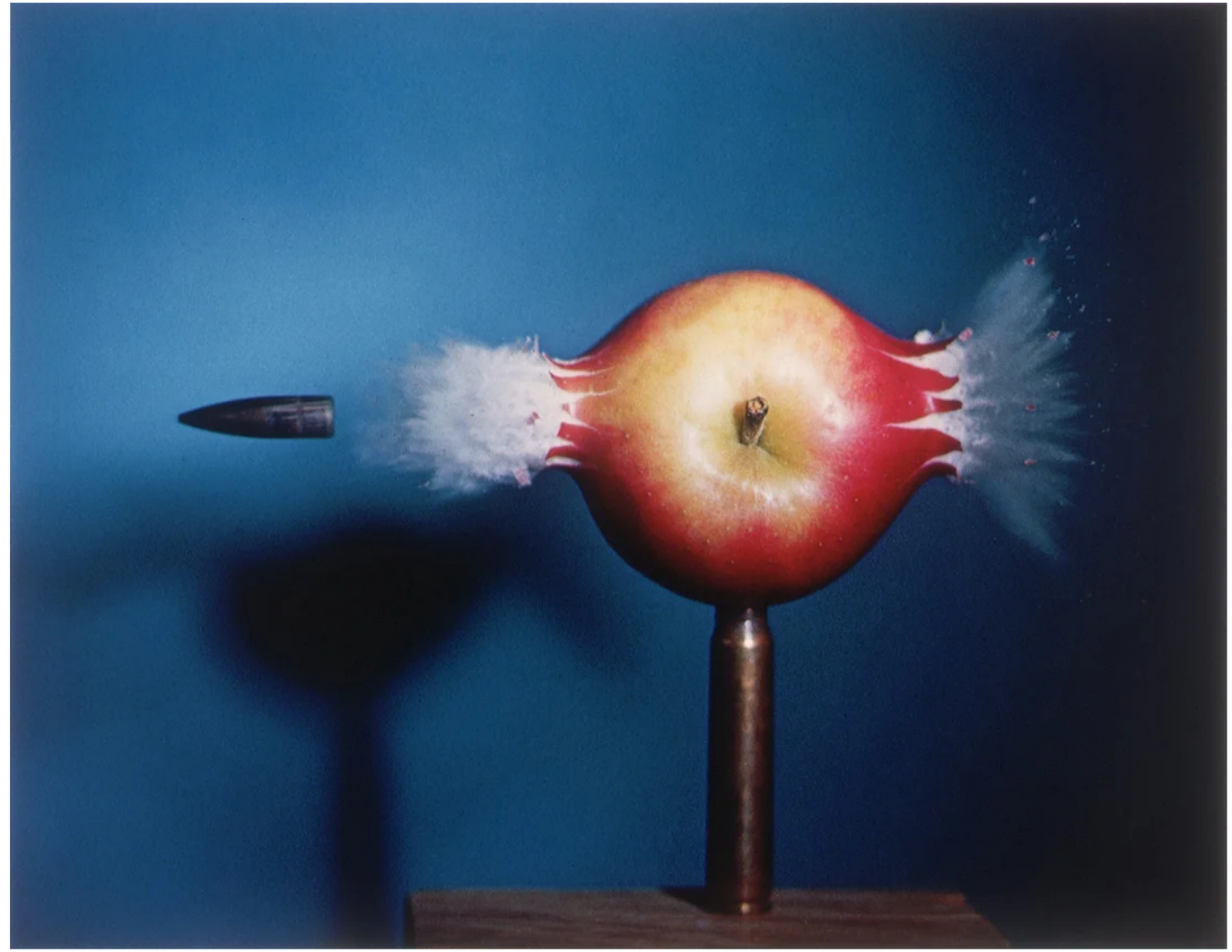
25 flashes per second

# Imaging bullet in flight

- speed about 1000 m/s,
- required spatial resolution about 1mm

$\Rightarrow$ exposures **shorter than 1 microsecond**

- MIT professor **Harold Edgerton** popularized using stroboscopes in science
- 1964: photograph *Bullet through Apple*, taken using a pulse of 1/3 microseconds



*Bullet through Apple*, Smithsonian American Art Museum

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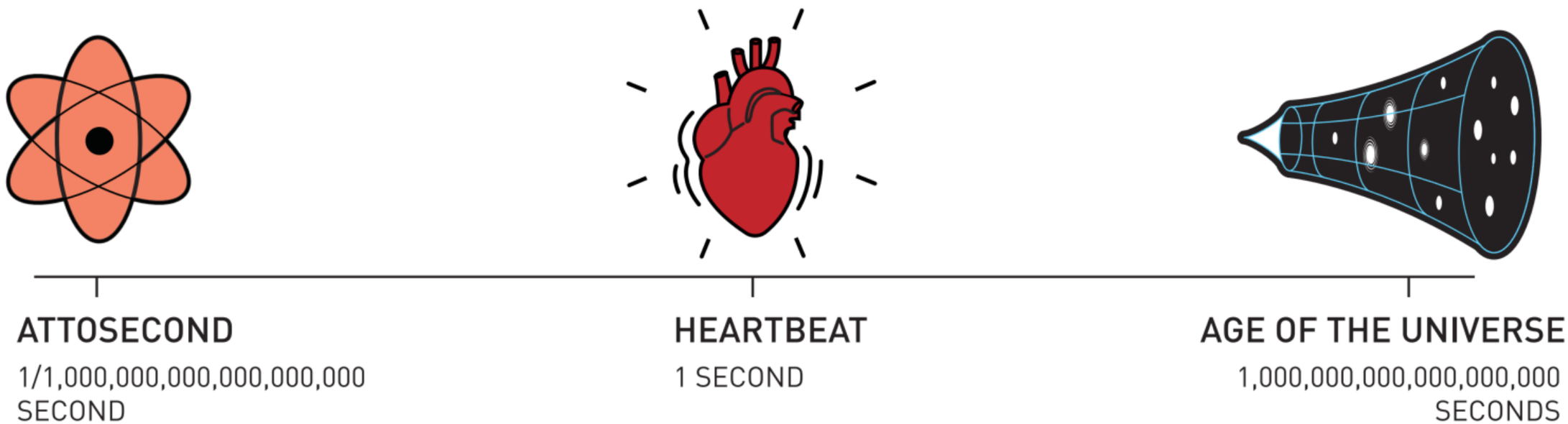
# Metric unit prefixes

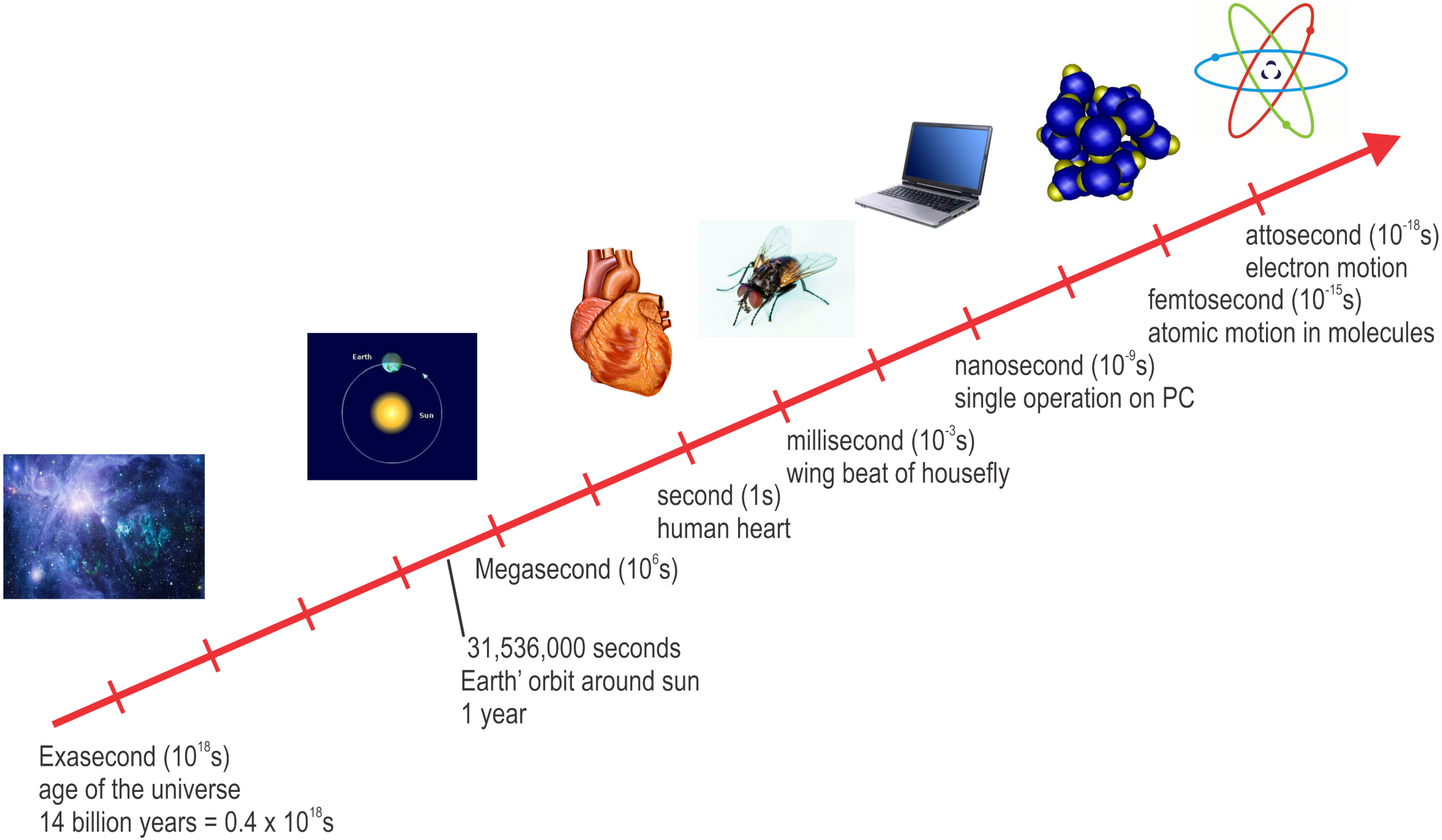
| Prefix | Exponent | Number | Scientific Notation | Name |
|---|---|---|---|---|
| Exa (E) | 18 | 1,000,000,000,000,000,000 | $10^{18}$ | quintillion |
| Peta (P) | 15 | 1,000,000,000,000,000 | $10^{15}$ | quadrillion |
| Tera (T) | 12 | 1,000,000,000,000 | $10^{12}$ | trillion |
| Giga (G) | 9 | 1,000,000,000 | $10^{9}$ | billion |
| Mega (M) | 6 | 1,000,000 | $10^{6}$ | million |
| kilo (k) | 3 | 1,000 | $10^{3}$ | thousand |
| hecto (h) | 2 | 100 | $10^{2}$ | hundred |
| deca (da) | 1 | 10 | $10^{1}$ | ten |
| | 0 | 1 | $10^{0}$ | one |
| deci (d) | -1 | 0.1 | $10^{-1}$ | one tenth |
| centi (c) | -2 | 0.01 | $10^{-2}$ | one hundredth |
| milli (m) | -3 | 0.001 | $10^{-3}$ | one thousandth |
| micro (μ) | -6 | 0.000001 | $10^{-6}$ | one millionth |
| nano (n) | -9 | 0.000000001 | $10^{-9}$ | one billionth |
| pico (p) | -12 | 0.000000000001 | $10^{-12}$ | one trillionth |
| femto (f) | -15 | 0.000000000000001 | $10^{-15}$ | one quadrillionth |
| atto (a) | -18 | 0.000000000000000001 | $10^{-18}$ | one quintillionth |

# The attosecond

- attosecond is a **billionth of a billionth** of a second, $1\,\mathrm{as} = 10^{-18}\,\mathrm{s}$
- number of of attoseconds in a second is about equal to the number of seconds in the **age of the universe**



# Why attosecond pulses? Timescales of natural processes

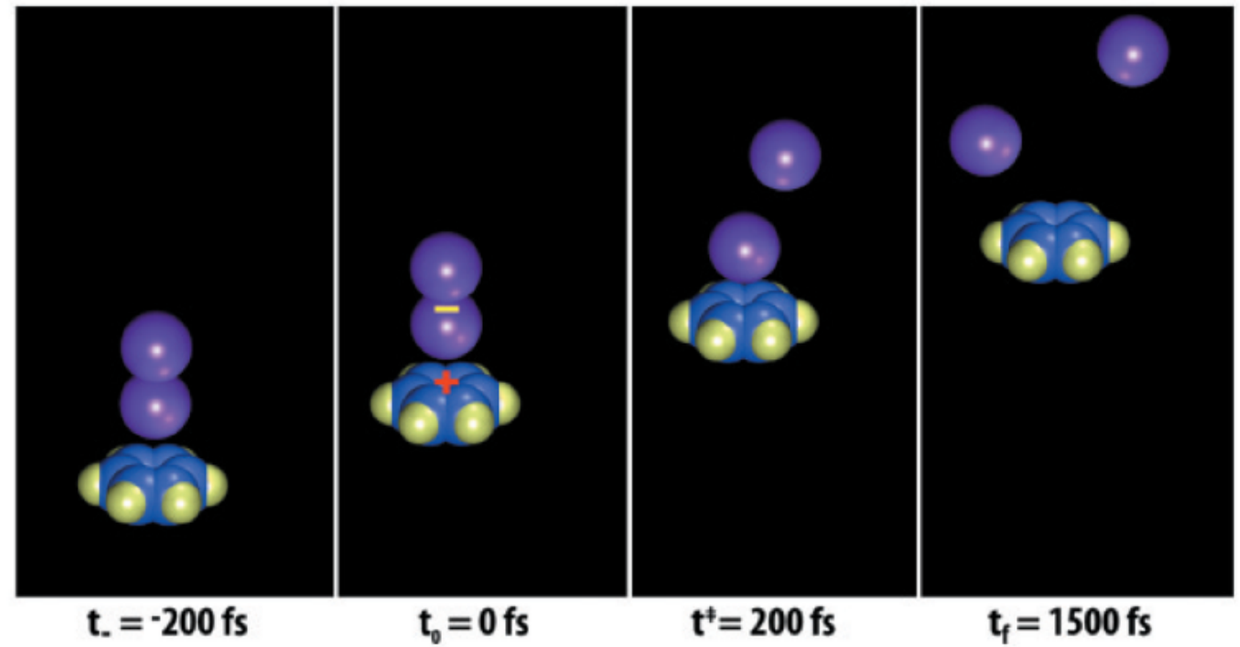


# Detour: Femtophysics and femtochemistry

**Imaging the motion of atoms and molecules:**

- spatial resolution $10^{-11}$ m (atom size $10^{-10}$ m)
- speed at room temperature: about 1000 m/s
- exposure time should not exceed $10^{-14}$ s = 10 femtoseconds

$\Rightarrow$ use **femtosecond laser pulses**




Reaction splitting iodine molecule by electron exchange with benzene, Baskin and Zewail (2001)

**1999 Nobel Prize in Chemistry:**

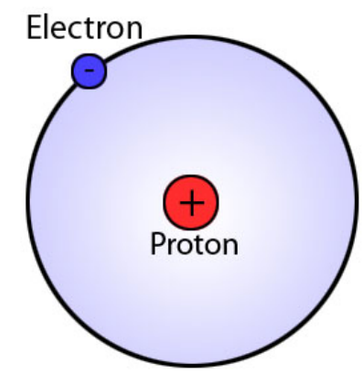
**Ahmed H. Zewail** (California Institute of Technology)

*"for his studies of the transition states of chemical reactions using femtosecond spectroscopy"*

# Electron dynamics in matter

- electrons are **much lighter** than atomic nuclei (proton mass is about 2000 times electron mass)

$\Rightarrow$ electrons move **much faster** than atoms



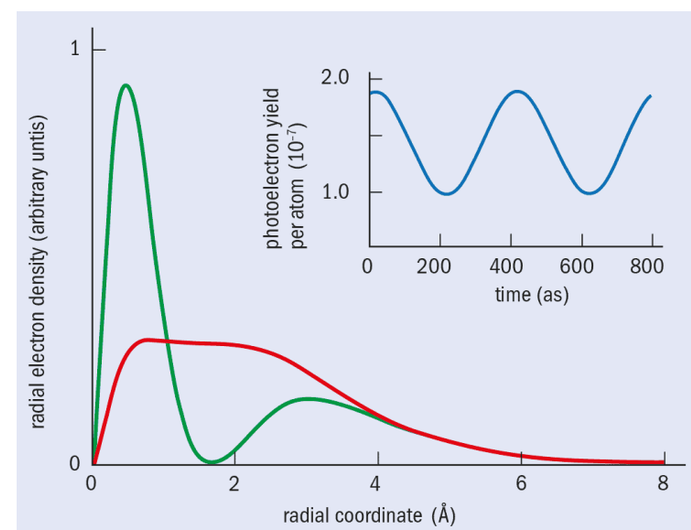

## Classical (Bohr) model of hydrogen:

- electron orbits proton at radius of 53 picometers ($53 \times 10^{-12}$ m)
- orbital period: **150 attoseconds**

## Quantum mechanics of hydrogen:

- real-time dynamics created by superposition of different eigenstates
- example: superposition of 1s and 2s states oscillates with period

$$T = h/|E_1 - E_2| \approx 400 \text{ attoseconds}$$




**Imaging electron dynamics requires attosecond light pulses**

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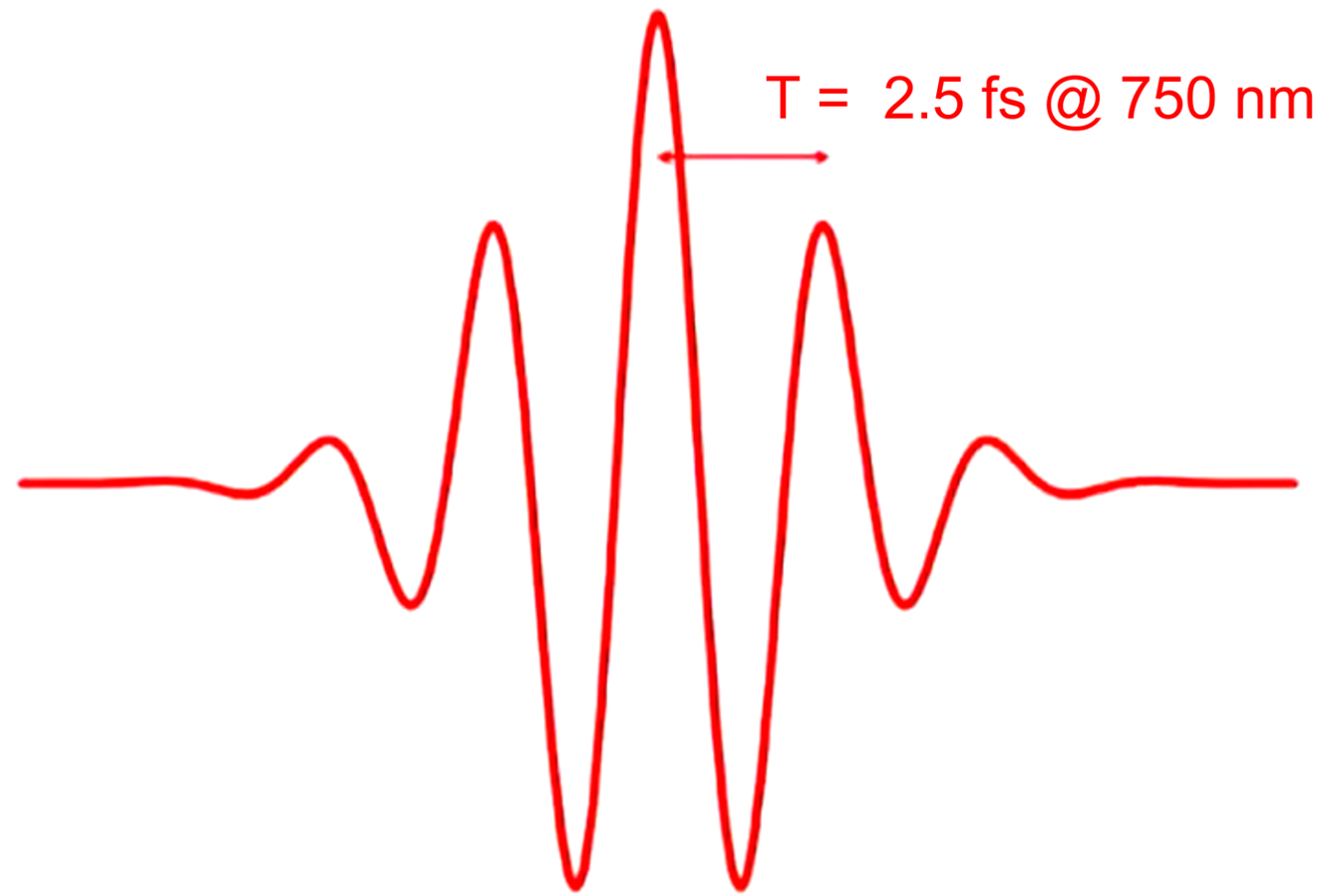
# Are sub-femtosecond pulses possible?

- light is (electromagnetic) **wave**
- short pulse is wave packet
- pulse cannot be shorter than one oscillation period $T = \lambda/c$
- typical laser systems have periods of a few femtoseconds
- at first glance, sub-femtosecond pulses seem **impossible**



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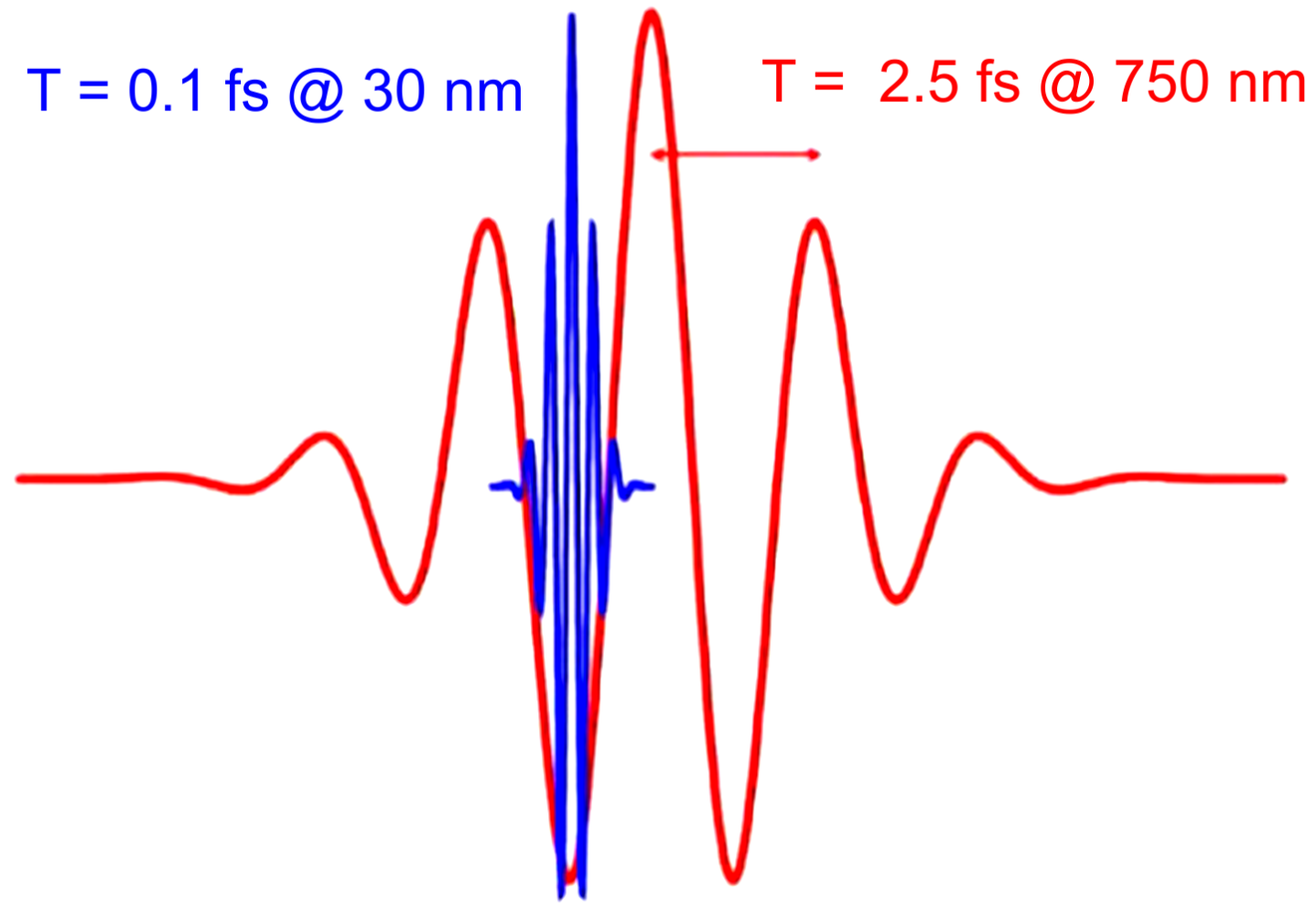
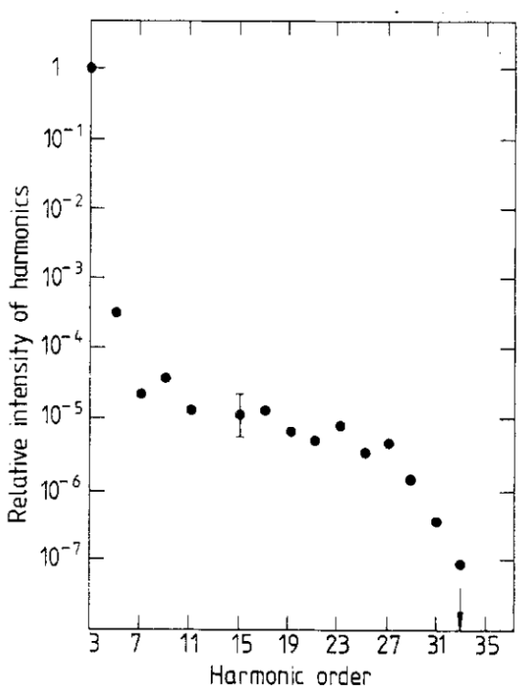
**Need coherent XUV radiation, light with much shorter period/wavelength!**

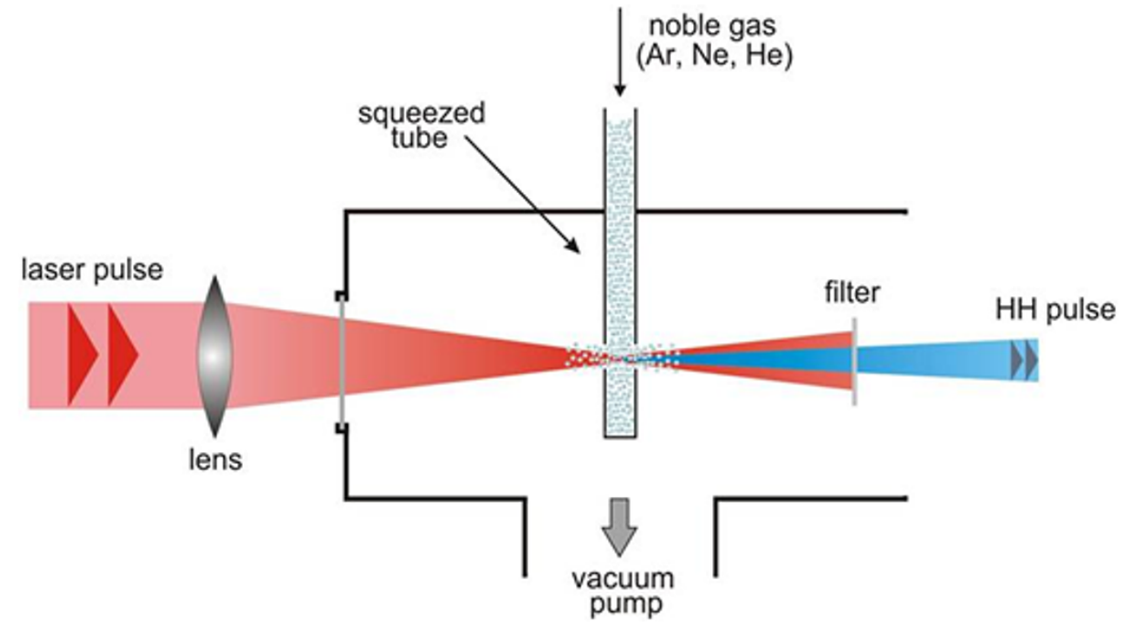


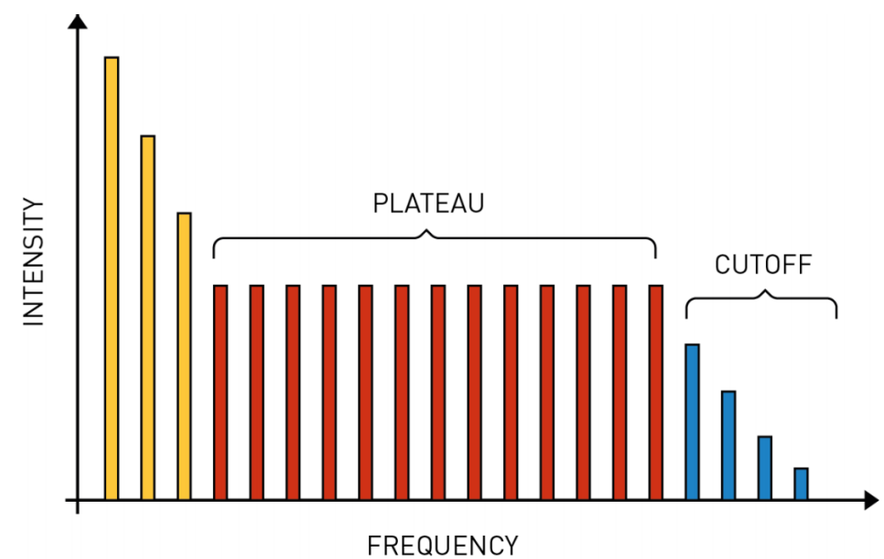
# Anne L'Huillier: High-harmonic generation

- in 1987, L'Huillier sent light of powerful laser through nobel gases (Ar, Kr, Xe)
- interaction between laser and gas atoms generates light at much higher frequencies
- specifically, it generates **high harmonics**, waves at **integer multiples** of original frequency

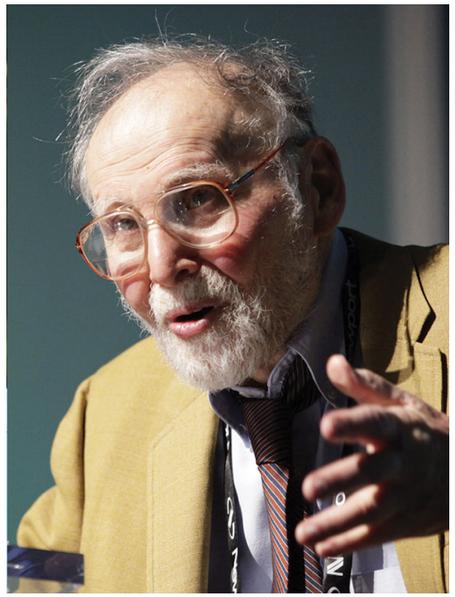



M. Ferray et al., J. Phys. B 21, L31 (1988)



# Detour 1: 2018 Physics Nobel Prize



**Arthur Ashkin**
Bell Laboratories

*"for the optical tweezers and their application to biological systems"*

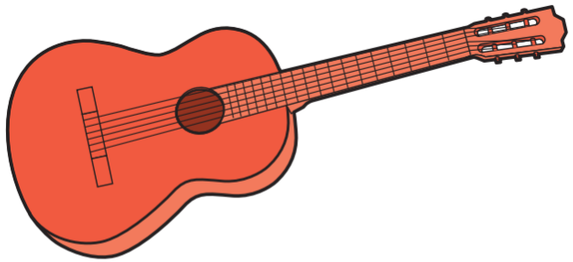
**Gérard Mourou**
École Polytechnique Palaiseau

**Donna Strickland**
University of Waterloo

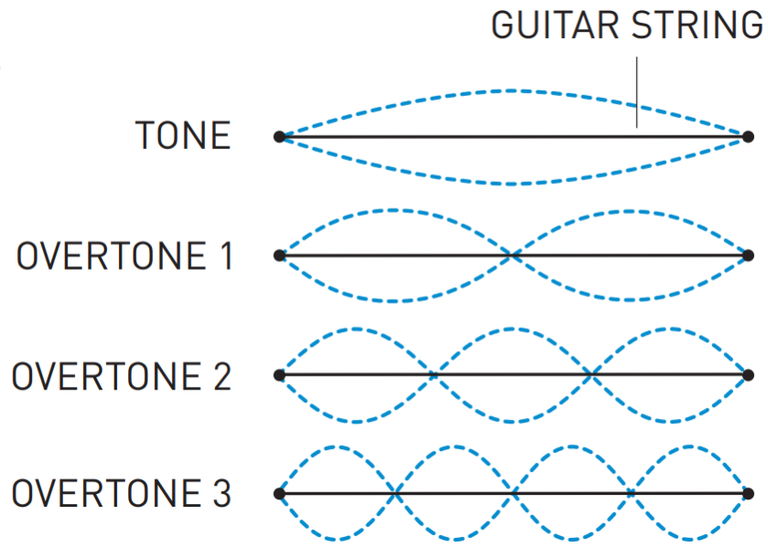
*"for their method of generating high-intensity ultra-short optical pulses"*

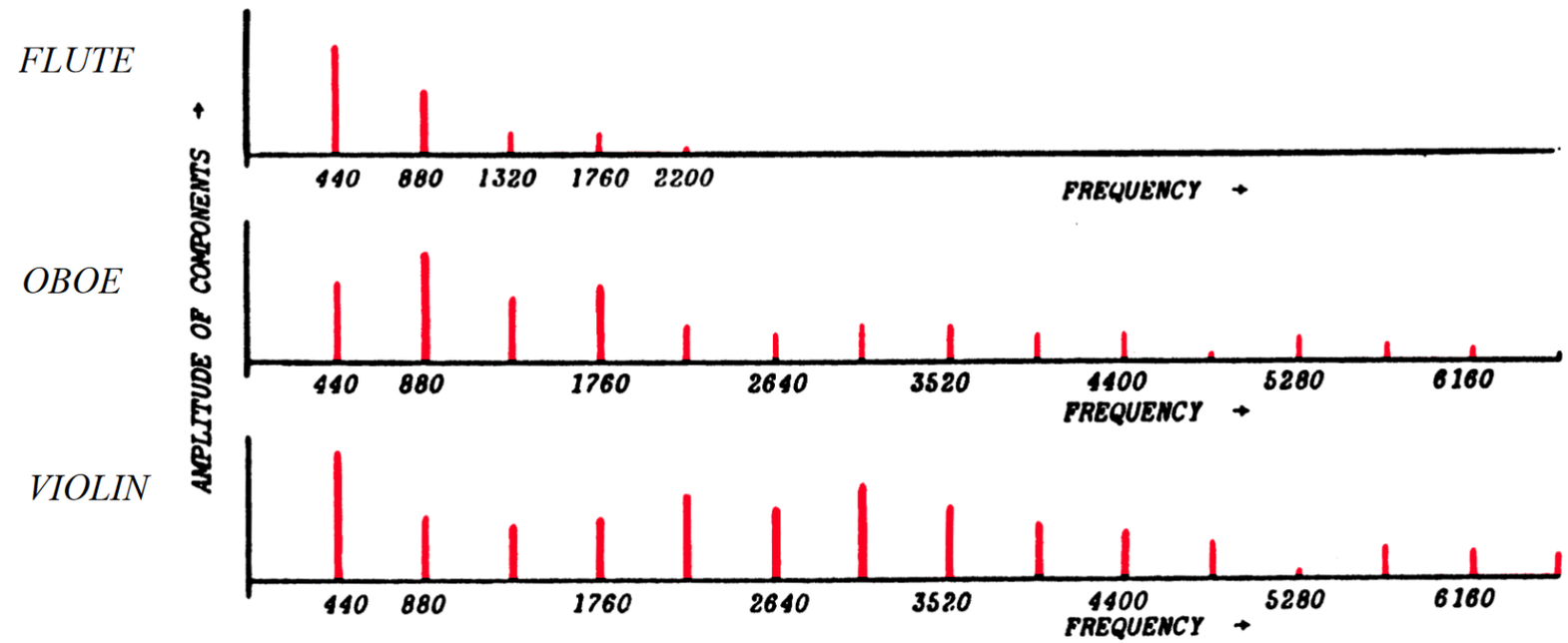
**Chirped pulse amplification**

# Detour 2: Overtones - harmonics of sound waves

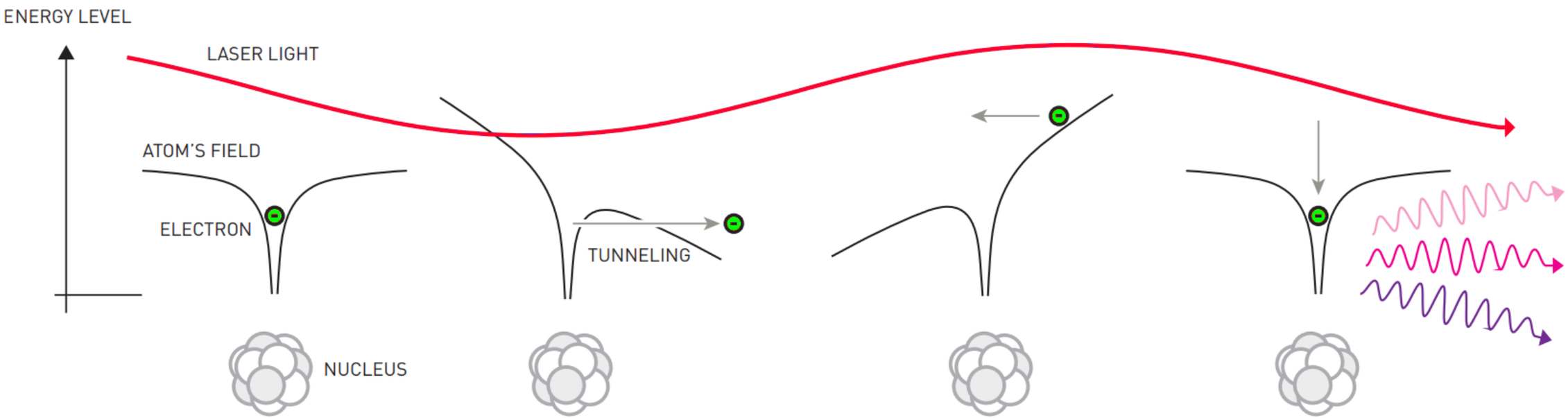


Overtones have several cycles for each cycle in the fundamental tone. Overtones work the same way in light waves.



# How does high-harmonic generation work?




1 An electron that is bound to an atom's nucleus cannot normally leave its atom; it does not have enough energy to lift itself out of the well created by the atom's electrical field.

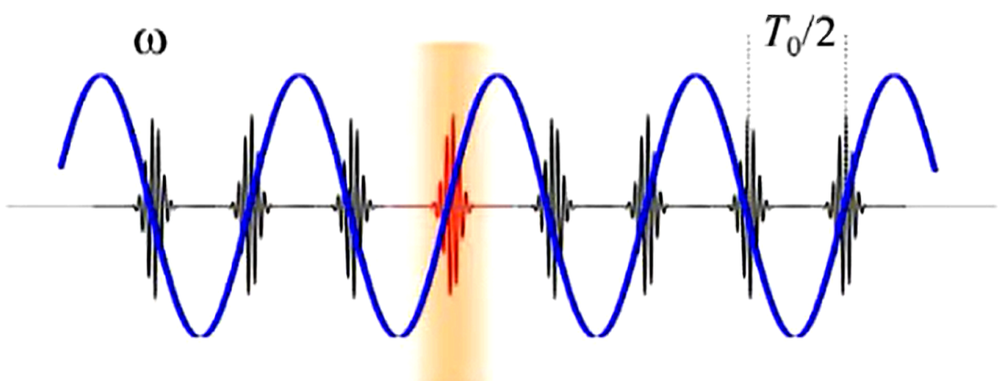
2 The atom's field is distorted when it is affected by the laser pulse. When the electron is only held by a narrow barrier, quantum mechanics allow it to tunnel out and escape.

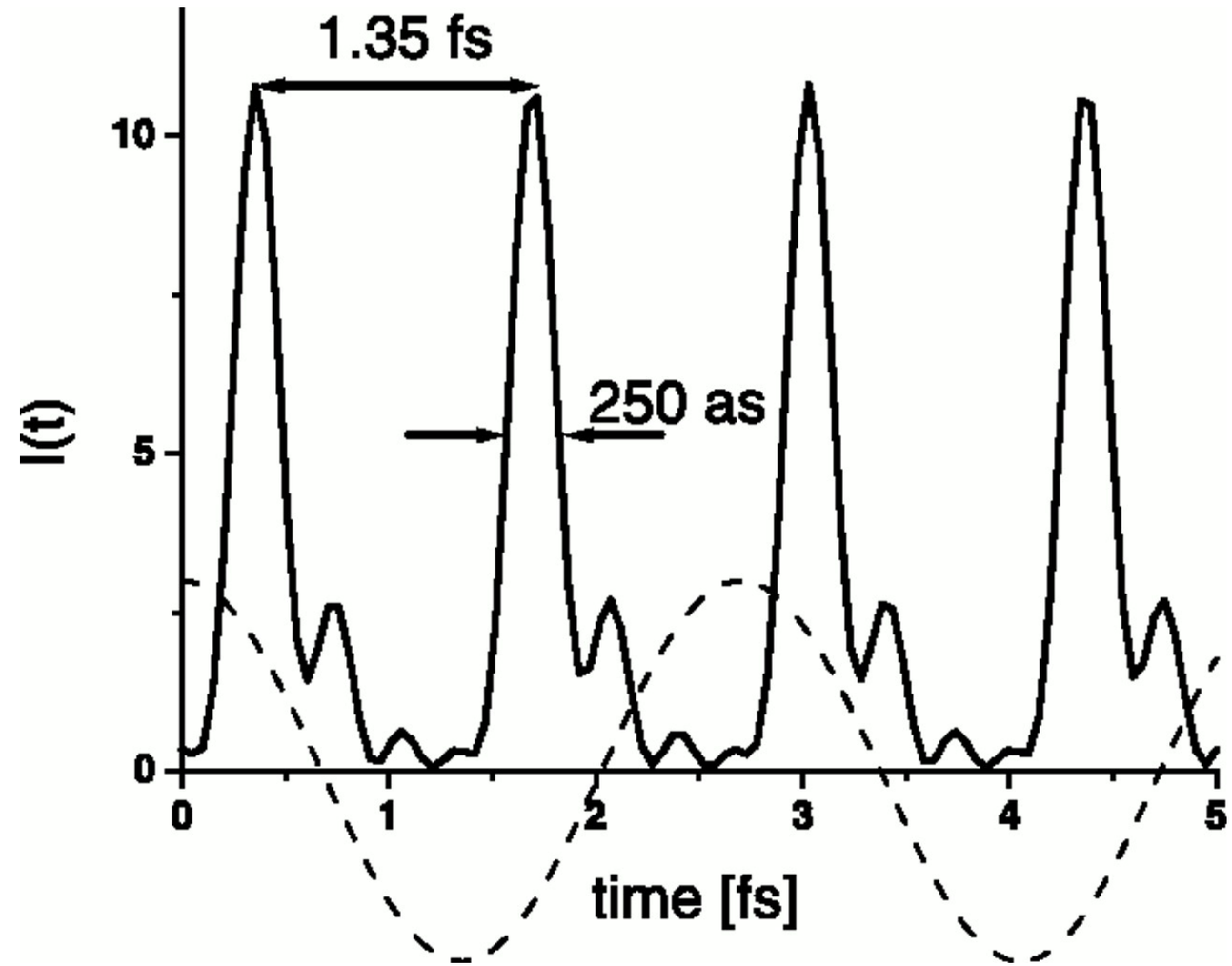
3 The free electron is still affected by the laser field and gains some extra energy. When the field turns and changes direction, the electron is pulled back in the direction it came from.

4 To reattach to the atom's nucleus, the electron must rid itself of the extra energy it gained during its journey. This is emitted as an ultraviolet flash, the wavelength of which is linked to that of the laser field, and differs depending on how far the electron moved.

# Pierre Agostini: Attosecond pulse trains

- in 1990's Pierre Agostini and coworkers developed techniques to analyze and measure attosecond pulses
- in 2001, Agostini's group produced train of pulses with duration of 250 attoseconds

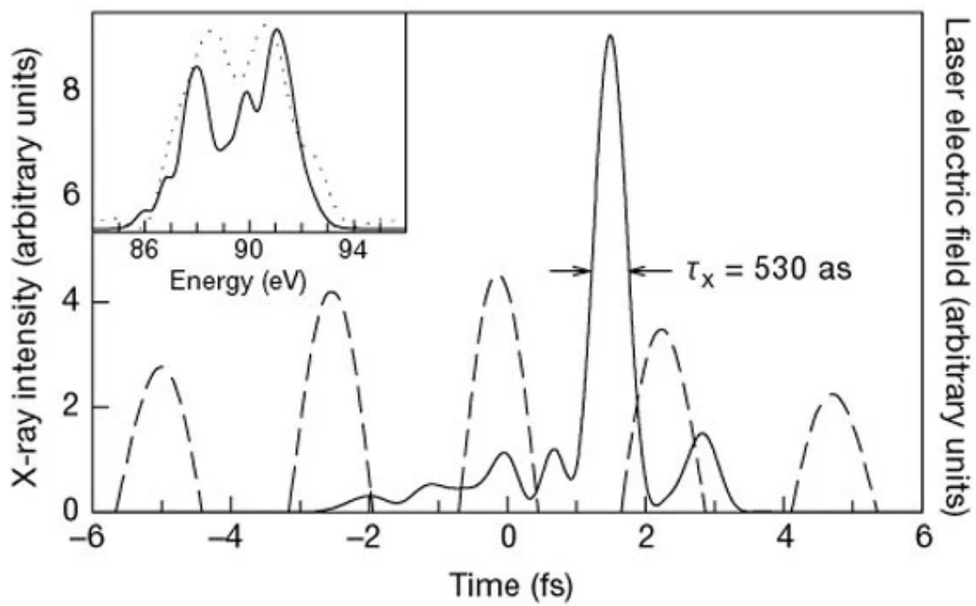


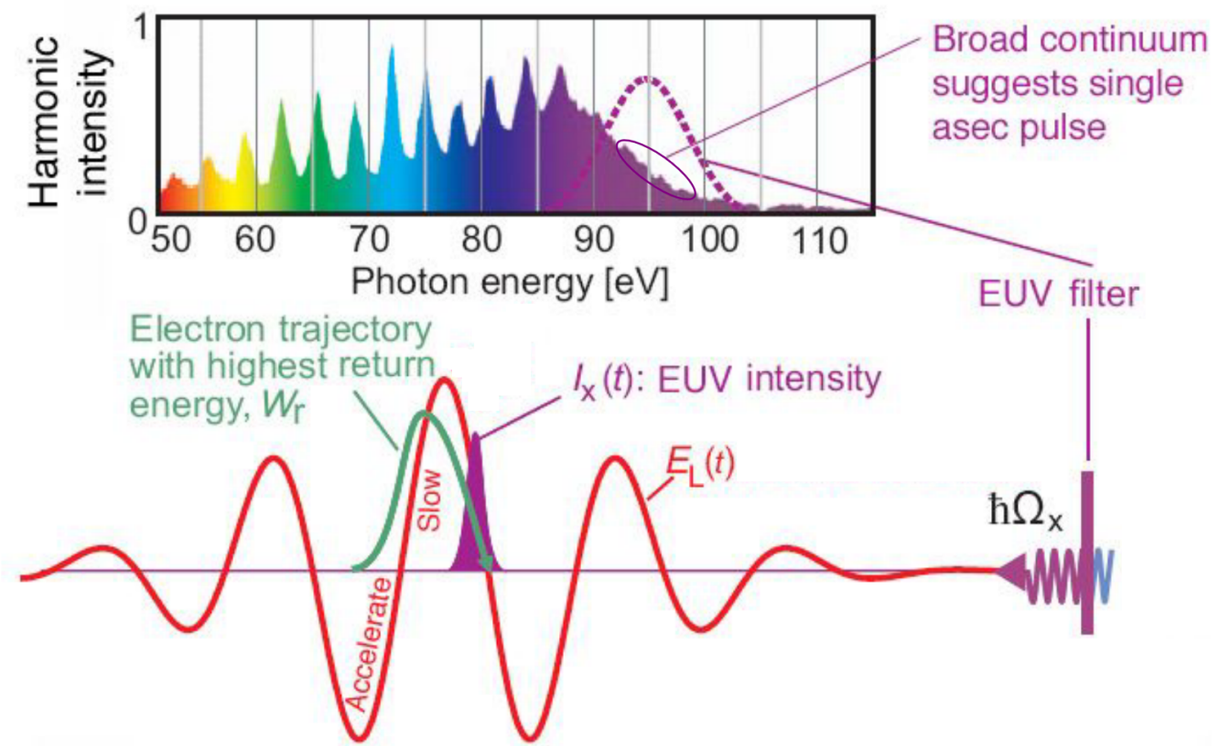

Paul et al., Science 292, 1689 (2001)

# Ferenc Krausz: Isolated attosecond pulses

- start with short, few-cycle laser pulse
- highest harmonics only generated in central laser cycle
- selecting these harmonics via spectral filter leads to single XUV pulse

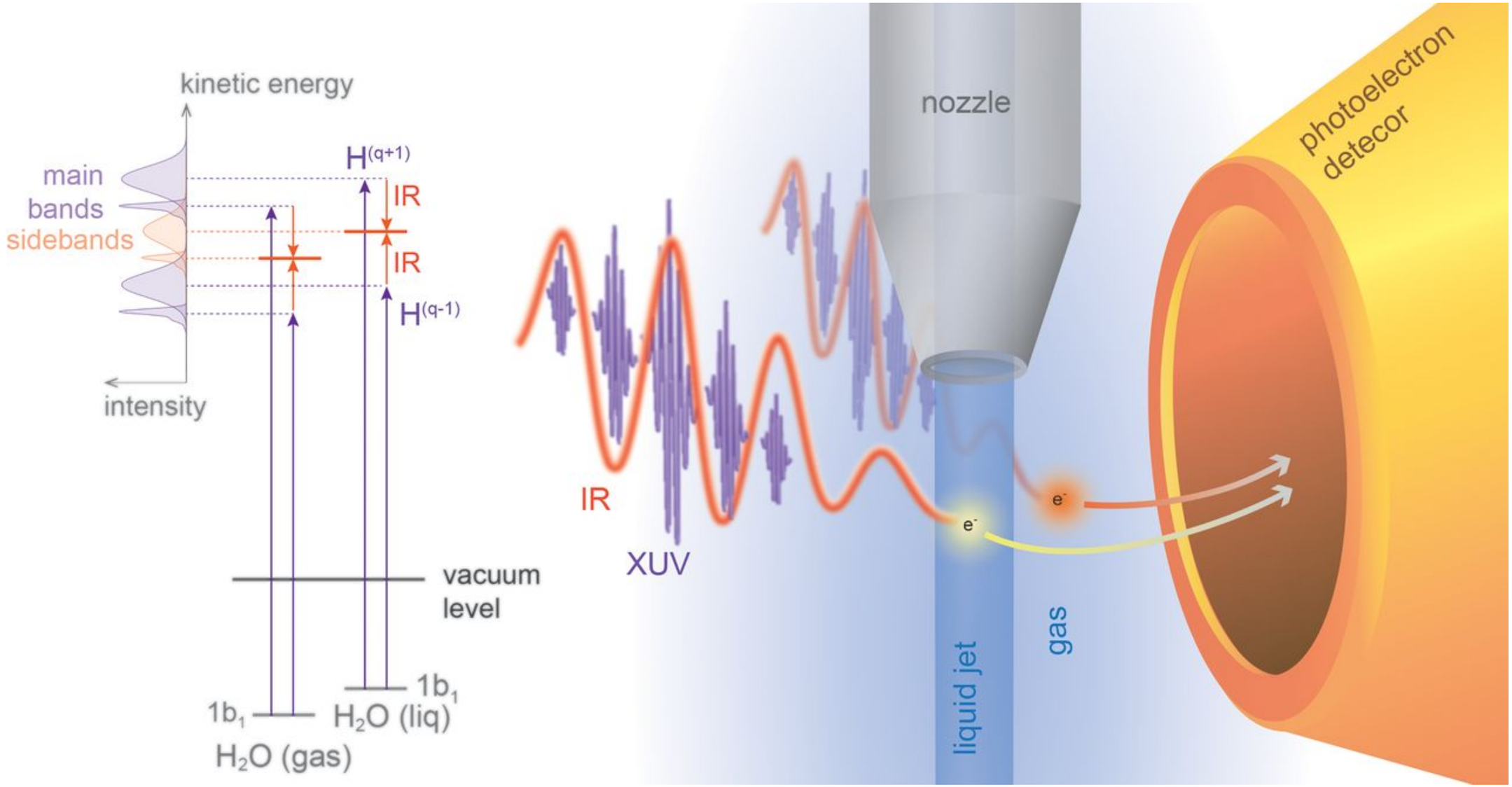



Hentschel et al., Nature 414, 509 (2001)



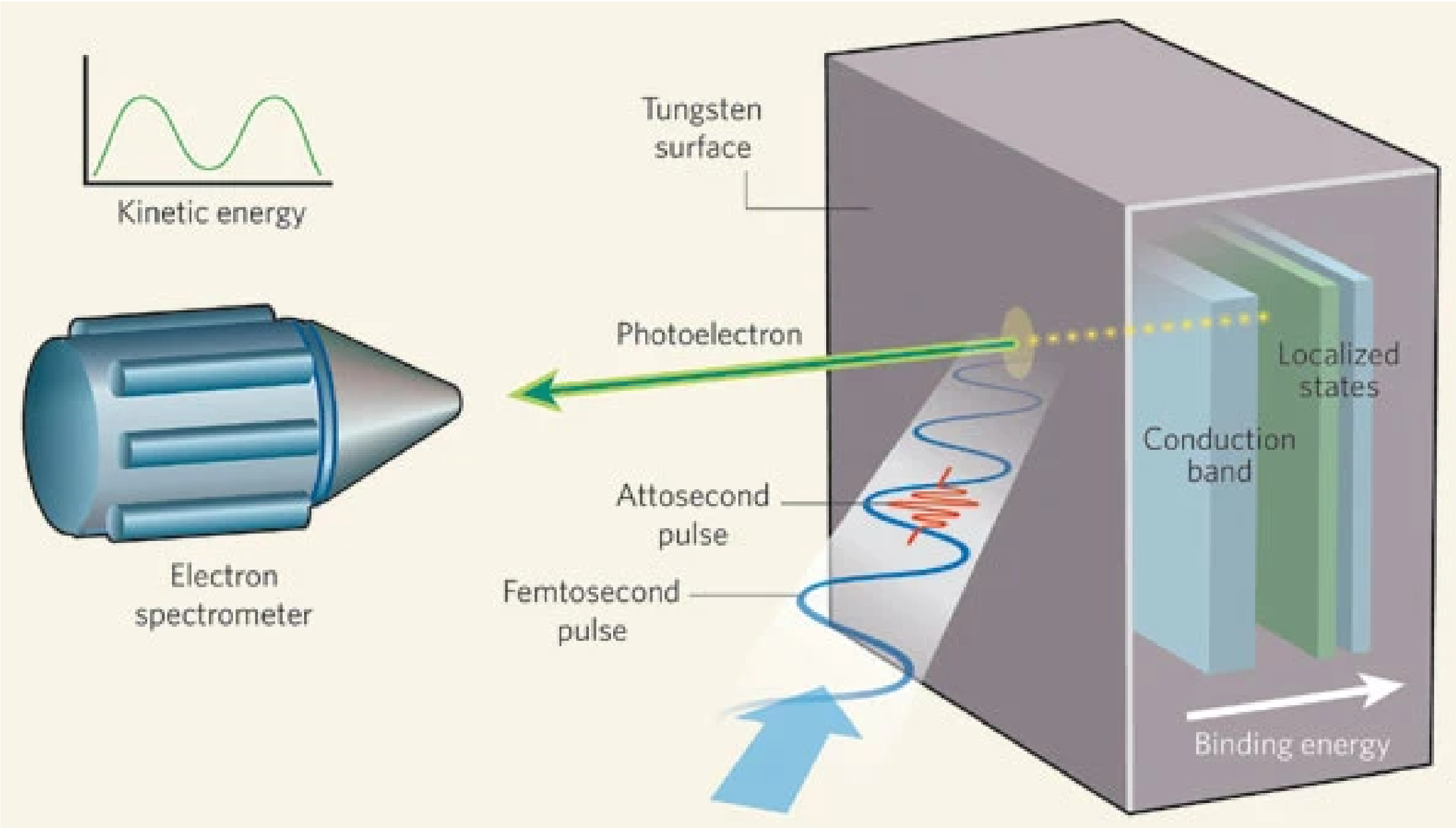

- in 2001, Krausz' group managed to create **isolated pulse of 650 as duration** (530 as in simulation)

# Attosecond spectroscopy of liquid water




Jordan et al., Science 369, 974 (2020)

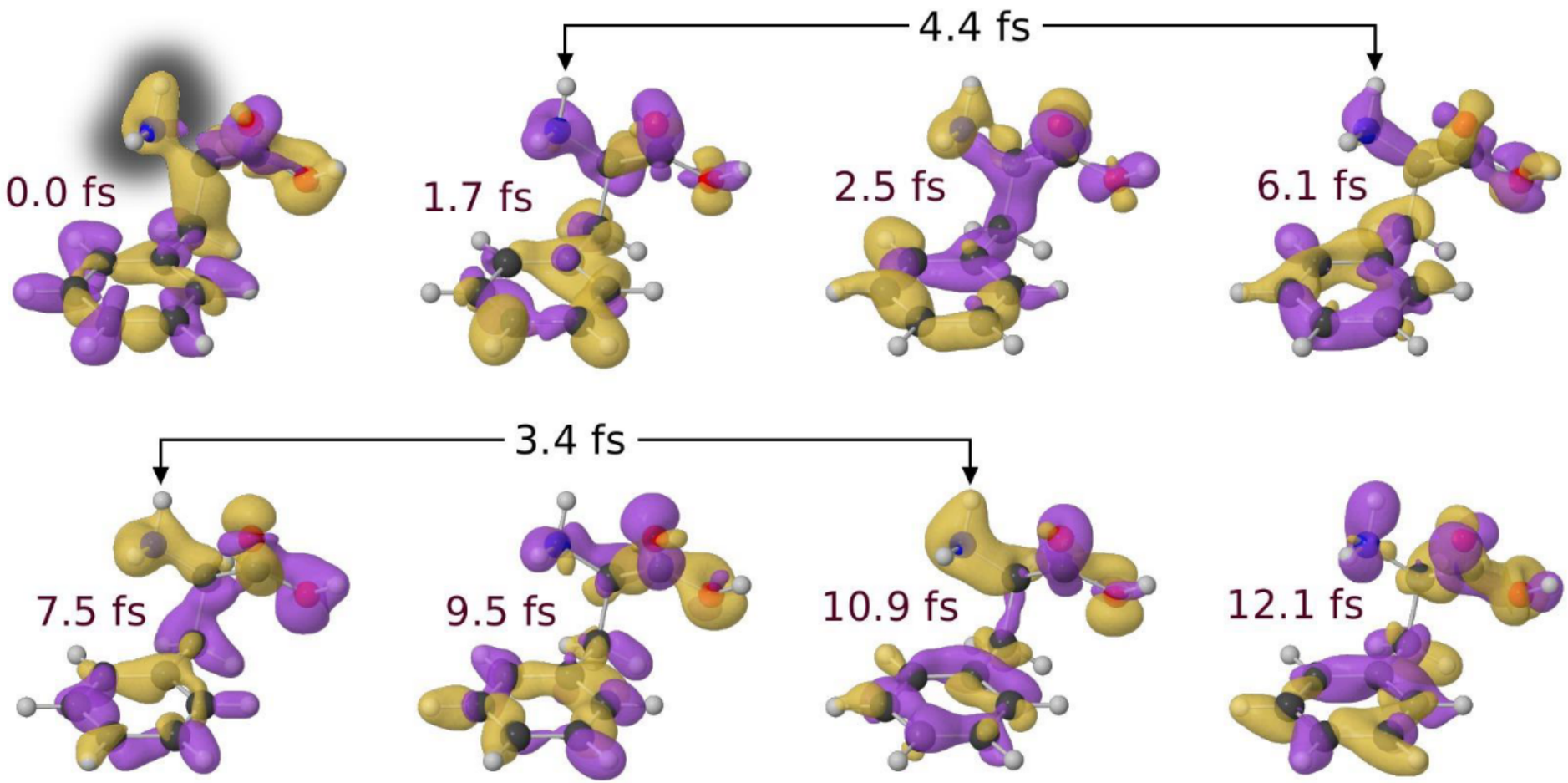
# Dynamics of the photoelectric effect



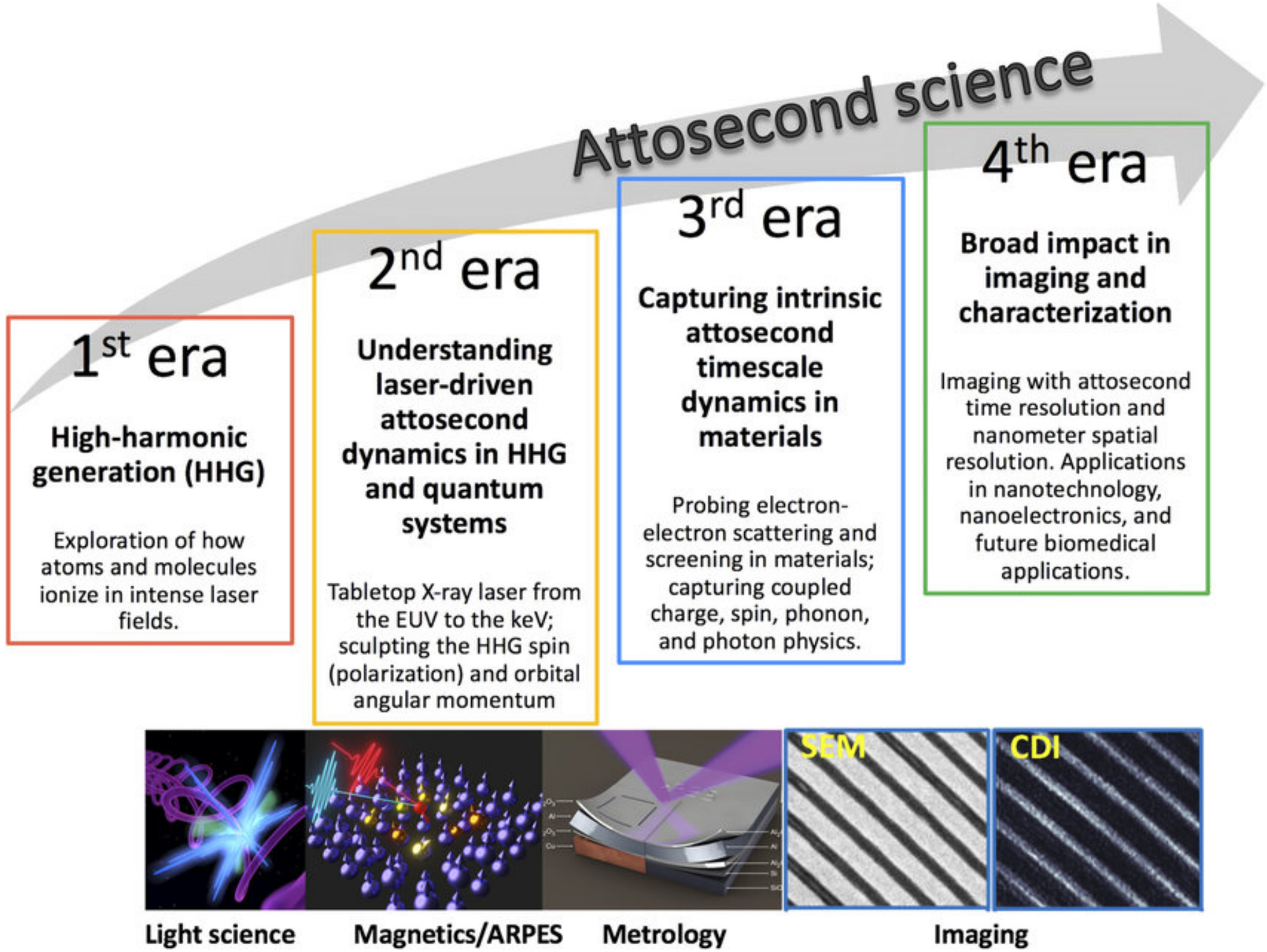

Cavalieri et al., Nature 449, 1029 (2007)

# Charge migration in bio-relevant molecules

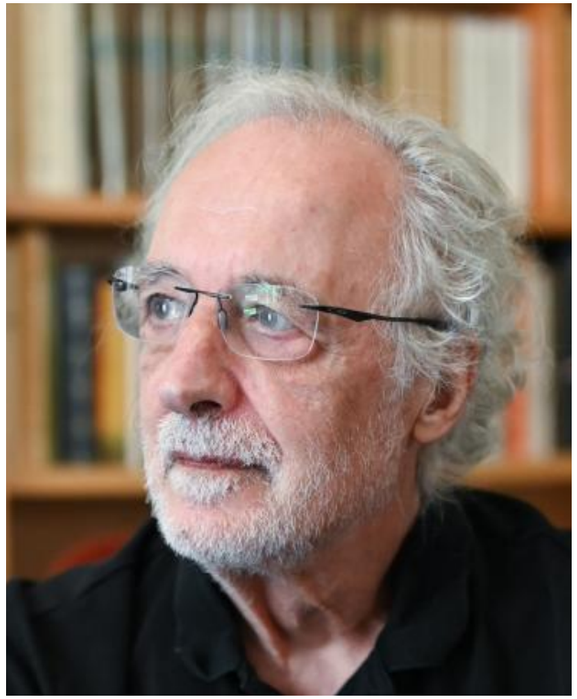
## Electron dynamics in phenylalanine initiated by attosecond pulses




Calegari et al., Science 346, 336 (2014)


Attosecond science
1st era
High-harmonic generation (HHG)
Exploration of how atoms and molecules ionize in intense laser fields.
2nd era
Understanding laser-driven attosecond dynamics in HHG and quantum systems
Tabletop X-ray laser from the EUV to the keV; sculpting the HHG spin (polarization) and orbital angular momentum
3rd era
Capturing intrinsic attosecond timescale dynamics in materials
Probing electron-electron scattering and screening in materials; capturing coupled charge, spin, phonon, and photon physics.
4th era
Broad impact in imaging and characterization
Imaging with attosecond time resolution and nanometer spatial resolution. Applications in nanotechnology, nanoelectronics, and future biomedical applications.
SEM
CDI
Light science
Magnetics/ARPES
Metrology
Imaging

# 2023 Physics Nobel Prize



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The Ohio State University, USA

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*"for experimental methods that generate attosecond pulses of light for the study of electron dynamics in matter"*