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

Nobel Prize in Physics (2023), awarded to:
Pierre Agostini
Ferenc Krausz
Anne L’Huillier

Luka
11 January 2024

Contents

- Introduction: why do we need short light pulses?
- How to generate them: high harmonics generation
- How to measure them: RABBIT
- Applications
  - Watching electron wavefunction oscillations.
  - Early cancer detection.

Further reading:

- Press release article, specifically the scientific background.
- Nobel lecture on youtube.
- Specific papers referenced in the slides.

Getting the Nobel prize

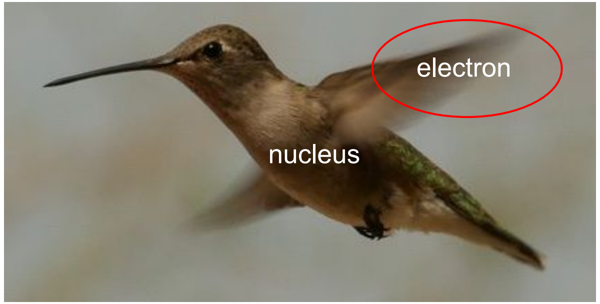
Having to explain your work to a general audience.

Having your life's work respectlessly reduced to less than 20 minutes by a random HEP student who understands nothing of photonics.

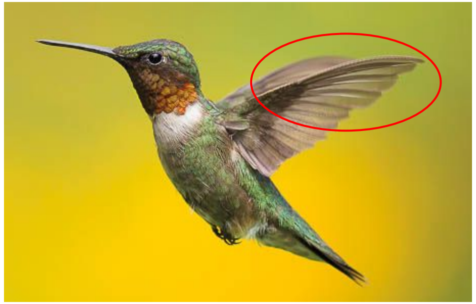
Ferenc Krausz during his Nobel prize lecture.

Short light pulses: why?

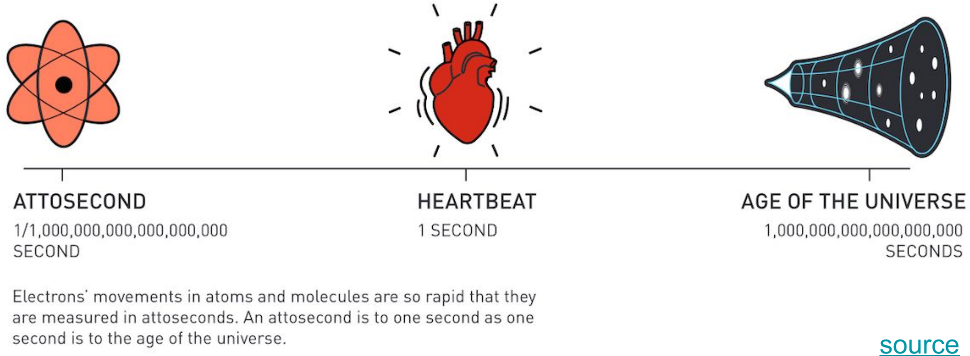
Slow shutter speed



Fast shutter speed



1 attosecond
= 1/1000 femtosecond
= 10^{-18} second



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Femtoseconds vs attoseconds

Femtosecond (10^{-15} s)

- Typical time scale of molecular vibrations and chemical reactions.
- Study of crystal vibrations, chemical transition states, molecular dynamics.

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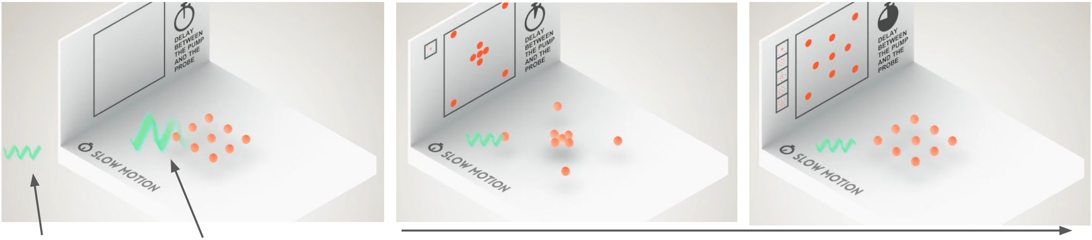


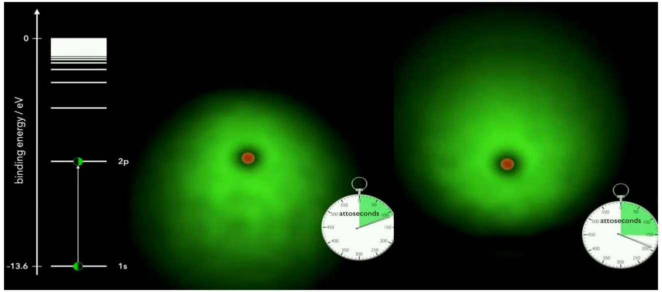
probe pump Repeat with varying delay between pump and probe

- Femtosecond pulses can be generated with ‘normal’ continuous lasers and clever optics (mode locking).
- Fundamental limit: almost-single-cycle pulse.

Further reading: A. Zewail, *Femtochemistry: Atomic-Scale Dynamics of the Chemical Bond*, J. Phys. Chem. A 2000, 104, 24, 5660-5694 (also Nobel prize in chemistry in 1999).

Attosecond (10^{-18} s)

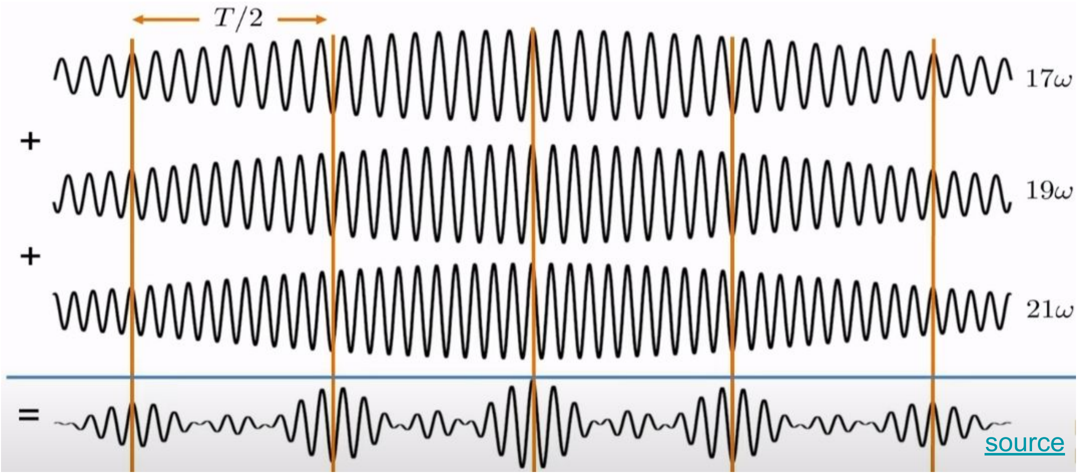
- Typical time scale of electron dynamics in atoms and molecules.



Dipole oscillations in 1s2p superposition. source

The basic principle: amplitude modulation by superposition

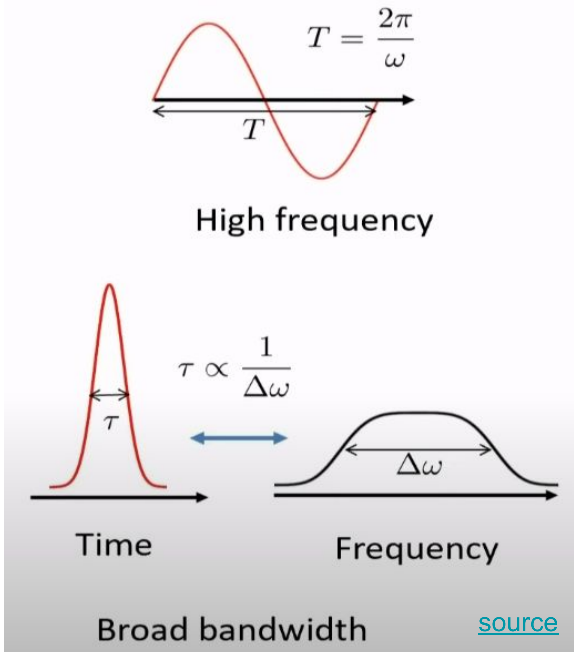
Superposition of waves with slightly different frequencies



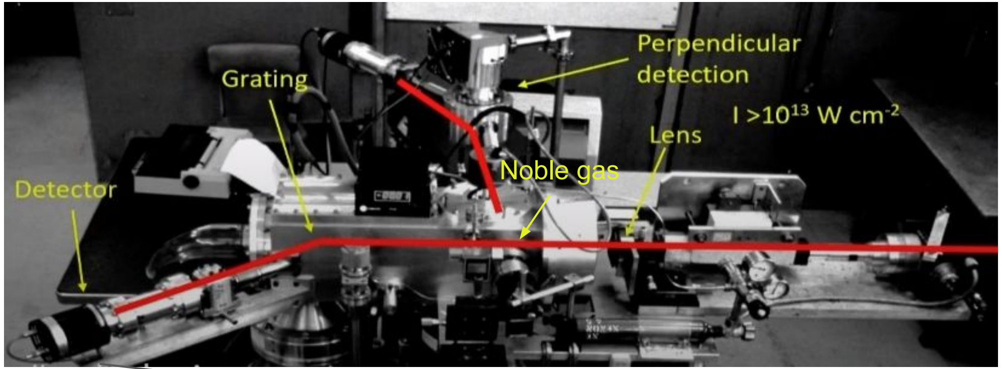
Problem: traditional lasers are unsuitable due to relatively low frequency (usually infra-red) and narrow bandwidth.

Solution: use superposition of higher-order harmonics, conveniently generated by HHG mechanism (see next slides).

Requirements for short pulses



High-harmonic generation



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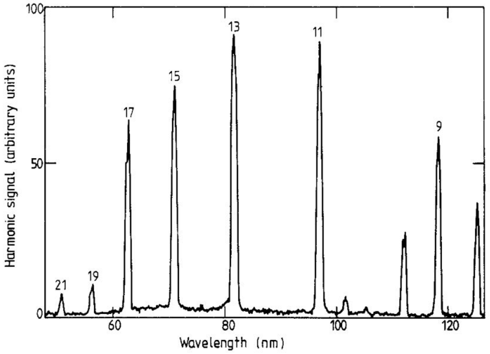
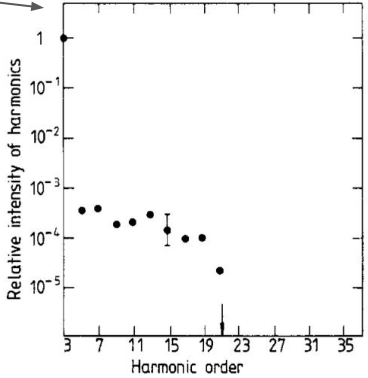
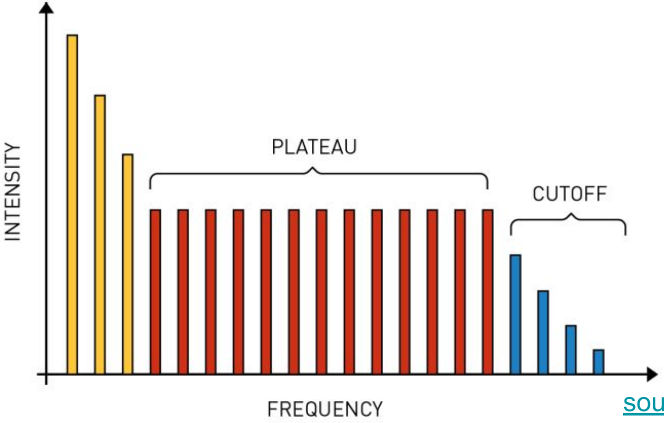




Conclusions:

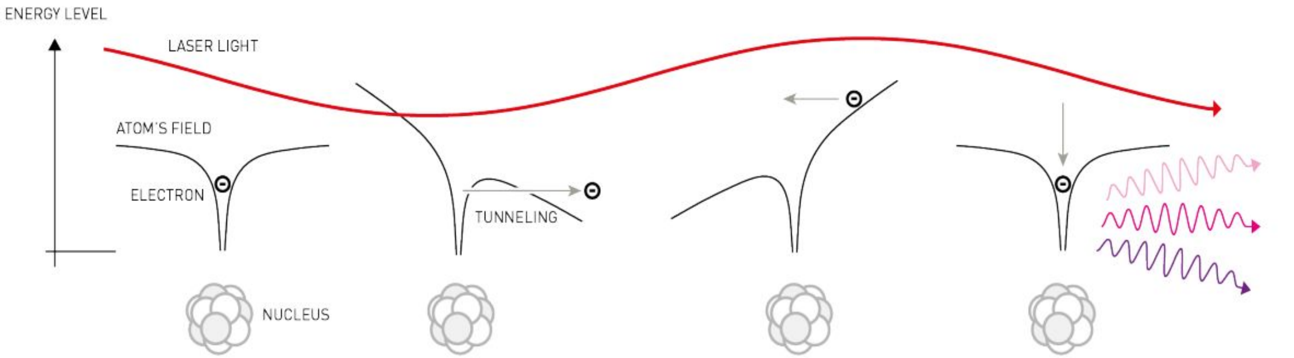
- Surprising 'plateau' of higher harmonics contributing with comparable intensity.
- Turn out to be phase-matched aka coherent (i.e. they are like lasers).
- Good candidate for making pulses by superposition.

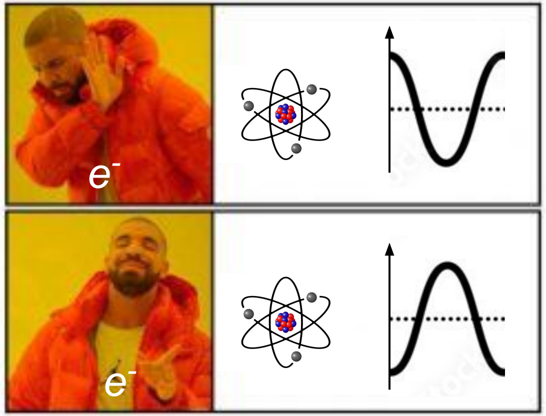


source

Further reading: A. L'Huillier et al., Multiple-harmonic conversion of 1064 nm radiation in rare gases, 1988 *J. Phys. B: At. Mol. Opt. Phys.* **21** L31.

The rescattering model for high-harmonic generation





<u>The 'rescattering' or 'three-step model'</u>:

- Tunneling ionization.
- Acceleration of free electron away from remaining ion.
- On phase shift of the laser field, free electron recombines with ion, converting kinetic energy into photons.

Note: model yields correct quantitative results both with semi-classical and fully quantum mechanics approach.

The RABBIT technique for measuring attosecond pulse duration

RABBIT: reconstruction of attosecond beating by interference of two-photon transitions

- Superimpose attosecond pulse train with original IR laser, with tunable time delay.
- Illuminate a material with this superimposed laser beam to generate photoelectrons.
- Measure kinetic energy of photoelectrons, proportional to energy of absorbed photons.

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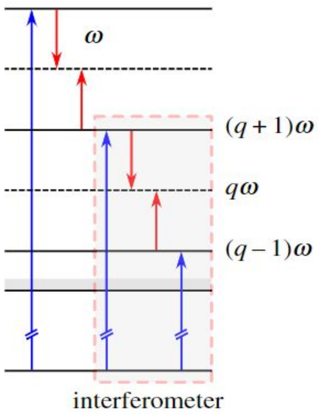

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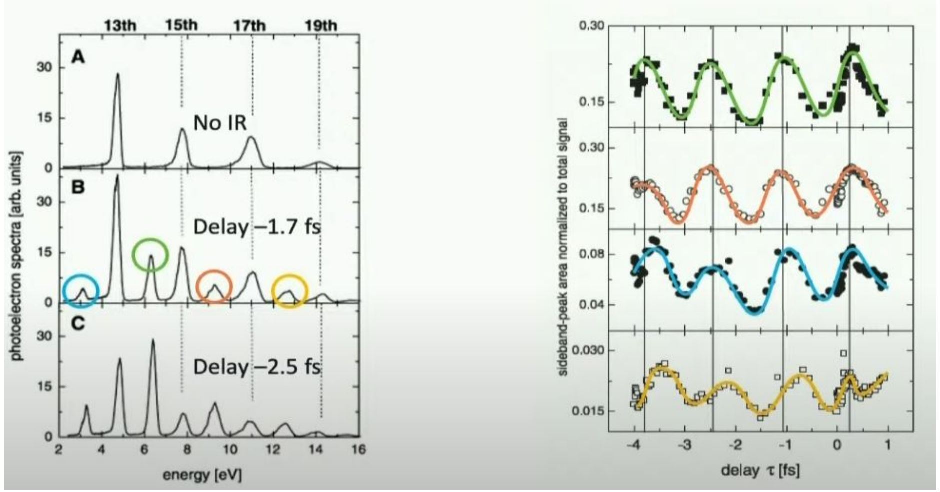
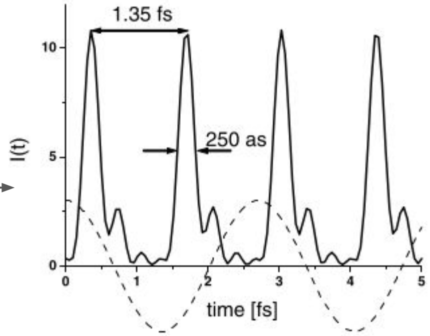
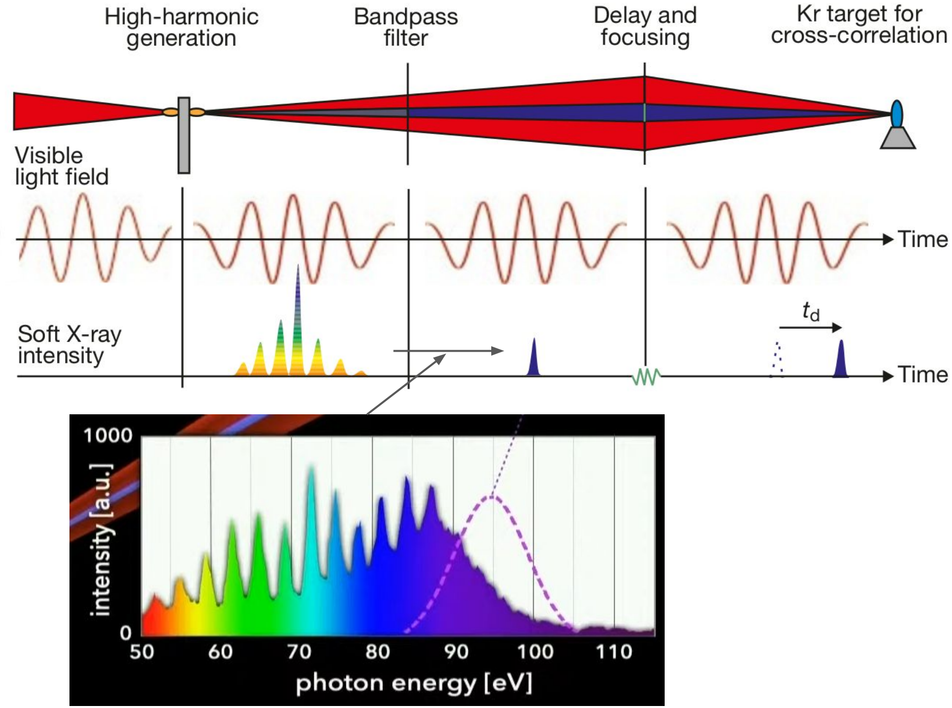




Further reading: P. Agostini et al., *Observation of a Train of Attosecond Pulses from High Harmonic Generation*, Science **292**, 1689-1692 (2001).

Single pulse generation



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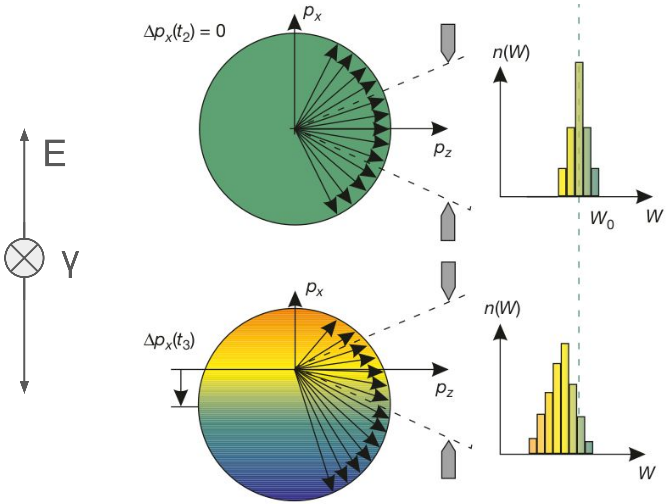
Differences w.r.t. train of pulses:

- Use extremely short (O(fs)) visible light laser pulse instead of multicycle infrared laser pulse as driver.
- Bandpass filter to remove lower frequencies.

Different measurement method:



Further reading: F. Krausz et al., Attosecond metrology. *Nature* **414**, 509–513 (2001).

Application: watching electron wavefunction oscillations

Single-cycle “conventional” laser pulse for ionization.

Attosecond HHG pulse for characterization, with tunable time delay w.r.t. ionization pulse.

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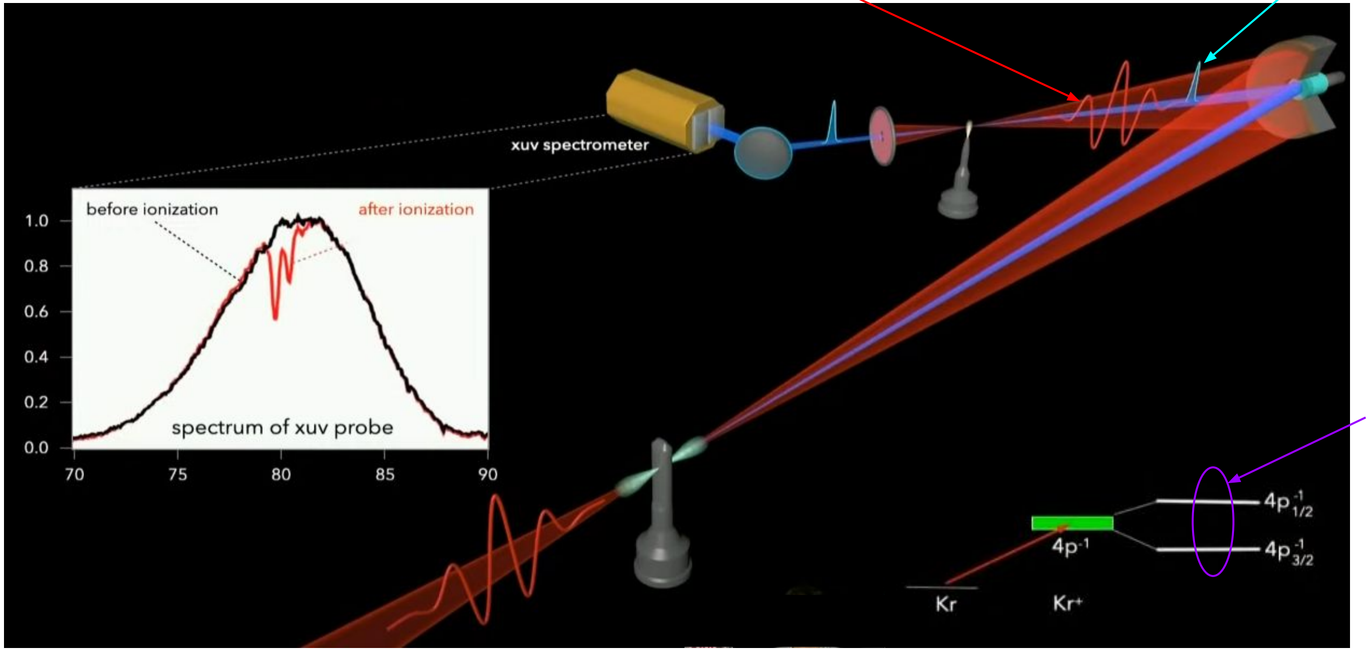
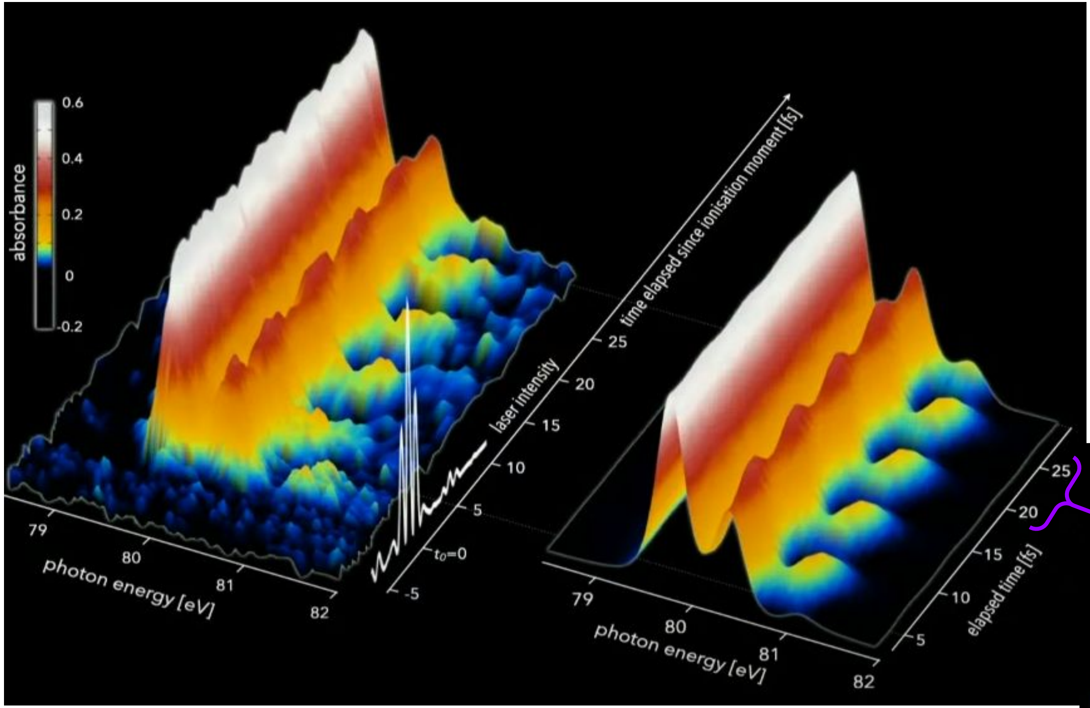


The hole (e^- deficiency) is created in a superposition of two states → oscillation!

Application: watching electron wavefunction oscillations (2)



Measurement

Prediction

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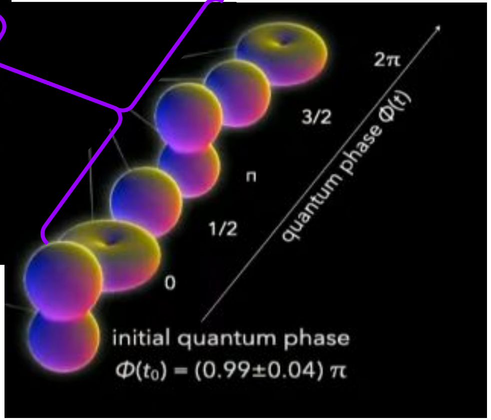
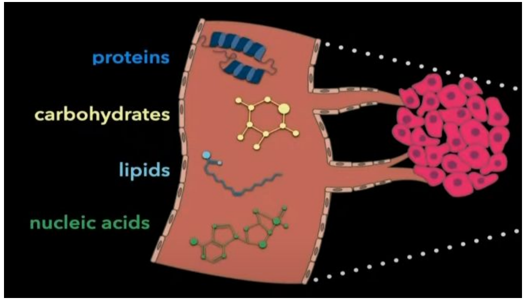
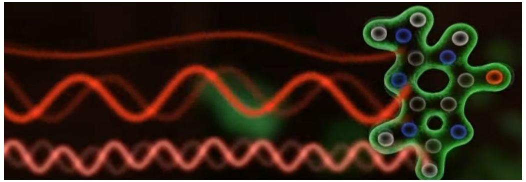
1 period = 6 fs

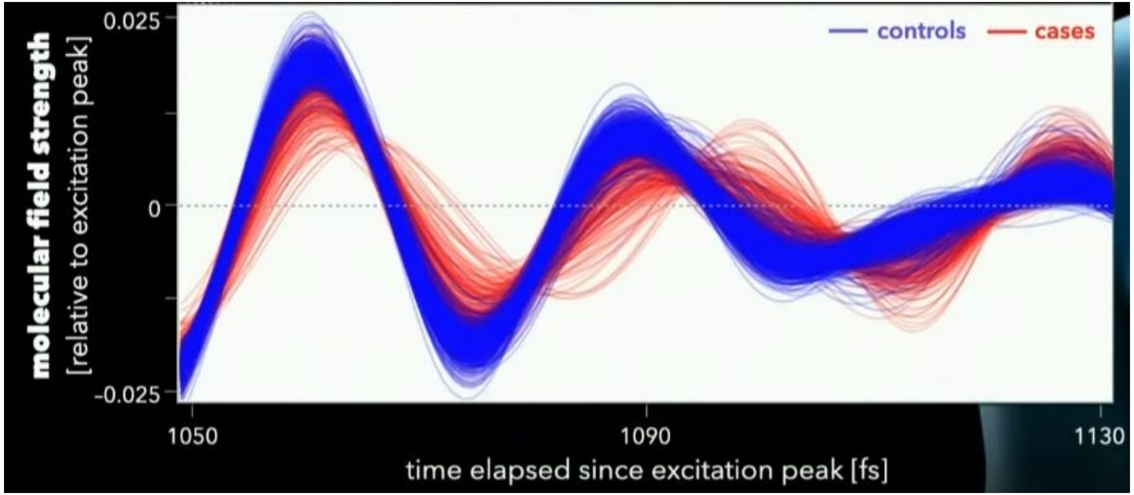


Application: early cancer detection

Spectrogram of full molecular content of blood plasma







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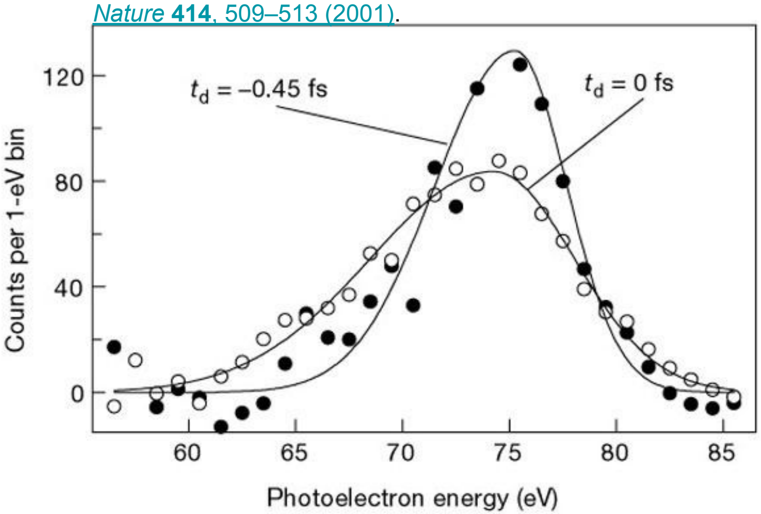
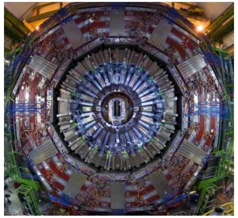
Excitation with single-cycle IR laser pulse.

Full time-domain response excitation captured with attosecond pulse scanning.

The end



"Clearly some systematics are missing"



"This is an excellent fit"