Experimental realization of attosecond pulses for probing electron dynamics in matter

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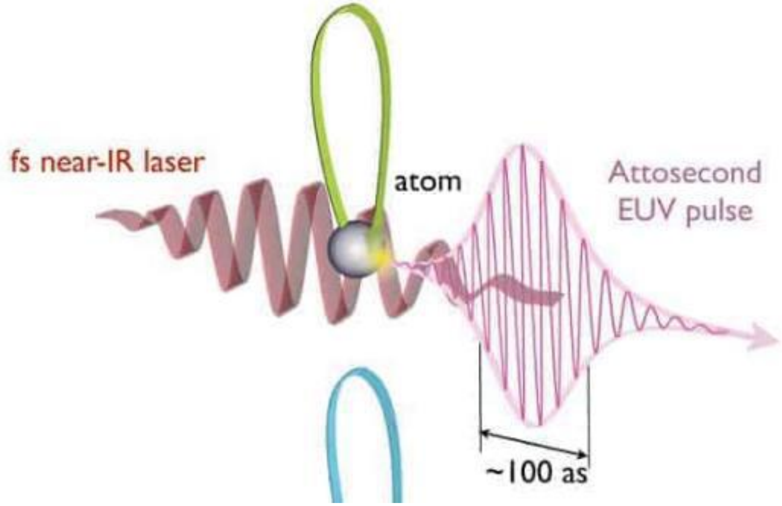


The Nobel Prize in Physics 2023

Ill. Niklas Elmehed © Nobel Prize Outreach

Pierre Agostini

Prize share: 1/3

Ill. Niklas Elmehed © Nobel Prize Outreach

Ferenc Krausz

Prize share: 1/3

Ill. Niklas Elmehed © Nobel Prize Outreach

Anne L'Huillier

Prize share: 1/3

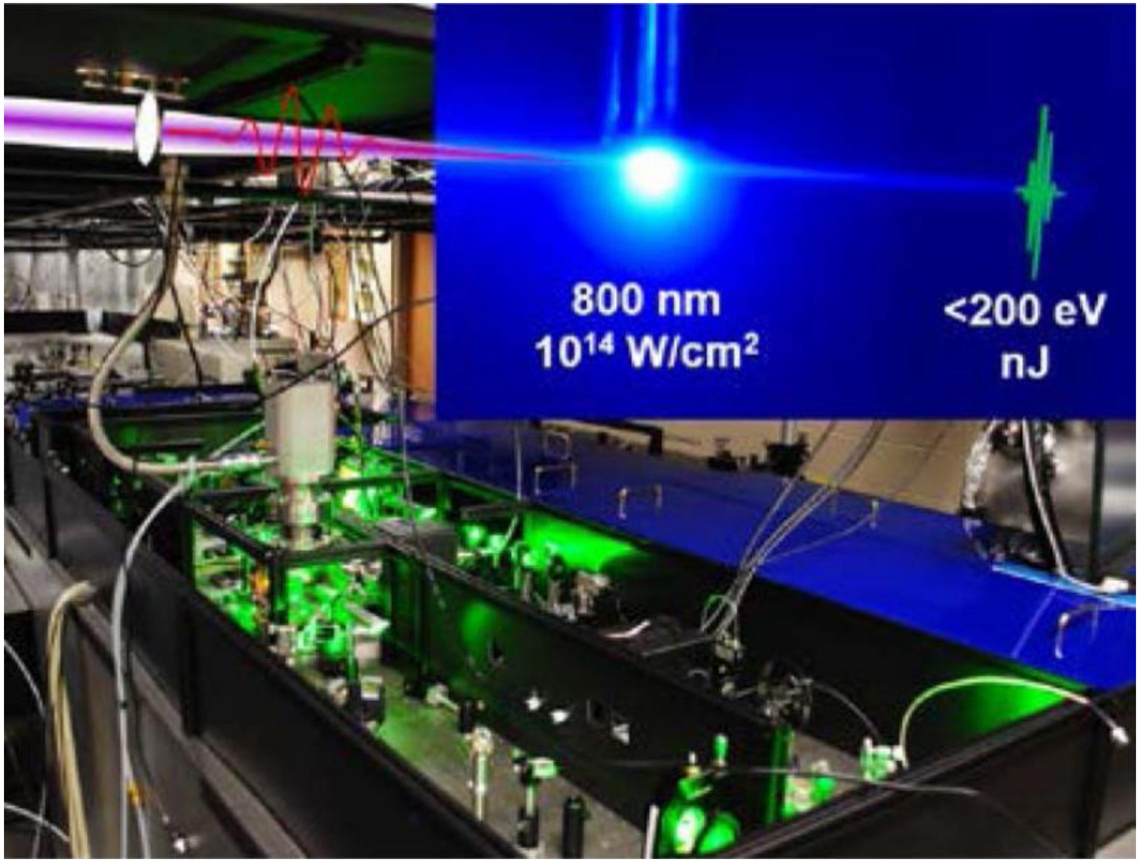
The Nobel Prize in Physics 2023 was awarded to Pierre Agostini, Ferenc Krausz and Anne L'Huillier "for experimental methods that generate attosecond pulses of light for the study of electron dynamics in matter"

This year's laureates have created flashes of light that are short enough to take snapshots of electrons' extremely rapid movements.

Anne L'Huillier discovered a new effect from laser light's interaction with atoms in a gas.

Pierre Agostini and **Ferenc Krausz** demonstrated that this effect can be used to create shorter pulses of light than were previously possible!

Outline



Attosecond Pulses!

- **What is attosecond, and why bother?**
- **Why was it difficult to breach the femtosecond barrier?**
- **What new ideas were invoked?**
- **How the breakthrough was achieved?**
- **What is the relevance in the context of modern-day science?**
- **What next?**

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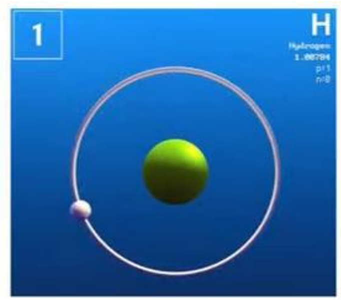
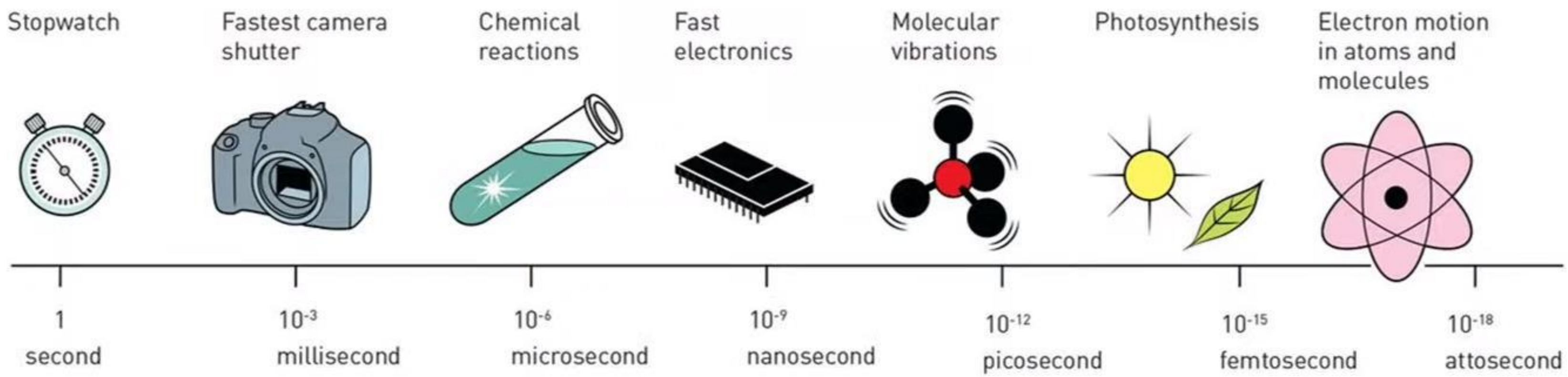
Time scales of various phenomena

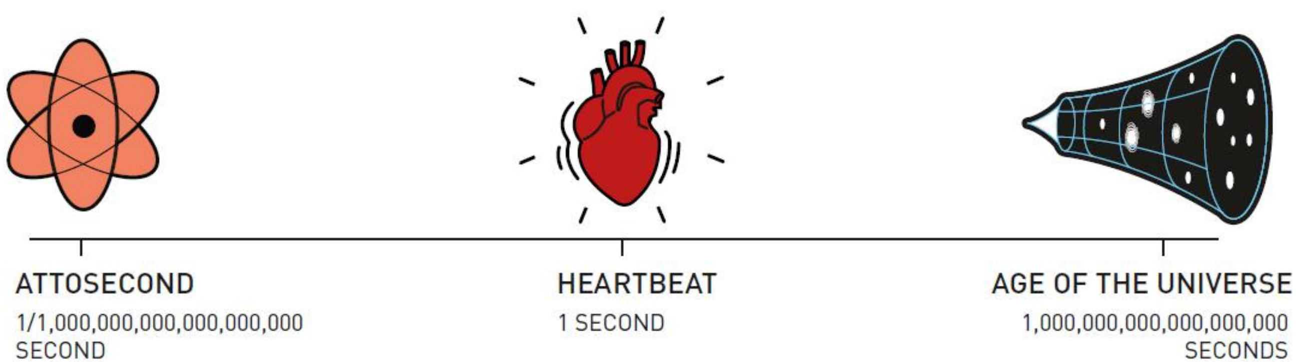


Bohr orbit time (n = 1)

$$\tau = \frac{2\pi a_0}{v} = \frac{2\pi a_0}{\alpha c}$$

$$\tau = \frac{2\pi a_0}{c/137} = 152 \text{ asec}$$





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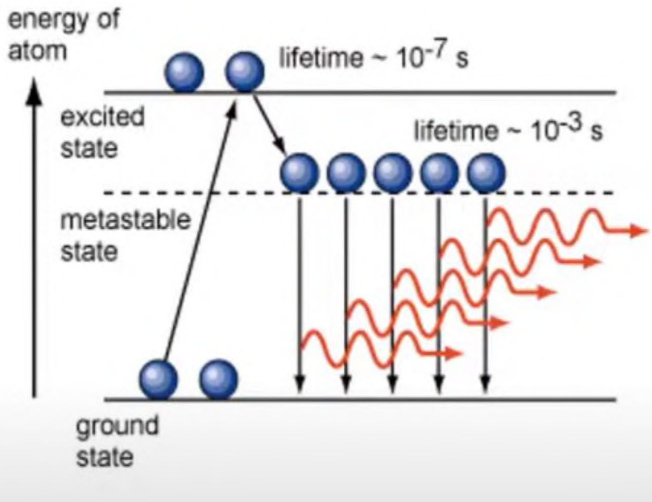
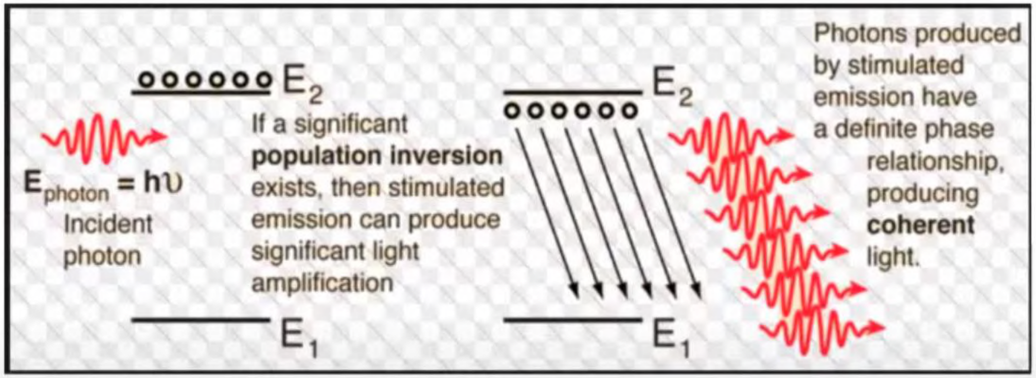
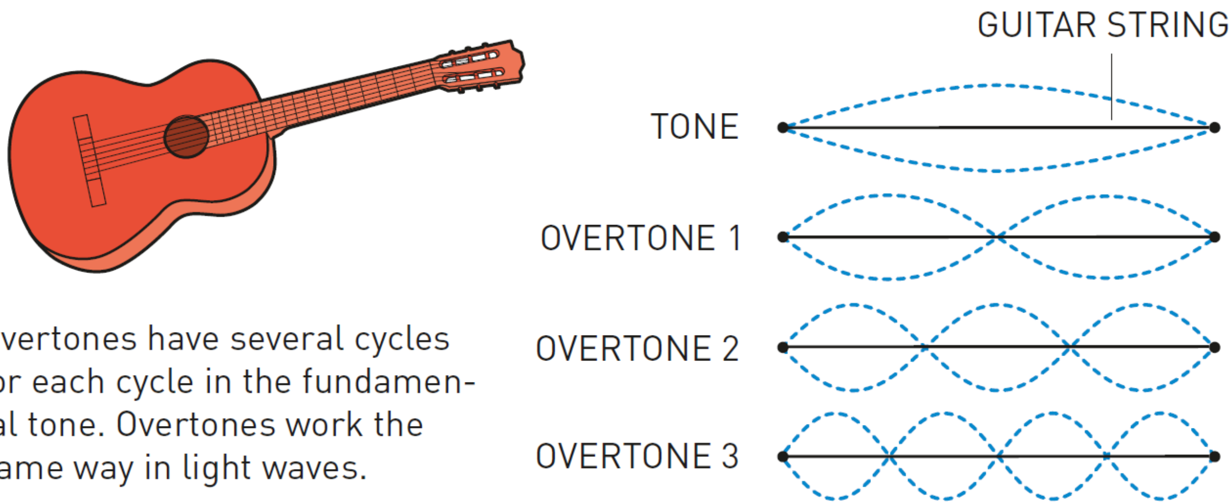
Lasers & Overtones







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

Timeline of Laser pulse evolution

CPA: Chirped Pulse Amplification
KLM: Kerr Lens Mode-Locking

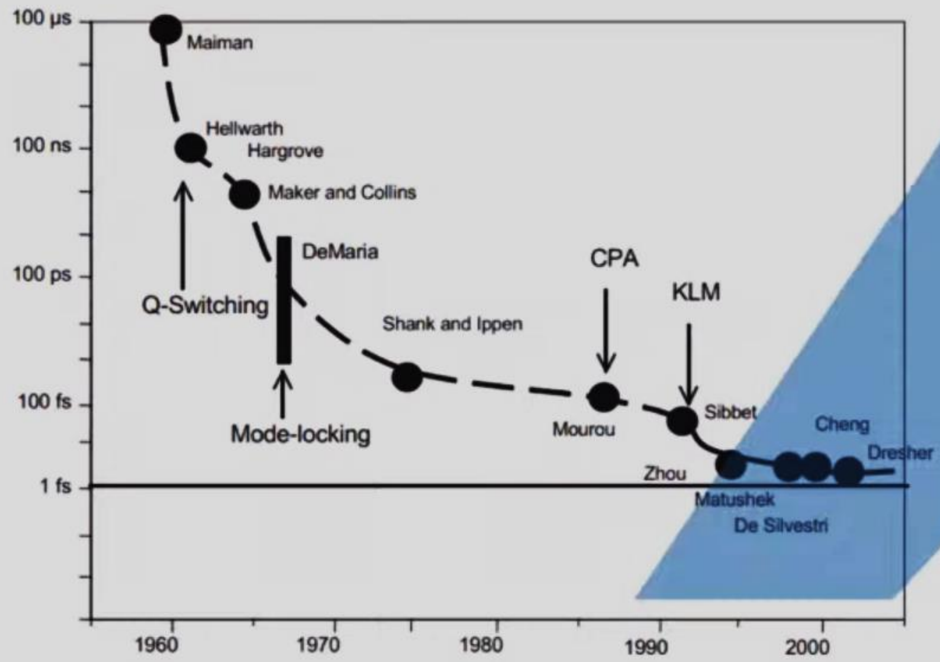


Figure 1. Timeline of the laser pulse length evolution from the free-running laser of Maiman to the recent refinements. A few names and techniques mark the essential steps in this evolution. The existence of a femtosecond barrier is clearly visible on the graph.

1987
Anne L'Huillier fires a laser at a noble gas to produce "overtones" of light.

1990s
L'Huillier and others explore the mechanism behind the overtones.

1994
Pierre Agostini begins to develop a pulse-measurement technique known as "RABBIT."

2001
Agostini produces a train of 250-attosecond pulses.
Ferenc Krausz isolates a single 650-attosecond pulse.

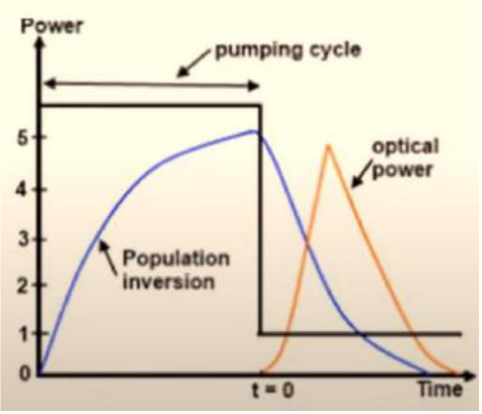
2000s
Attosecond pulses are used to explore the motion of electrons in various materials.

2023
L'Huillier, Agostini and Krausz awarded Nobel Prize in Physics.

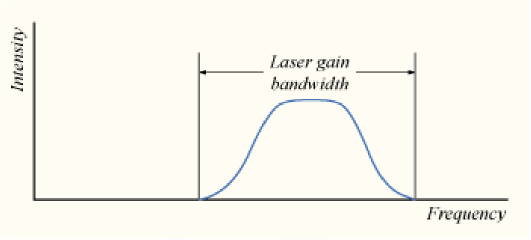
Pierre Agostini and Louis F DiMauro, Rep. Prog. Phys. 67 (2004) 813–855

Q-switching & Mode Locking techniques (ns to ps)

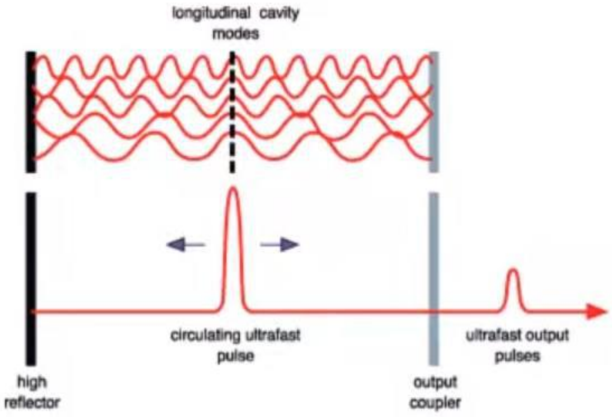
Q-switching: Q-switching is a method for generating intense short pulses (sometimes called "giant pulses") of light with a laser. In the first phase, the gain medium is pumped, while the extraction of energy as laser light is prevented by keeping the resonator losses high (that is, the Q factor is kept low). The resonator losses are then suddenly reduced. As the gain is then substantially higher than the resonator losses, the intracavity power rises exponentially.



Mode Locking: The basis of the technique is to induce a fixed phase relationship between the longitudinal modes of the laser's resonant cavity due to standing waves. Constructive interference between these modes can cause the laser light to be produced as a train of pulses. The laser is then said to be "phase-locked" or "mode-locked".

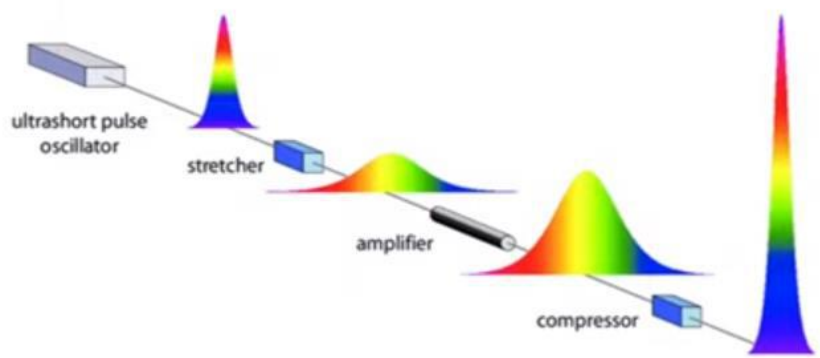


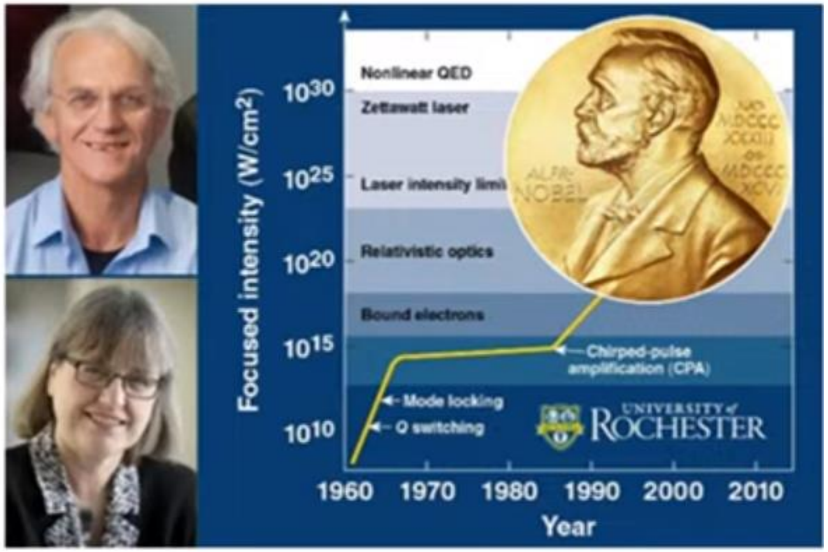
A typical helium–neon laser has a gain bandwidth of about 1.5 GHz (a wavelength range of about 0.002 nm at a central wavelength of 633 nm, lasing modes are 3), whereas a titanium-doped sapphire (Ti:sapphire) solid-state laser has a bandwidth of about 128 THz (a 300 nm wavelength range centered at 800 nm with 250000 modes).



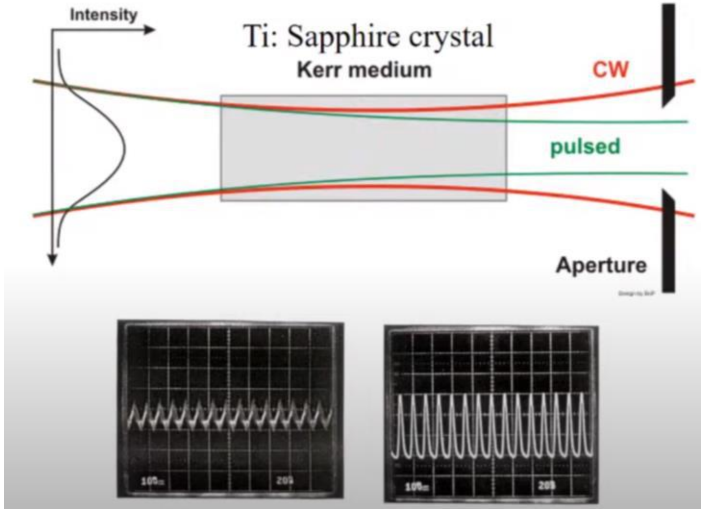
Chirped Pulse Amplification & Kerr Mode Locking (fs)

Chirped Pulse Amplification: First stretch the pulse, then amplify, and now compress! 2018 Nobel Prize in Physics, Gerard Mourou & Donna Strickland, Univ. of Rochester)





Kerr Lens Mode Locking: Because of the non-uniform power density distribution in a Gaussian beam the refractive index changes across the beam profile; the refractive index experienced by the beam is greater in the center of the beam than at the edge. Thus a rod of an active Kerr medium functions as a lens for high intensity light. In the laser cavity short bursts of light will then be focused differently from continuous waves.



Input and Mode-Locked Output

Inspired by public lecture on "Nobel Prize in Physics" by Prof. Reji Philip, RRI Bangalore

The "femtosecond barrier"

- The Ti:Sapphire crystal has an emission bandwidth of about 100 THz (700 - 1000 nm), which can support a mode-locked laser pulse down to 4.5 fs (not less!)

- No other material has been found that has a larger bandwidth with high gain in the visible spectral region.

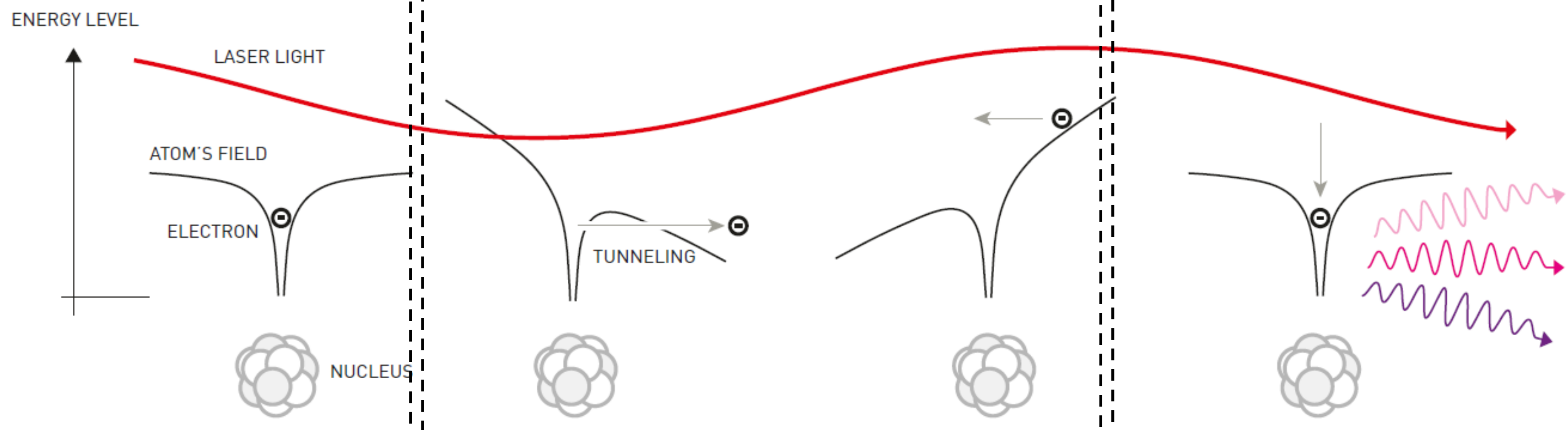
- The fs barrier can be beaten (for creating attosecond light pulses) only by finding a way to generate a series of periodically spaced frequencies that cover a much larger spectral range

- Two methods have been found to be useful for this:
  (i) High Harmonic Generation (HHG) in gases
  (ii) Cascaded Stimulated Raman Scattering

Inspired by public lecture on "Nobel Prize in Physics" by Prof. Reji Philip, RRI Bangalore

Laser light interacts with atoms in a gas

Ionization | Process after Ionization | Recombination



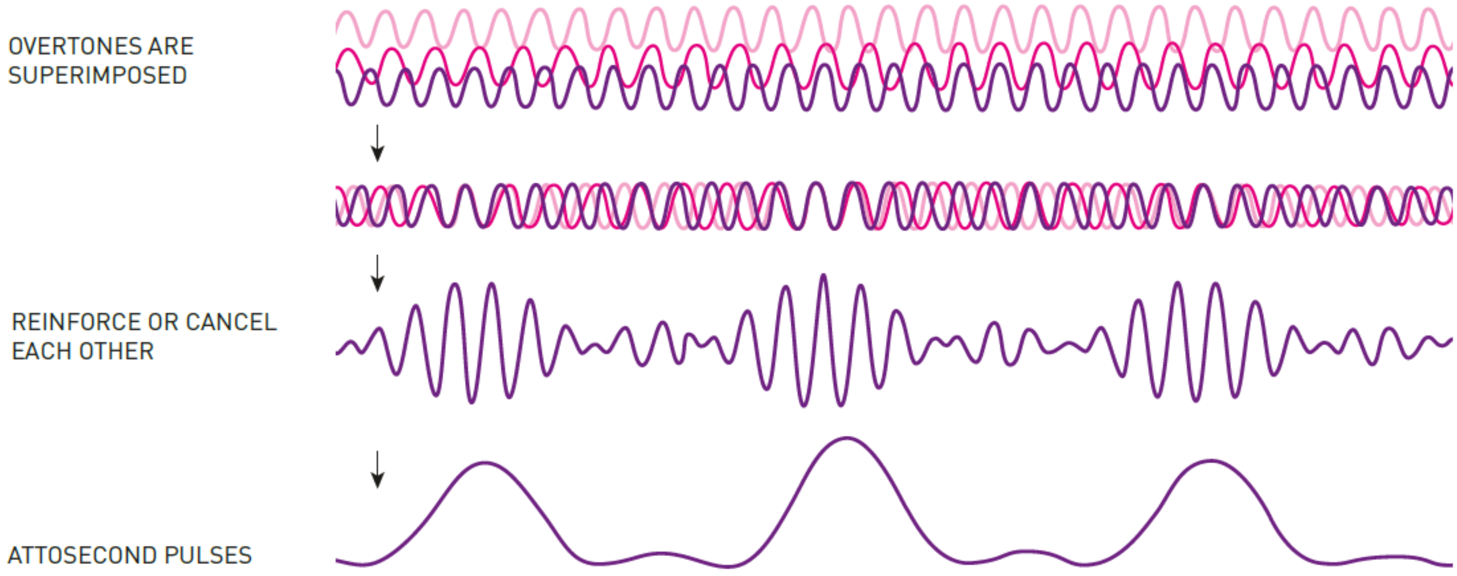
1 An electron that is bound to an atom's nucleus cannot normally leave its atom; it does not have enough energy to lift itself out of the well created by the atom's electrical field.

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.

Superposition of higher harmonics leads to attosecond pulses



Since light pulses are limited by their carrier frequency, visible pulses are bound to a few femtoseconds. XUV or x-ray pulses, on the other hand, allows the transition across the femtosecond border, entering the attosecond timescale.

Professor Anne L'Huillier

- Multiphoton ionization,
- High Harmonic Generation
- Contributed to full quantum theory of the process (along with Lewenstein & Corkum)
- Explanation of photo-ionization discrepancy by considering "shake-up" (Auger) electrons

Physical Review Letters

Multiply Charged Ions Formed by Multiphoton Absorption Processes in the Continuum

A. L'Huillier, L. A. Lompre, G. Mainfray, and C. Manus
Service de Physique des Atomes et des Surfaces, Centre d'Etudes Nucléaires de Saclay, F-91191 Gif-sur-Yvette Cedex, France

(Received 12 March 1982)

Singly, doubly, triply, and quadruply charged krypton ions are formed by multiphoton absorption processes in krypton atoms. They are induced by a 50-psec laser pulse at 1.064 μm in the 10^{13}–10^{14}-W-cm^{-2} intensity range. The percentage ratio between the numbers of doubly and singly ionized Kr atoms is 10% at 8×10^{13} W cm^{-2}. It is shown that Kr^{2+} ions result from a direct 33-photon absorption process. At 1.5×10^{13} W cm^{-2}, the 33-photon absorption rate is only about 60 times less than the 13-photon absorption rate which gives singly charged ions.

- The continuum component of the electron wave packet interferes with the component that remains bound to the parent ion. The coherent overlap of these two components generates an electric dipole.
- The centre of mass of the total electron distribution oscillates back and forth as the continuum wave function propagates.
- This electric dipole acts as an atomic antenna lasting just a small fraction of an optical cycle of the driving radiation, leading to high harmonic emission.

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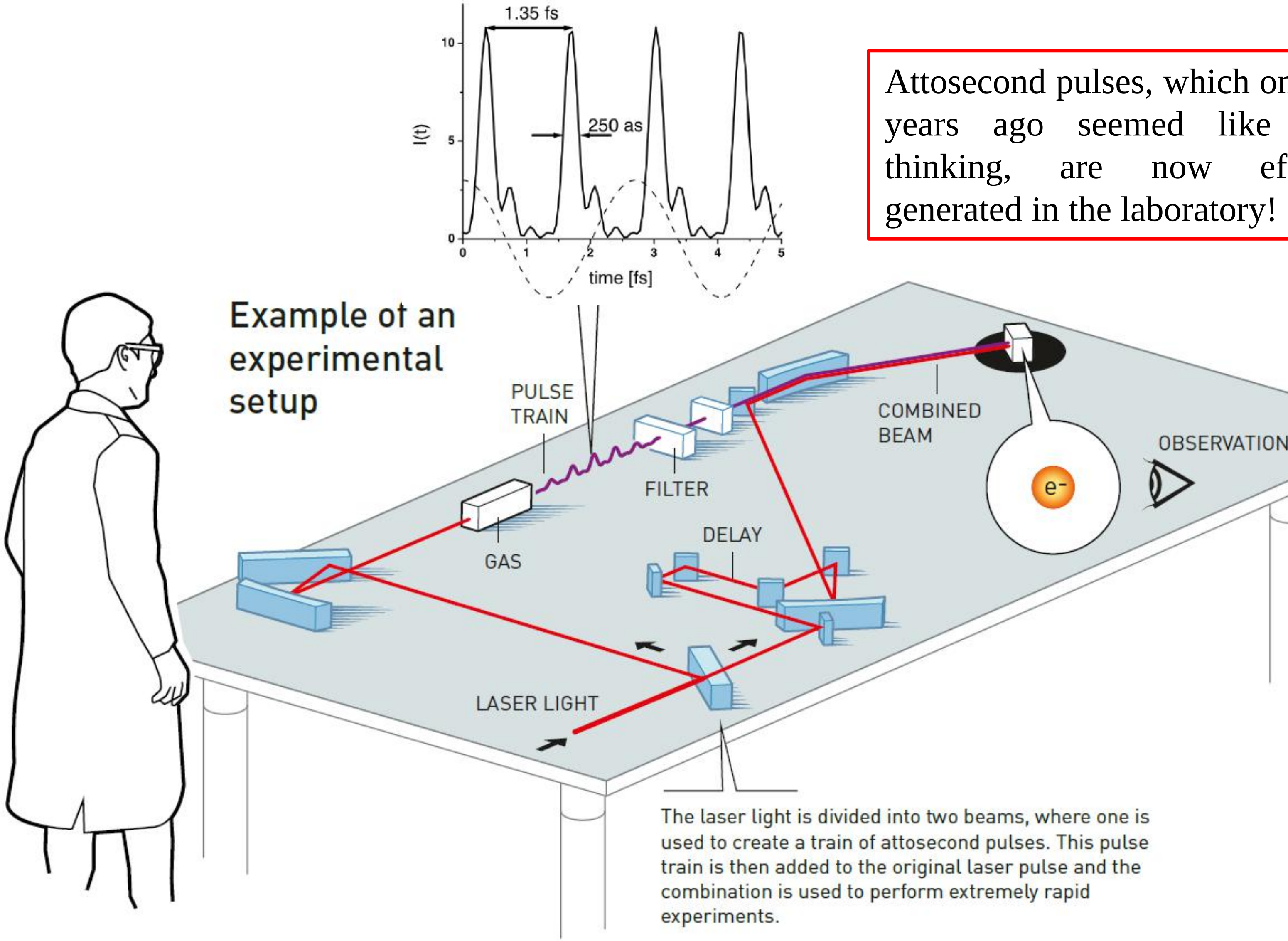
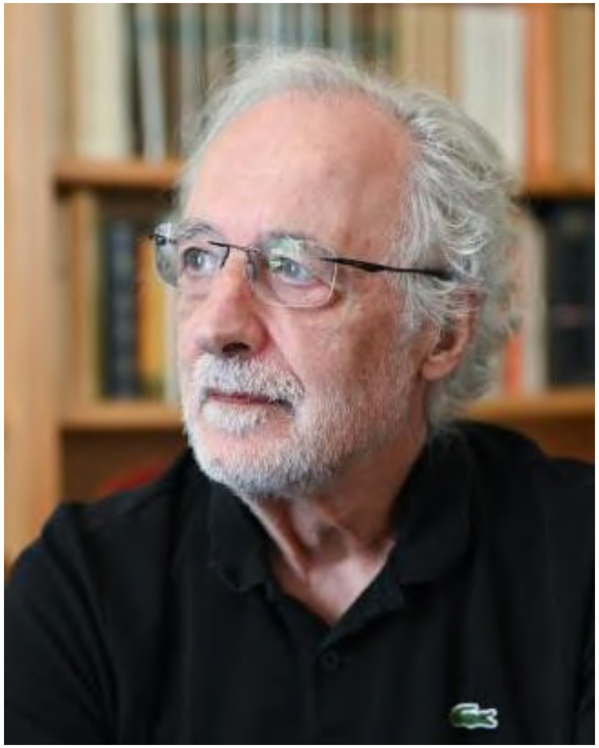
Experimental realization: typical experimental

Attosecond pulses, which only a few years ago seemed like wishful thinking, are now effectively generated in the laboratory!



Professor Pierre Agostini



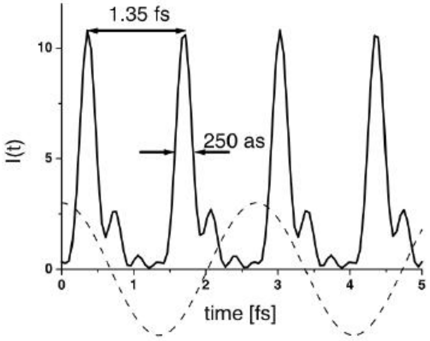
- Experimental realization of a train of attosecond pulses
- Above Threshold Ionization (ATI)
- Investigation of the principle of frequency modulation in a two-color photon field (RABBITT – Reconstruction of Attosecond Beating by Interference of Two-photon Transitions)

Science

Observation of a Train of Attosecond Pulses from High Harmonic Generation

P. M. Paul,[1] E. S. Toma,[2] P. Breger,[1] G. Mullot,[3] F. Augé,[3] Ph. Balcou,[3] H. G. Muller,[2*] P. Agostini[1]

In principle, the temporal beating of superposed high harmonics obtained by focusing a femtosecond laser pulse in a gas jet can produce a train of very short intensity spikes, depending on the relative phases of the harmonics. We present a method to measure such phases through two-photon, two-color photoionization. We found that the harmonics are locked in phase and form a train of 250-attosecond pulses in the time domain. Harmonic generation may be a promising source for attosecond time-resolved measurements.



Professor Ferenc Krausz

- Few-cycle laser pulse generation
- Broad HH spectrum generation
- Generation of 1st isolated attosecond light flashes and its measurement using cross-correlation
- Application of attosecond to record electron dynamics

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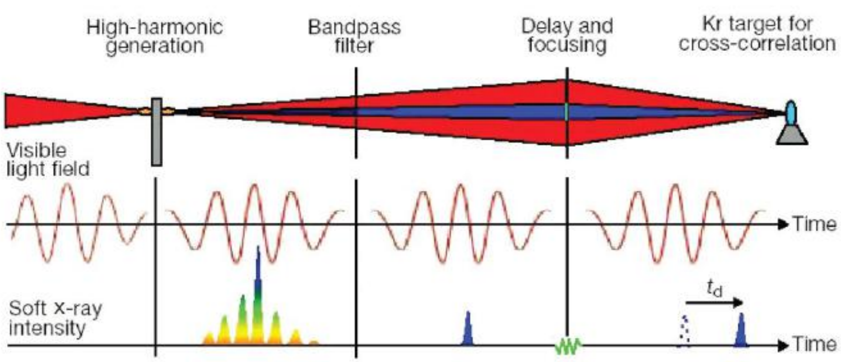
Article | Published: 29 November 2001

Attosecond metrology

M. Hentschel, R. Kienberger, Ch. Spielmann, G. A. Reider, N. Milosevic, T. Brabec, P. Corkum, U. Heinzmann, M. Drescher & F. Krausz

Nature **414**, 509–513 (2001) | Cite this article



What next?!

Ultrafast laser pulses allow physicists and chemists to watch fast molecular motion as it happens. But some fundamental atomic processes are even faster! Stability & optimization of phase matching is still very challenging!

Some works are being done in the fields of:

Attosecond transient absorption spectroscopy

Charge Migration in molecules

Charge transfer in organic photovoltaics

nature › nature physics › news & views › article

News & Views | Published: 03 August 2023

Optical physics

Quantum optics meets attosecond science

Dong Hyuk Ko & P. B. Corkum

Nature Physics (2023) | Cite this article

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Generating high harmonics or attosecond pulses of light is normally thought of as a classical process, but a theoretical study has now shown how the process could be driven by quantum light.

Thank you!