Science Lunch FZU, 18. 1. 2024

The Nobel Prize in Physics 2023:

high-order harmonics and attosecond pulse generation

Jaroslav Nejdl

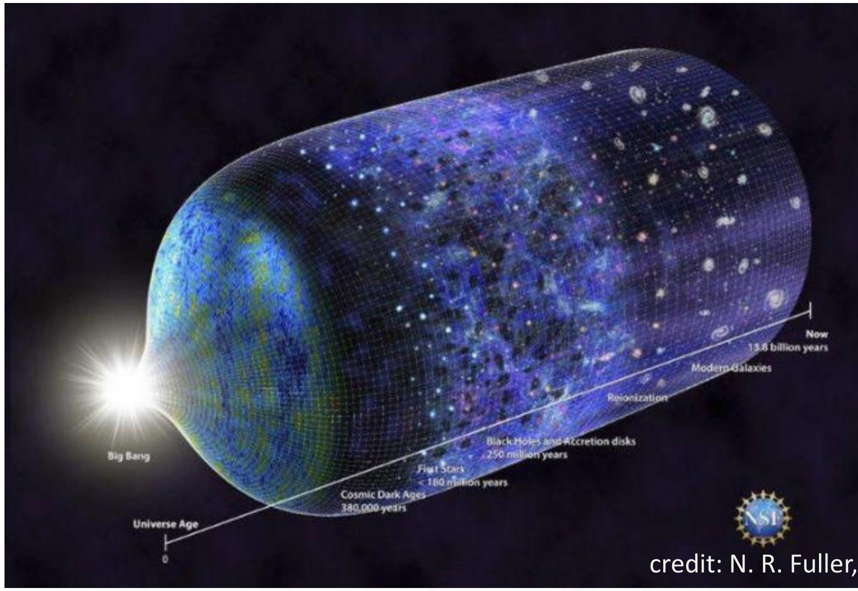
ELI Beamlines facility, Extreme Light Infrastructure ERIC

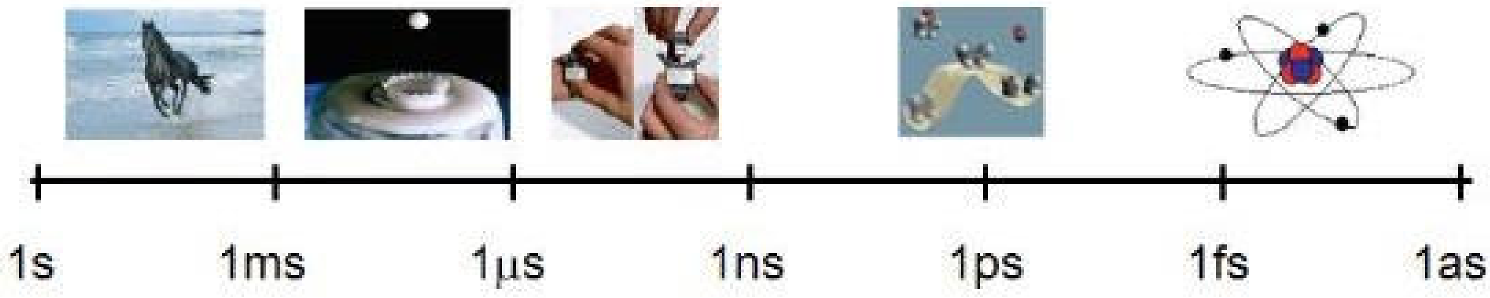
Dolní Břežany, Czech Republic


THE NOBEL PRIZE
IN PHYSICS 2023
Pierre Agostini
The Ohio State University, USA
Ferenc Krausz
Max Planck Institute of Quantum Optics & Ludwig-Maximilians-Universität München, Germany
Anne L'Huillier
Lund University, Sweden
Pierre Agostini
Ferenc Krausz
Anne L'Huillier
"for experimental methods that generate attosecond pulses of light for the study of electron dynamics in matter"
THE ROYAL SWEDISH ACADEMY OF SCIENCES

Time scales of our world

- **Universe:**
  14 billion years = $4x10^{17}$s
- **Solar system**
  165 years = $5x10^{9}$ s
- **Human heart beat** 1 s





1 as = 10^{-18} s

Exploring fast processes

- **We need to record the fast evolving objects still – short exposure**

Exploring fast processes

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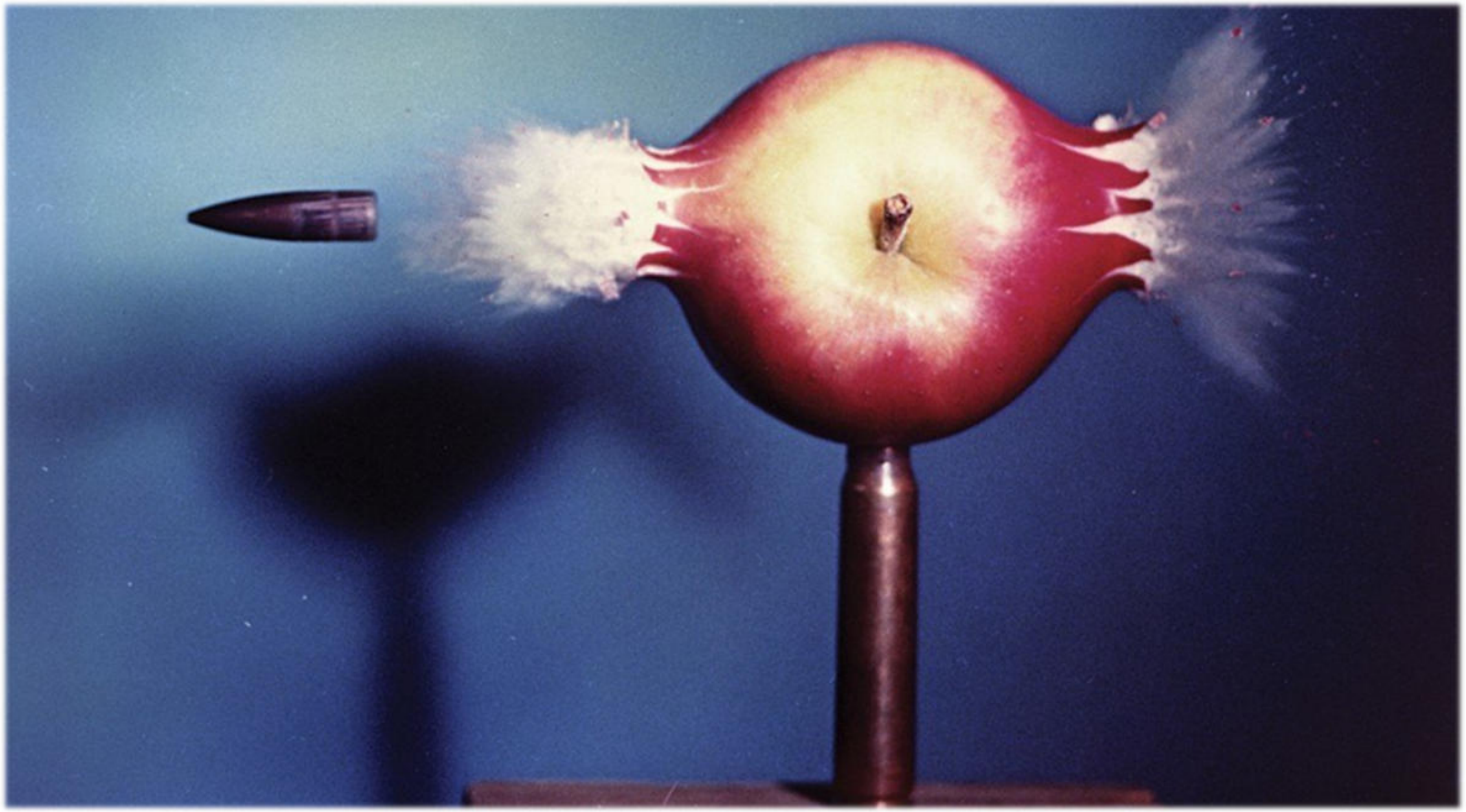
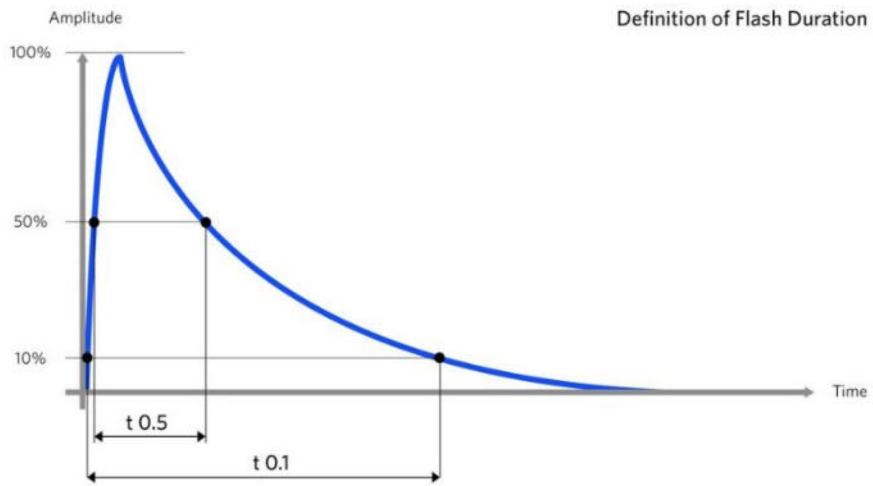
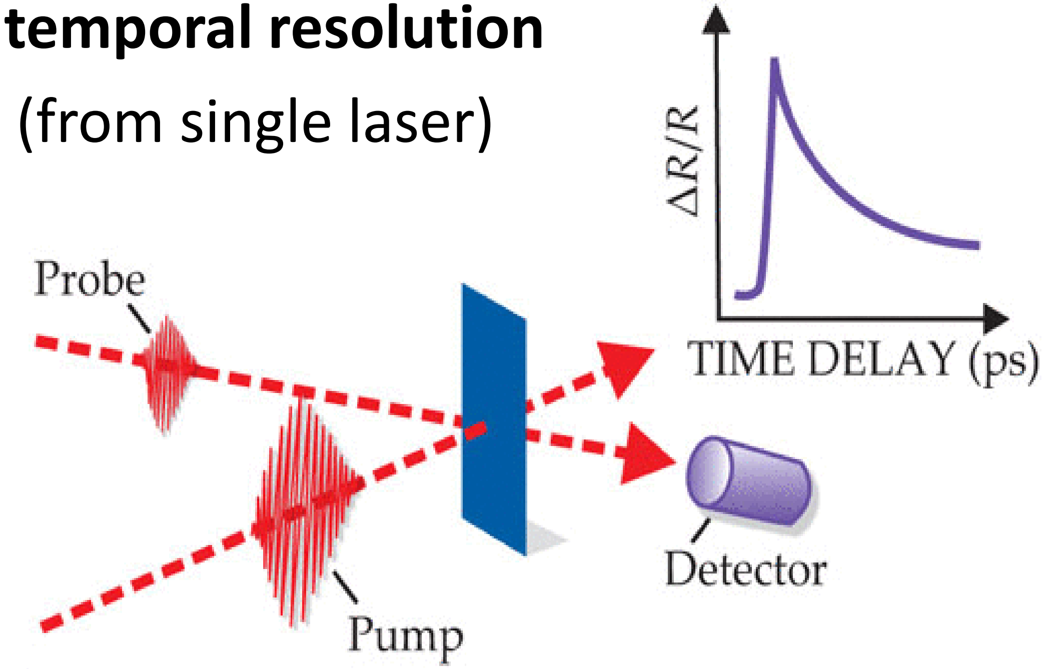


Harold Eugene Edgerton, 1964

Exploring fast processes

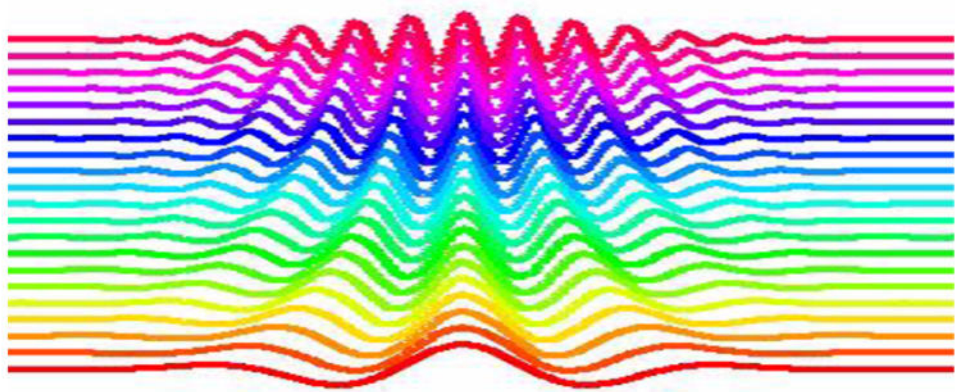
- For a good picture (not affected by motion blur)
  - Shortening the exposure times (> 1 μs)
  - Employing **flashes of light** with short duration
- Stroboscopic illumination of periodic motion (triggered by another light pulse)
- **Pulse duration** of the probe limits the **temporal resolution**
- Precise synchronization of both pulses (from single laser)



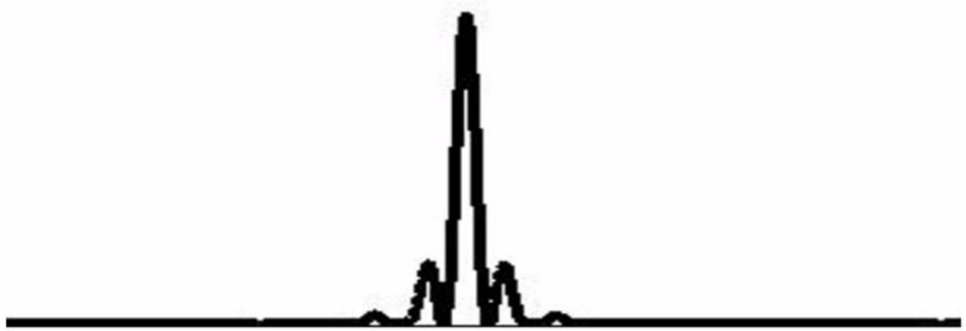


Generation of short (laser) pulses

Varying the phase of longitudinal modes (colors) of the oscillator



Resulting intensity:



$$\tau \, \Delta\nu \gtrsim \frac{1}{2}$$

$$\Delta t \, \Delta E \gtrsim \frac{h}{2}$$

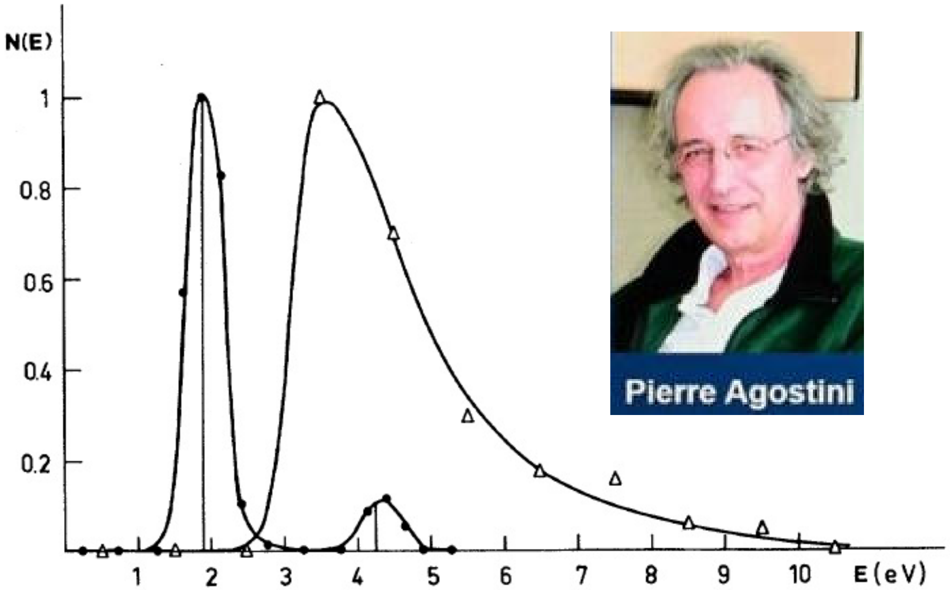
Synchronization of phases of the modes (modelocking) $\Rightarrow$ shortest possible (Fourier limited) pulse

Large bandwidth $\Rightarrow$ short central wavelength $\Rightarrow$ NIR -> VIS -> XUV

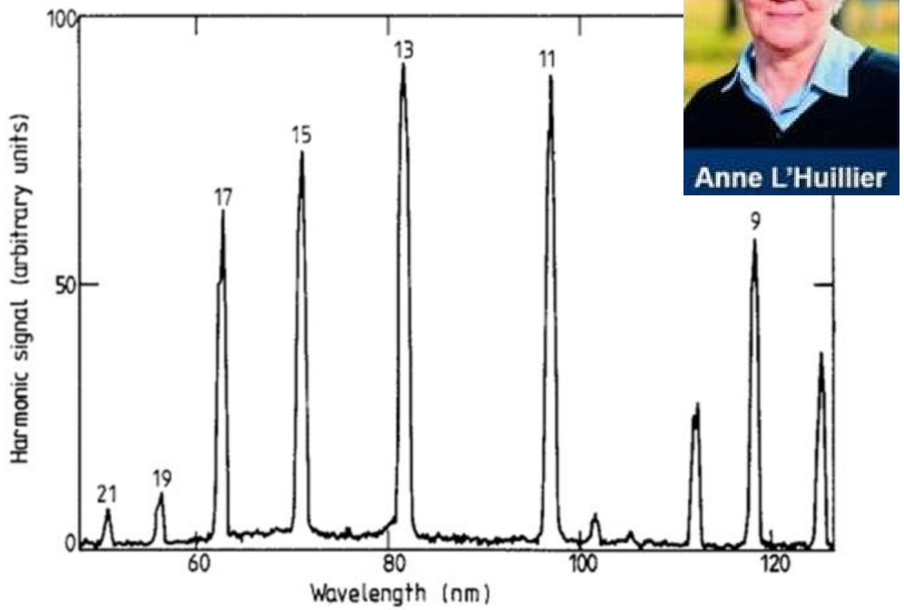
High-order harmonic generation

Contribution of the nobelists from CEA, France:

- P. Agostini – late 70’s – ionization of atoms by intense lasers (multi-photon ionization or tunel ionization)
- A. L’Huillier - late 80’s - High-order harmonic generation



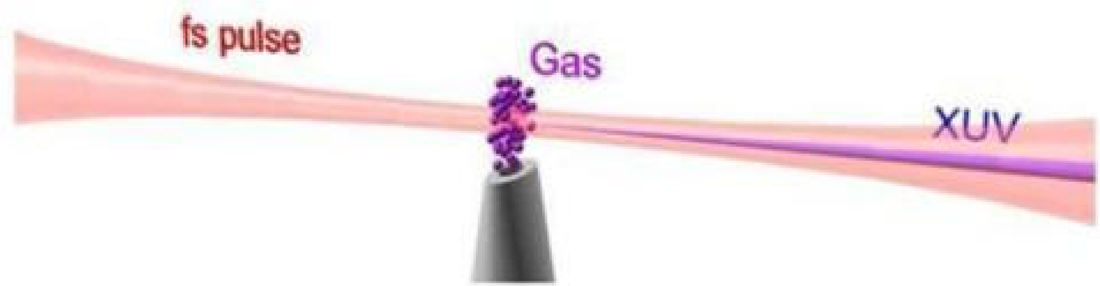
Electron spectrum at 4×10^{13} Wcm^{-2} (1064 nm) and 8×10^{12} Wcm^{-2} (532 nm) PRL **42**, 1127 (1979).

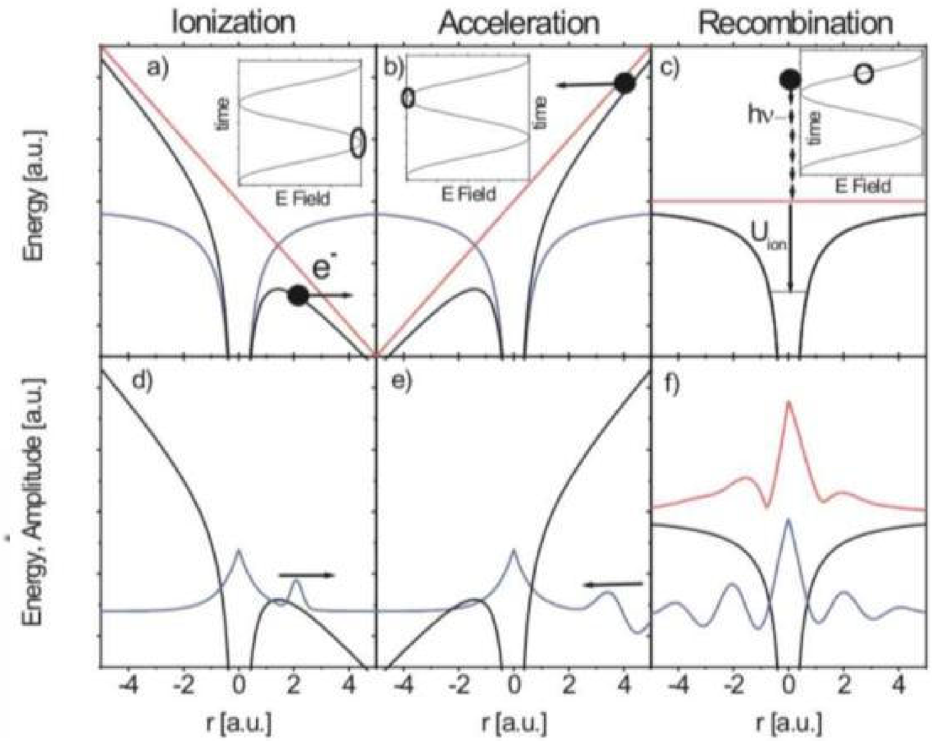


HHG in Xe generated by 1064 nm laser, J. Phys. B **21**, L31 (1988)

High-order harmonic generation

- Interaction of intense linearly polarized laser pulse with matter
  (Intensity 10^{13}-10^{15}W/cm^2)





- Three-step model:
  - Ionization of neutral atom by E-field of the pulse
  - Acceleration (propagation of the free electron in the field)
  - Recombination (only in the case of lin. poláriztation)

- Classically: P. B. Corkum, Phys. Rev. Lett., **71**, 1994 (1993)
- Quantum mechanically: M. Lewenstein et al., Phys. Rev. A **49**, 2117 (1994)

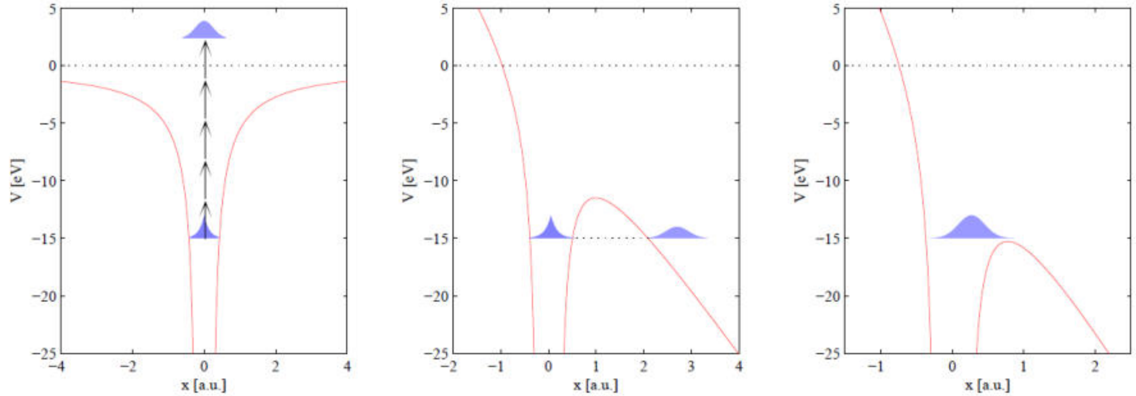
animation

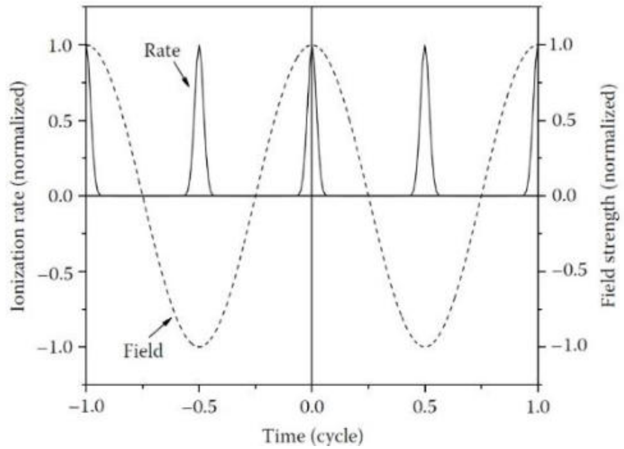
http://www.stanford.edu/~mguehr/research_HHG.html

High-order harmonic generation

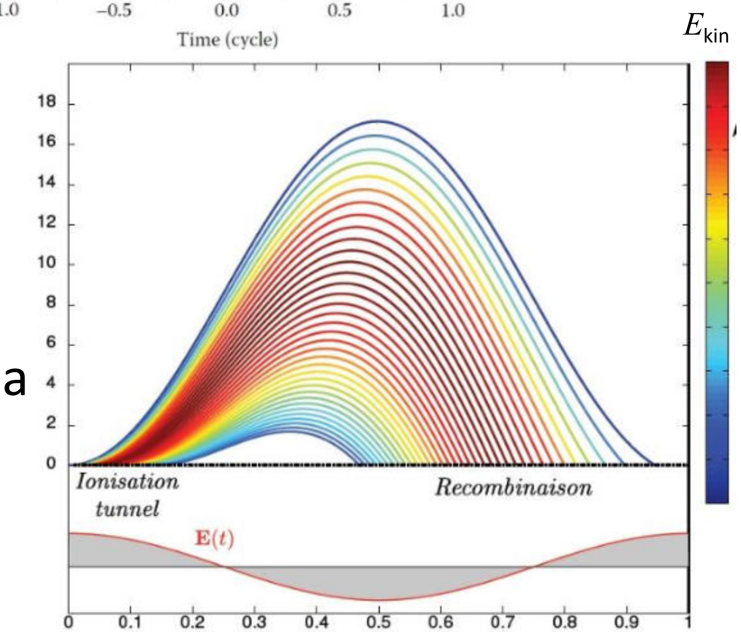
- Ionization

  Highly nonlinear function of the E-field and I_p





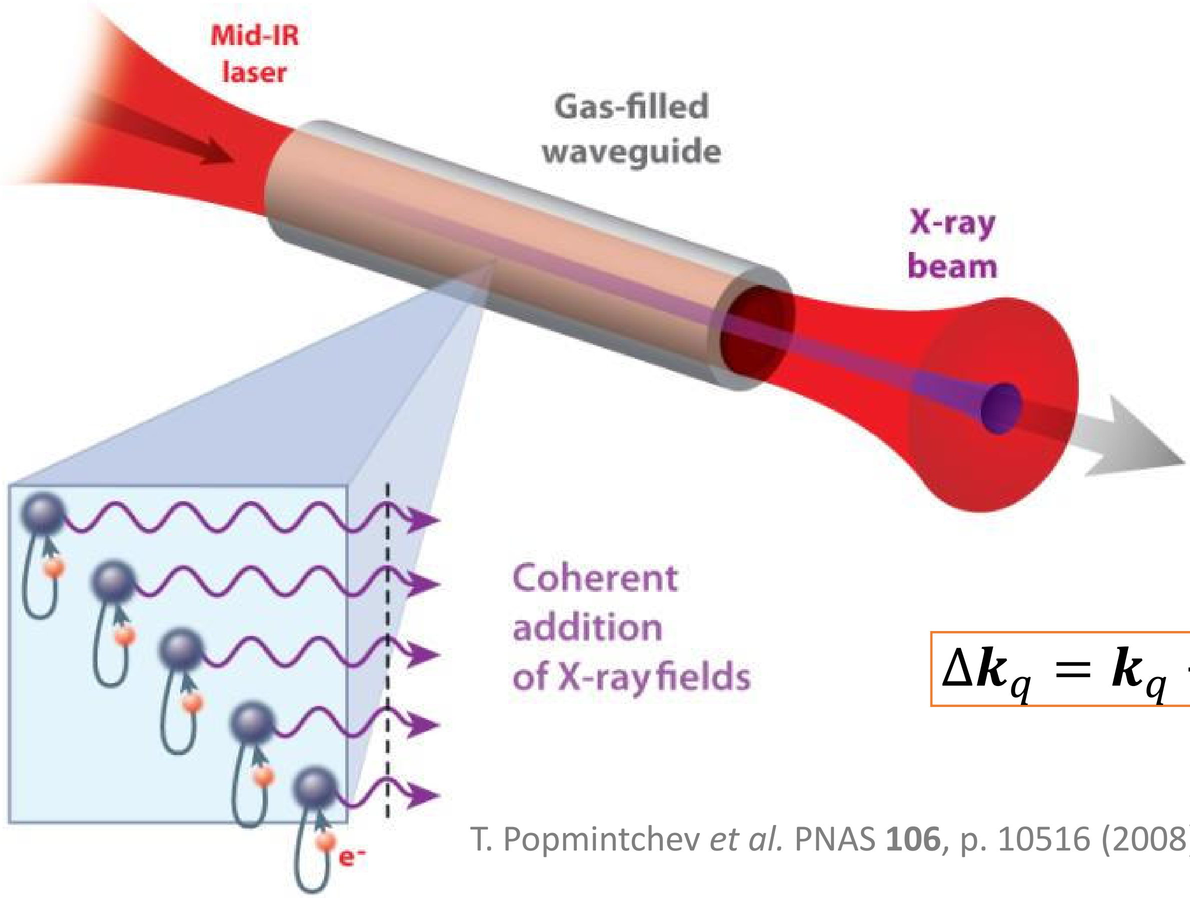
- Propagation of the free electron
  - To recombine, laser must be lin. polarized
  - el. Recombines only if it was ionized after E-field maxima
  - There are two trajectories with the same E_{kin}



$$E_{kin}^{max} = 3.17U_p \quad \Rightarrow \quad E_{cutoff} = I_p + 3.17U_p$$

High-order harmonic generation

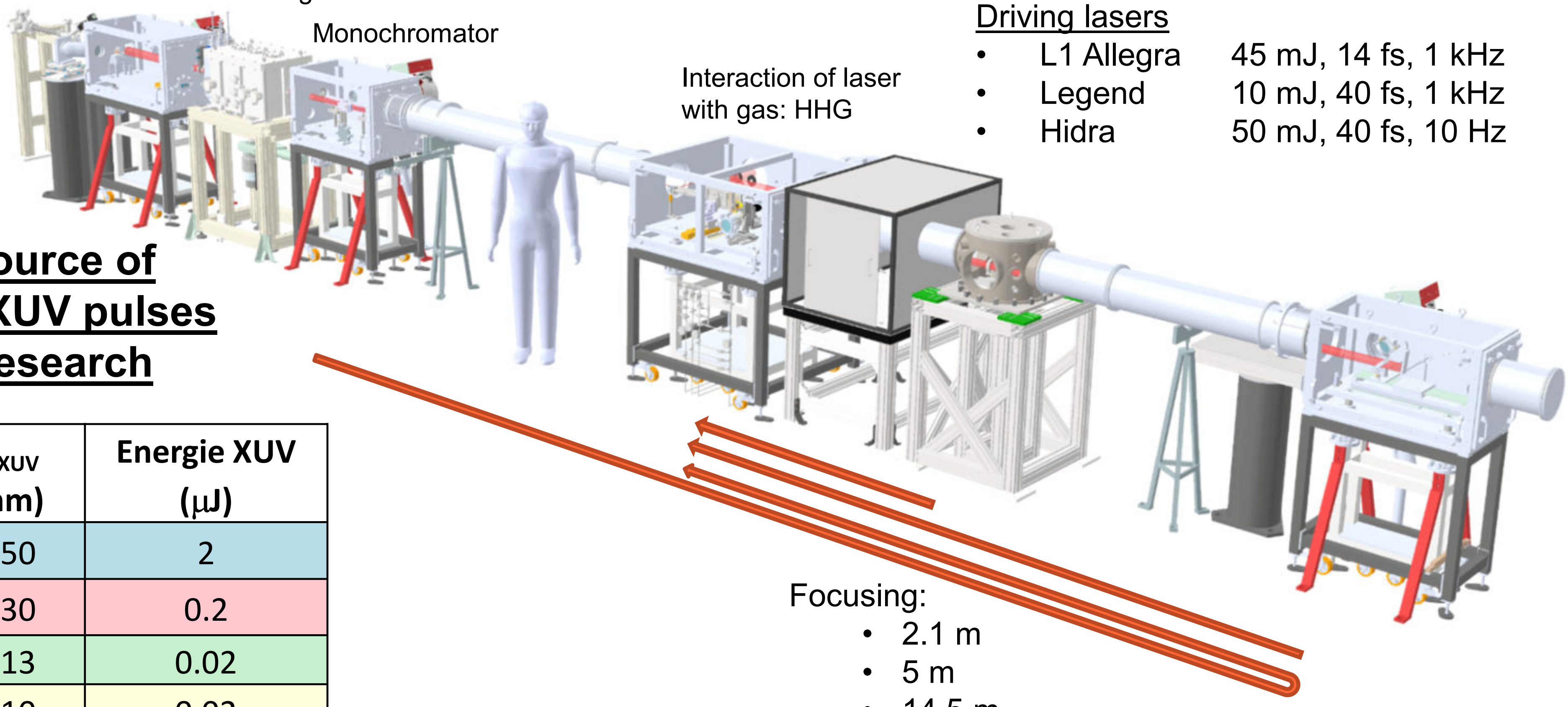
For efficient generation one needs to ensure phase-matching of all emitters



$$\Delta \boldsymbol{k}_q = \boldsymbol{k}_q - q\boldsymbol{k}_L - \boldsymbol{K}_{geo} \approx 0$$

T. Popmintchev *et al.* PNAS **106**, p. 10516 (2008)

HHG Beamline at ELI Beamlines



Driving lasers

- L1 Allegra 45 mJ, 14 fs, 1 kHz
- Legend 10 mJ, 40 fs, 1 kHz
- Hidra 50 mJ, 40 fs, 10 Hz

Intense source of coherent fs XUV pulses for user research

Plyn	λ_{XUV} (nm)	Energie XUV (μJ)
Xenon	≥50	2
Argon	≥30	0.2
Neon	≥13	0.02
Helium	≥10	0.02

Focusing:
- 2.1 m
- 5 m
- 14.5 m

High-order harmonic generation

- $\lambda = 1064$ nm $\rightarrow$ $T = 3.2$ fs

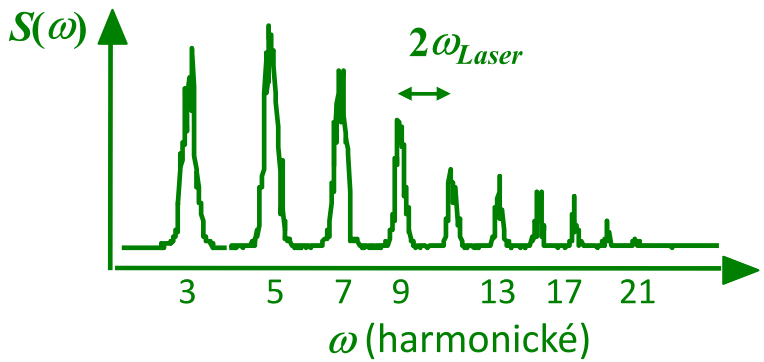
  $\rightarrow h\nu = 1.17$ eV

- $\lambda = 800$ nm $\rightarrow$ $T = 2.7$ fs

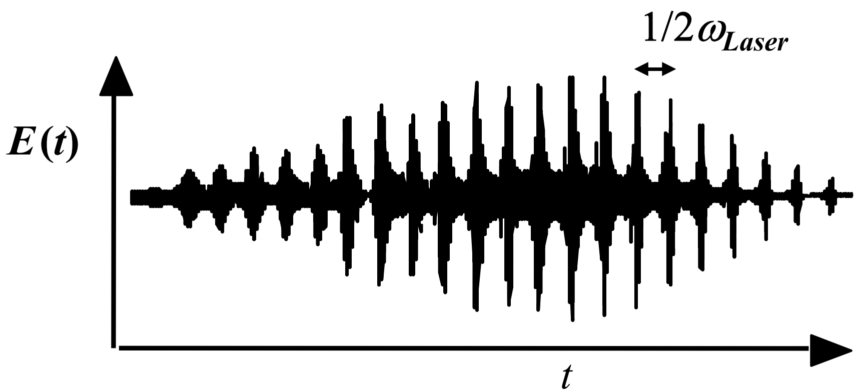
  $\rightarrow h\nu = 1.55$ eV

100 fs laser pulse: attosecond pulse train **(?)**

Measured spectrum



Electric field

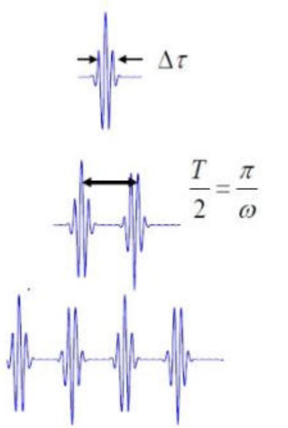


High-order harmonic generation

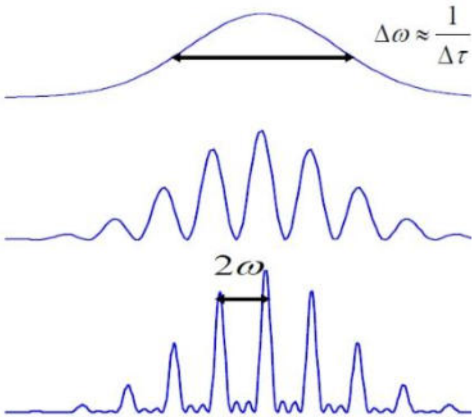
- All the atoms ionized
  - Generation stops
- Shortening the driving pulse
  - Generation of isolated sub-fs pulse

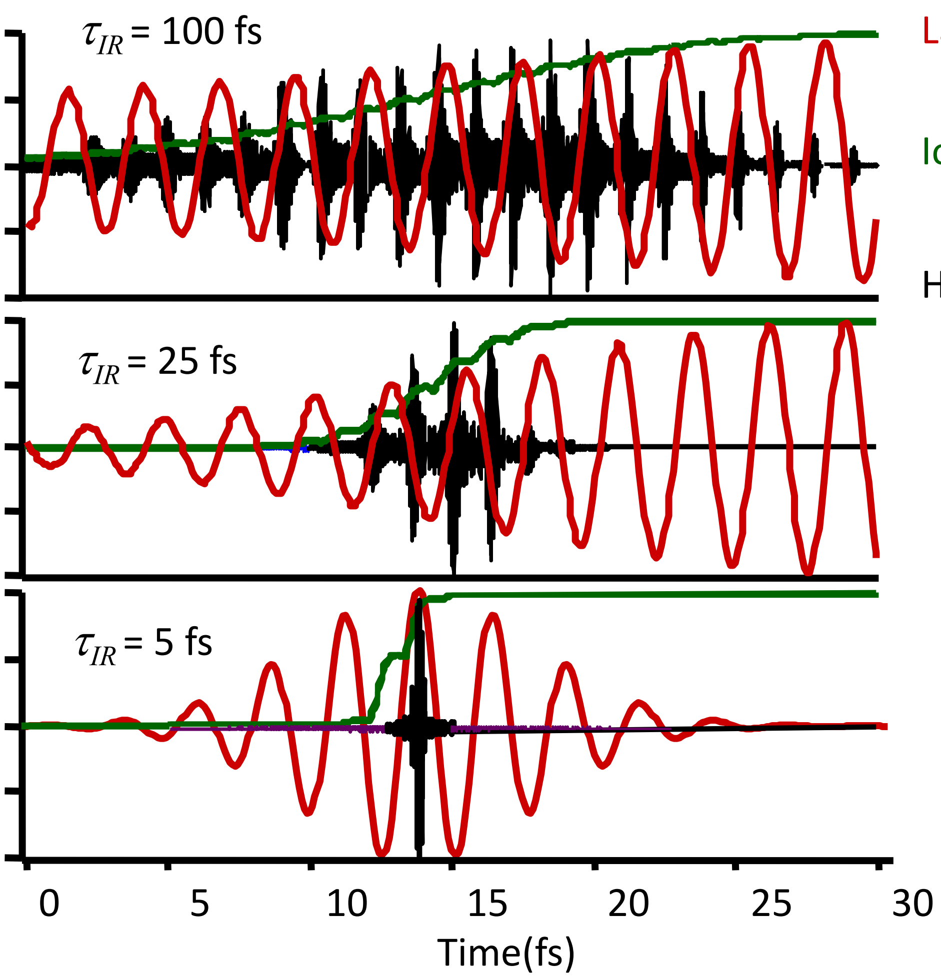
    F. Krausz 2001

Temporal profile:



Spectrum:

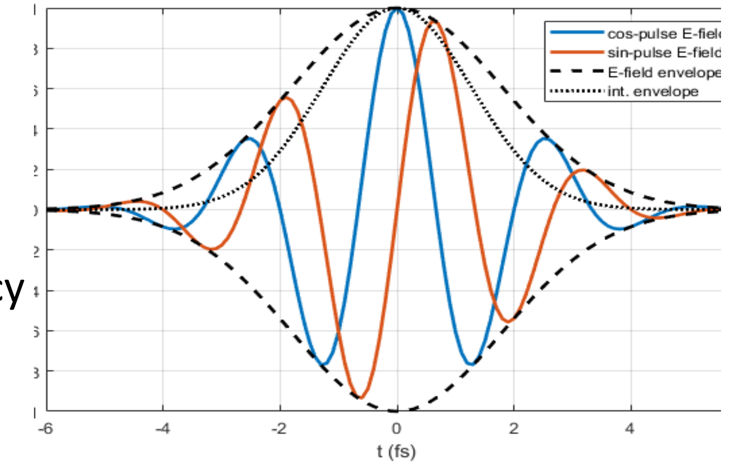




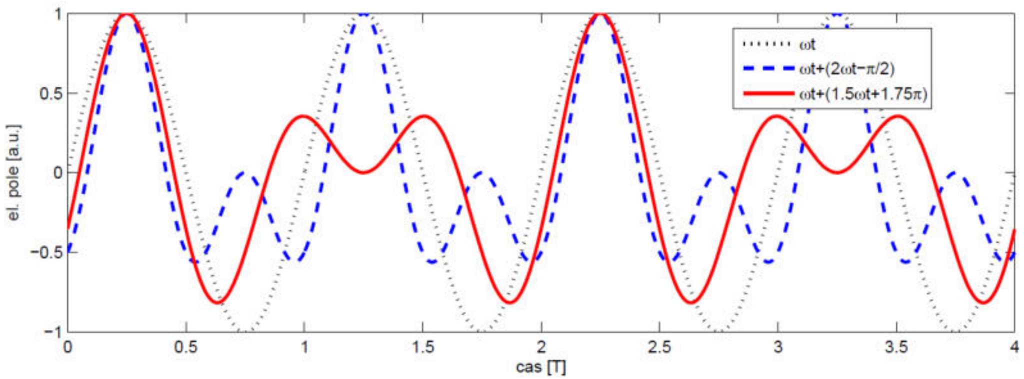
Prof. R. Trebino, Lectures on Ultrafast Optics, Georgia Institute of Technology

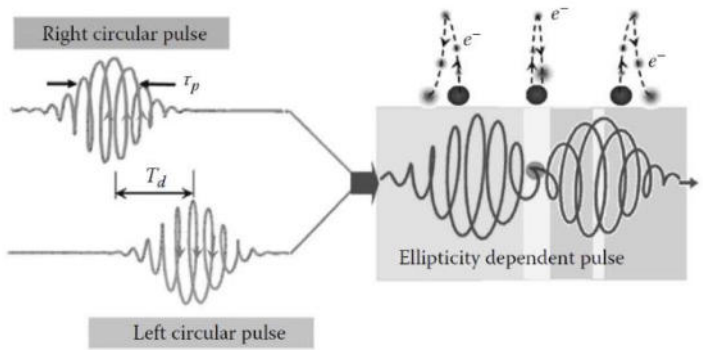
Single attosecond pulse generation

- We have to prevent repeating the microscopic three-step process
- Driver pulse with single optical cycle
  - Need for Carrier-Envelope Phase (CEP) stabilization



- Ionization gating
  - Modification of the driving field by „admixture“ of different frequency Ionization happens only at the highest peaks (not every half-cycle)
- Polarization gating
  - Superposition of two delayed circularly polarized (LH and RH) pulses to gate electron recombination





Single attosecond pulse generation

Research Article Vol. 25, No. 22 | 30 Oct 2017 | OPTICS EXPRESS 27506

Optics EXPRESS

Streaking of 43-attosecond soft-X-ray pulses generated by a passively CEP-stable mid-infrared driver

THOMAS GAUMNITZ, AROHI JAIN, YOANN PERTOT, MARTIN HUPPERT, INGA JORDAN, FERNANDO ARDANA-LAMAS, AND HANS JAKOB WÖRNER *

Laboratorium für Physikalische Chemie, ETH Zürich, Vladimir-Prelog-Weg 2, 8093 Zürich, Switzerland
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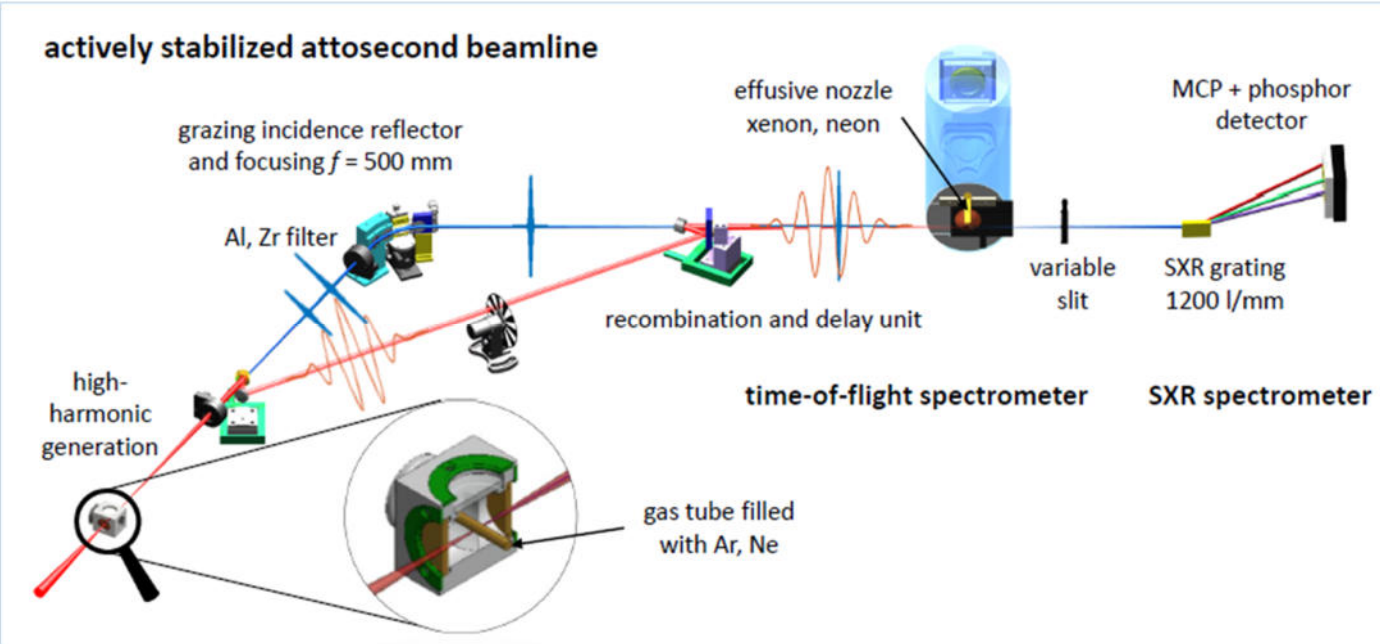
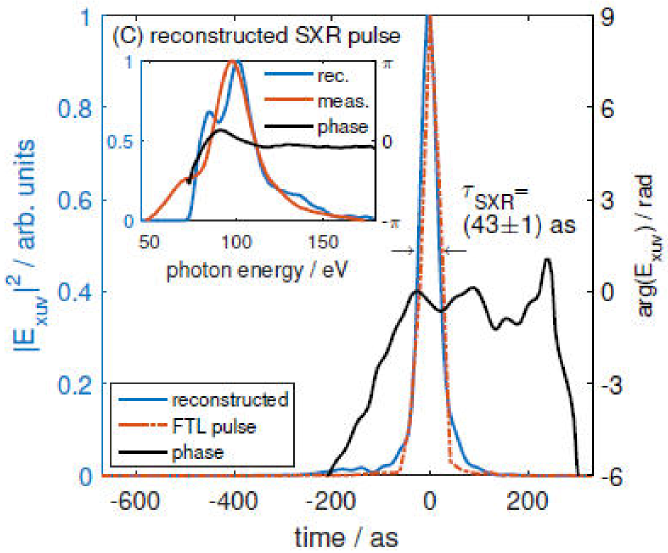
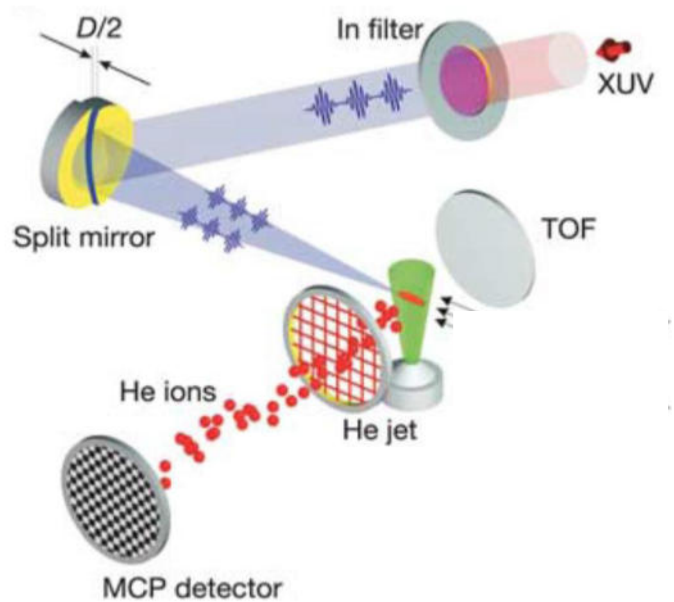


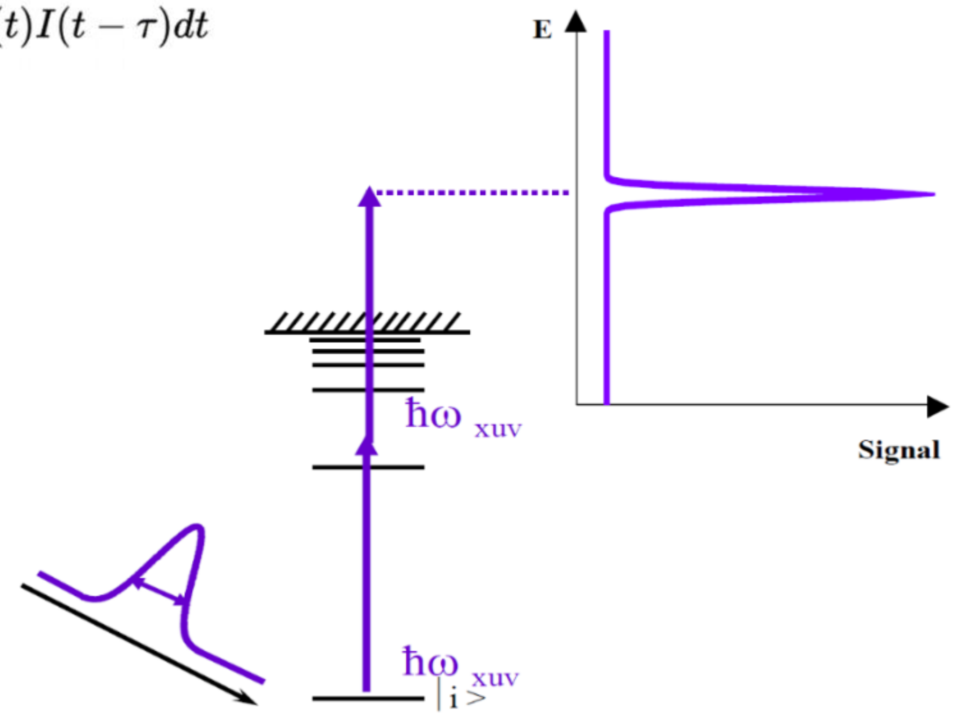


Metrology of attosecond pulses

- There is no good optics nor nonlinear crystals in the XUV range
- Two-photon ionization of noble gas – nonlinear process of the lowest order
  - Intensity autocorrelation $A(\tau) = \int_{-\infty}^{+\infty} I(t)I(t-\tau)dt$

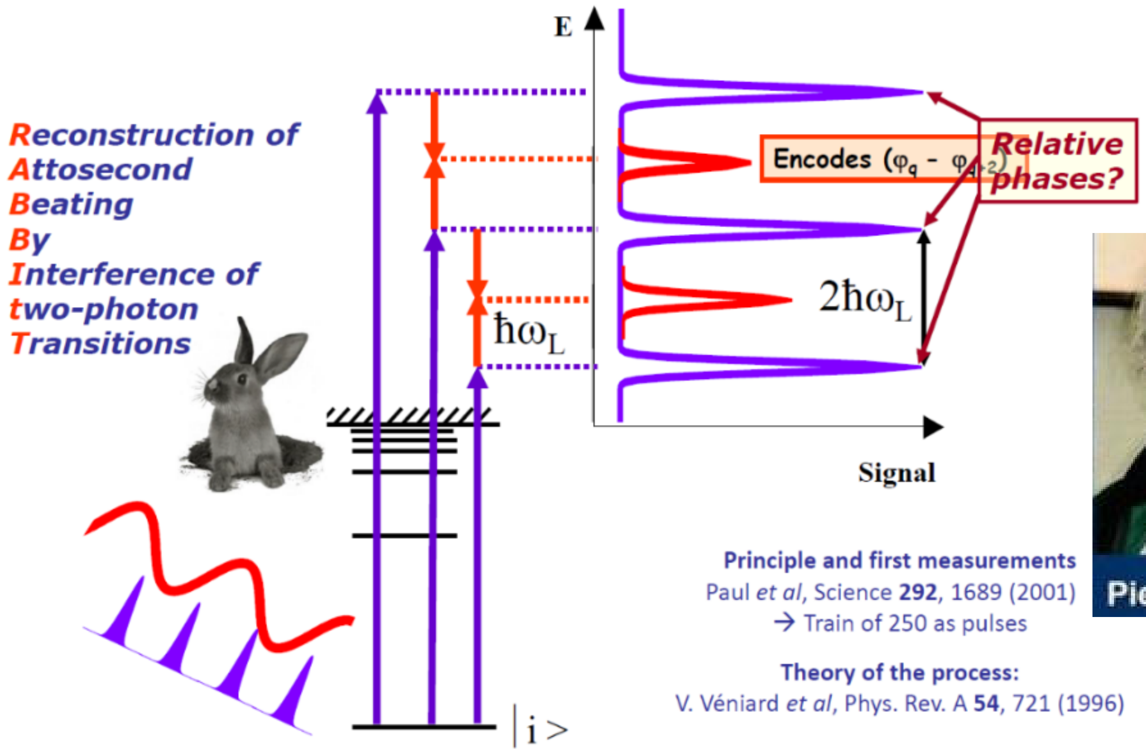


P. Tzallas et al. Nature **426**, 267 (2003)



Metrology of attosecond pulses

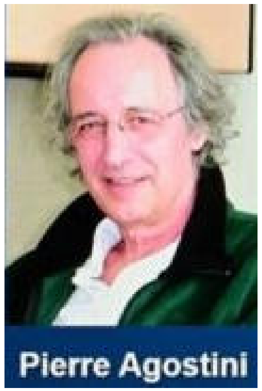
- Most often used methods employ cross-correlation (XUV-NIR):
  - RABBIT – for **attosecond pulse train** (P. Agostini 2001)

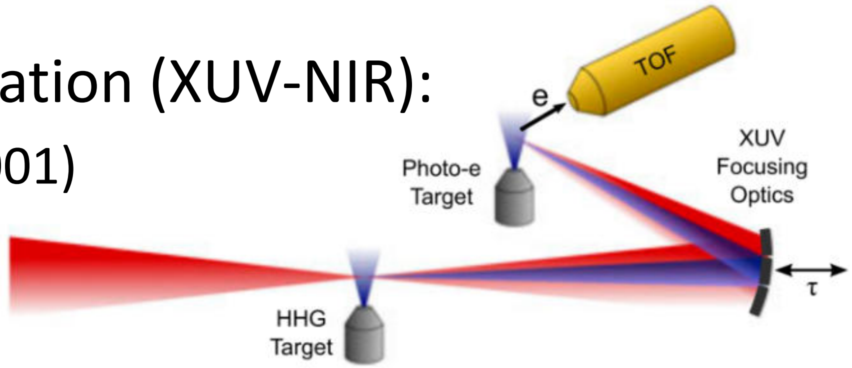


P. Salières, Erice Summer school 2023

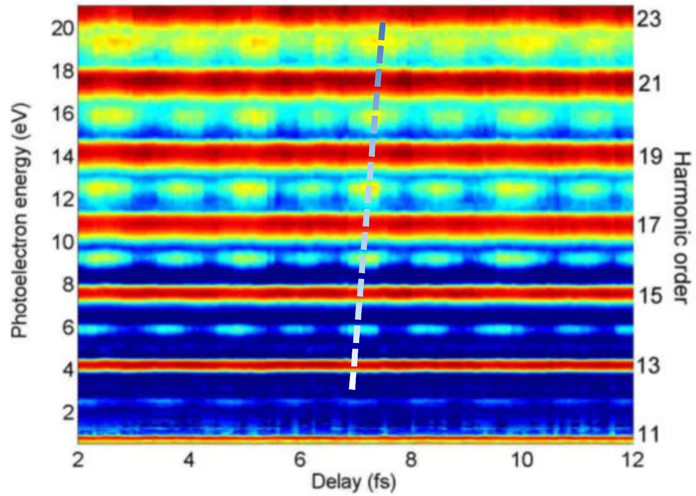
Principle and first measurements
Paul *et al*, Science **292**, 1689 (2001)
→ Train of 250 as pulses

Theory of the process:
V. Véniard *et al*, Phys. Rev. A **54**, 721 (1996)





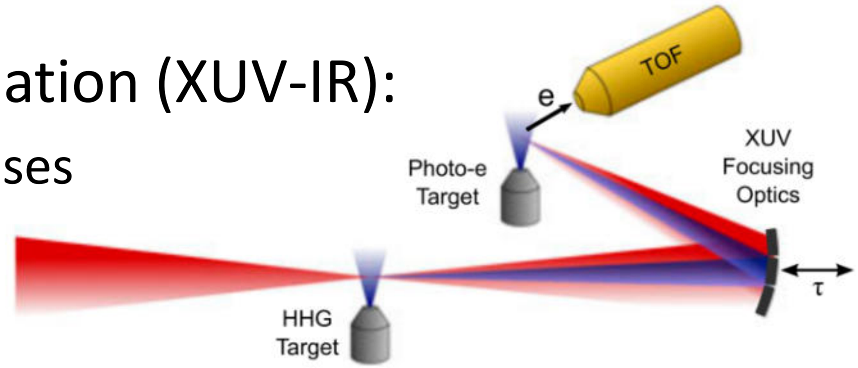
K.T. Kim et al., Nature Photonics **8**, 187 (2014)



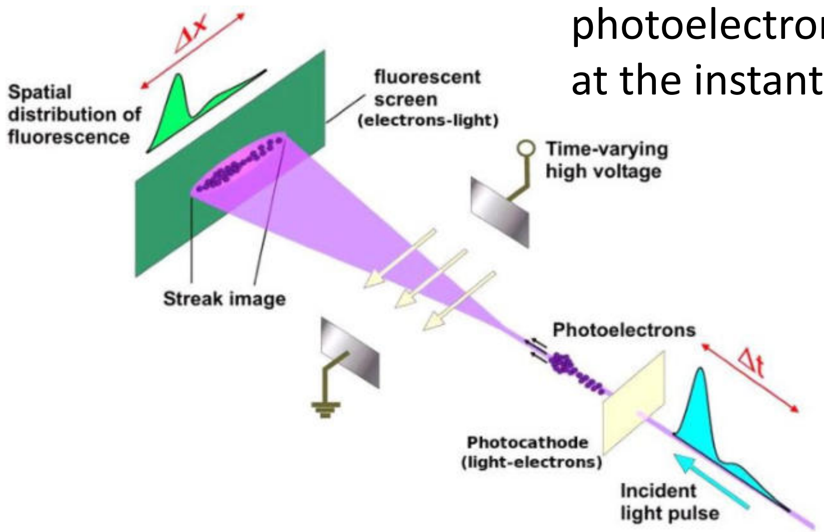
Typical RABBIT result

Metrology of attosecond pulses

- Most often used methods employ cross-correlation (XUV-IR):
  - Attosecond streaking – for isolated attosecond pulses
    - Atoms of the gas are the photocathode
    - Fast changing electric field = field of the laser pulse
    - Fluorescent screen = electron spectrometer

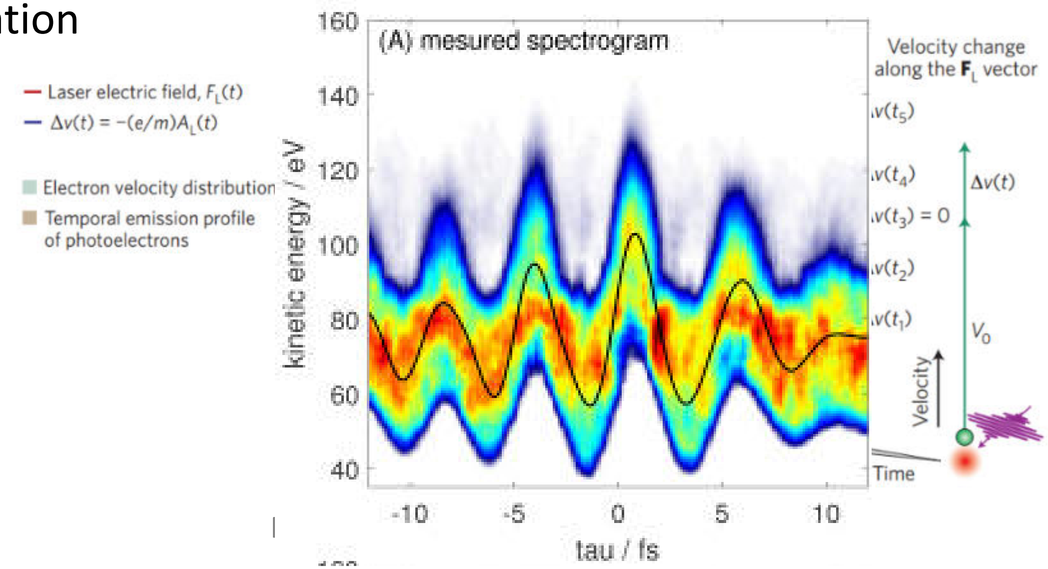


K.T. Kim et al., Nature Photonics **8**, 187 (2014)



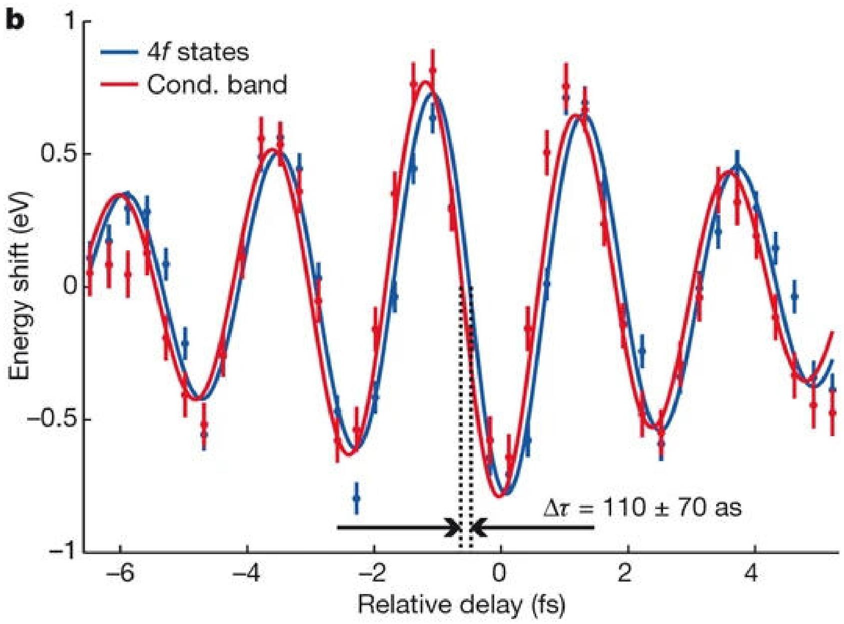
Conventional streak camera (Wikipedia)

photoelectrons (hν > I_p) gain momentum (energy) depending on phase of the field at the instant of ionization

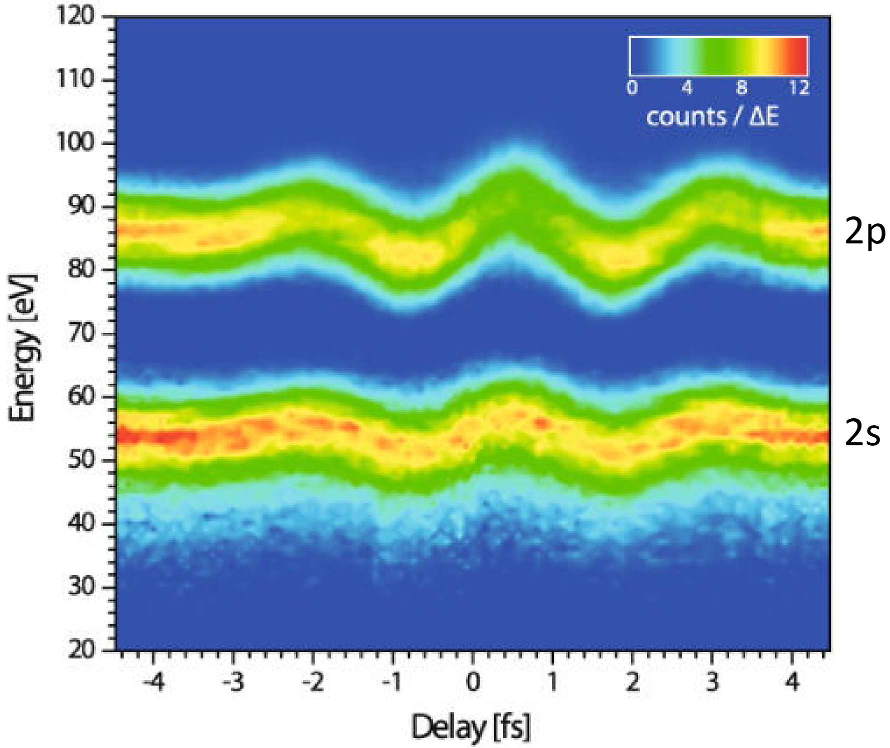


Applications of attosecond pulses

- Measuring the delay of photoionization (from different atomic shells)
  - Using attosecond streaking (with 200 as pulses)
  - Photoionization of Ne (2s and 2p) – delay 20 as
  - Other atoms, molecules and solids



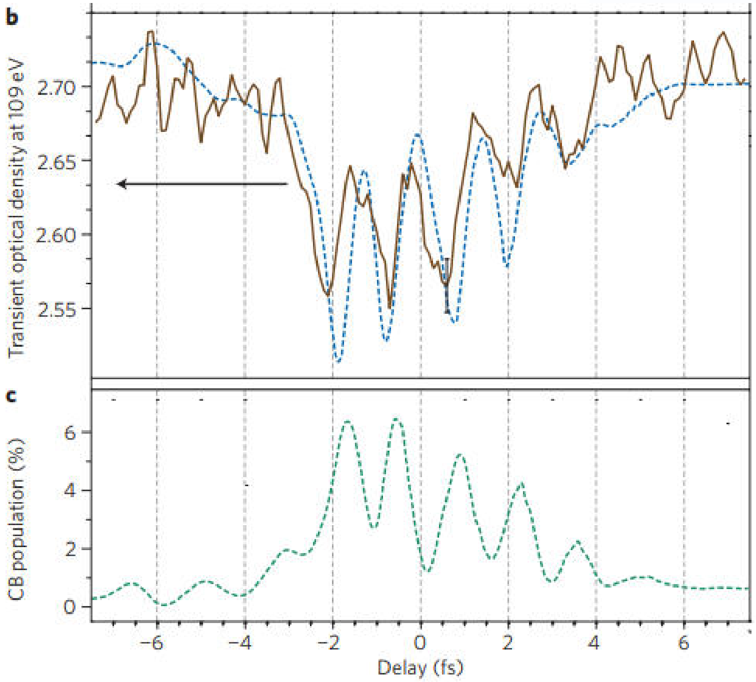
A. L. Cavalieri *Nature* **449**, 1029 (2007)

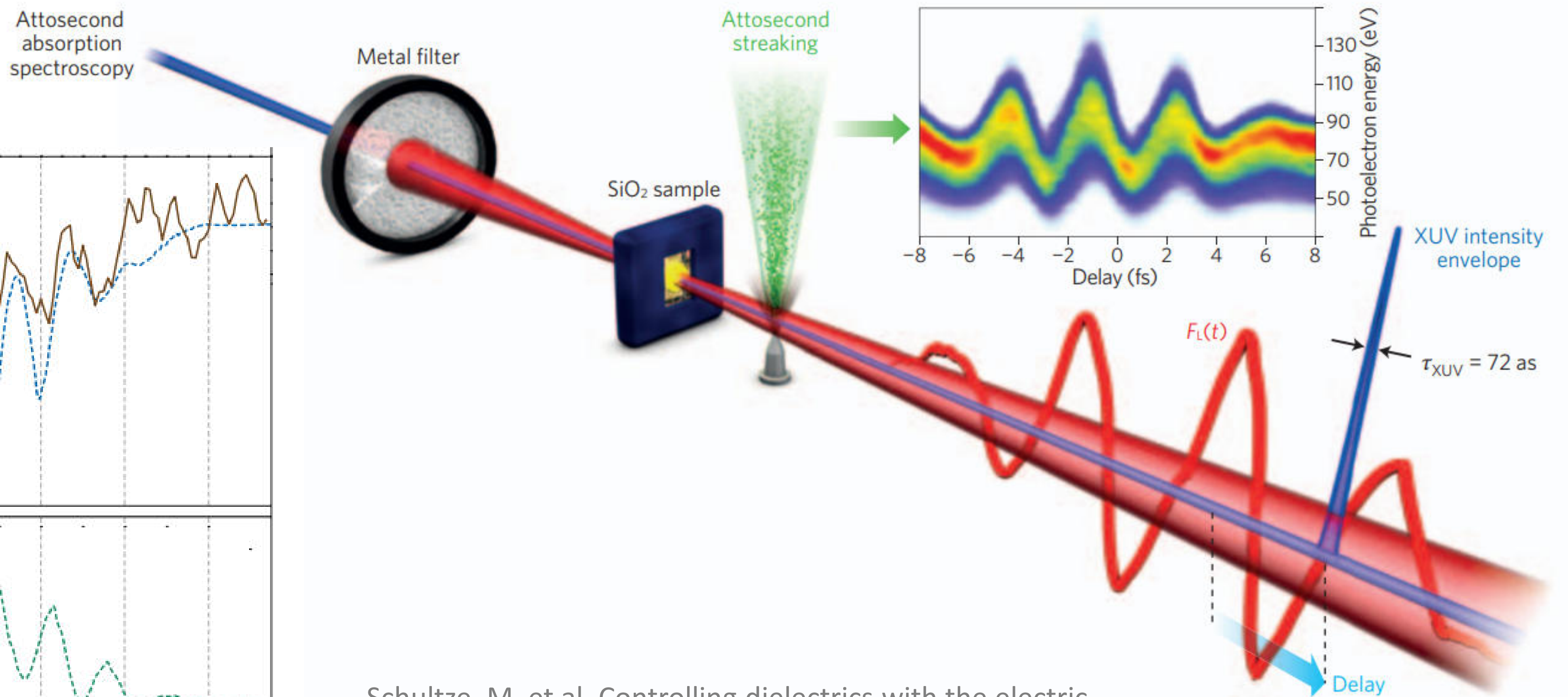


M. Schultze *et al.* Delay in Photoemission *Science* **328**,1658 (2010).

Applications of attosecond pulses

- „direct“ measurement of electric field (vect. potential) of a laser pulse
  - Modification of optical properties of SiO_2 by instant field of the laser

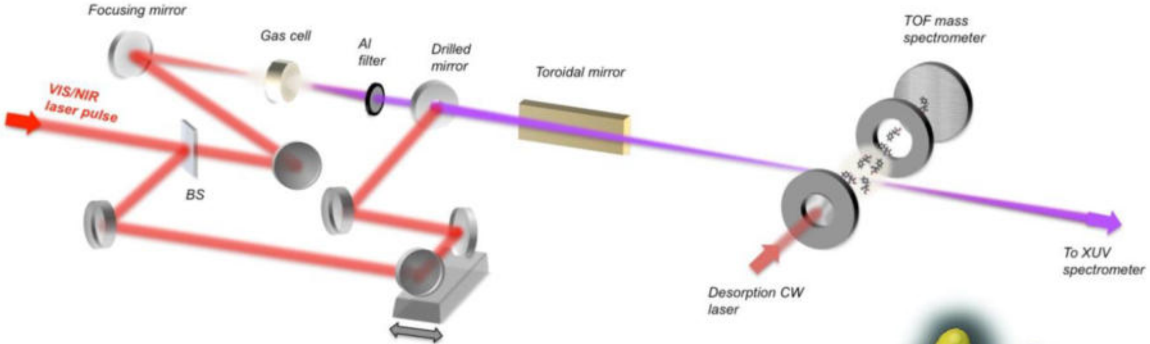


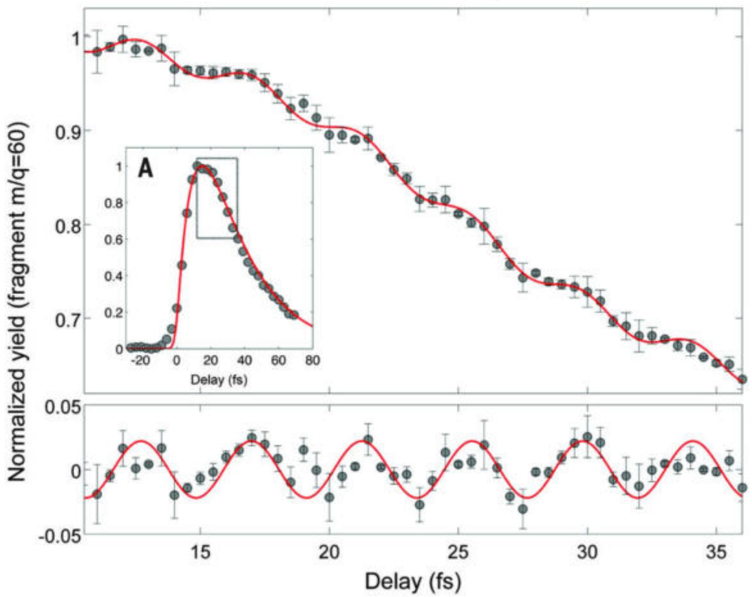


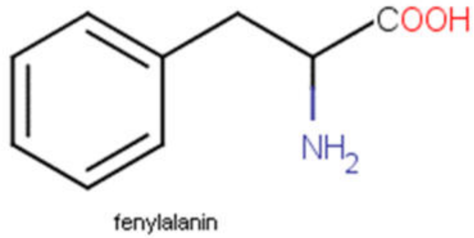
Schultze, M. et al. Controlling dielectrics with the electric field of light. Nature **493**, 75 (2013)

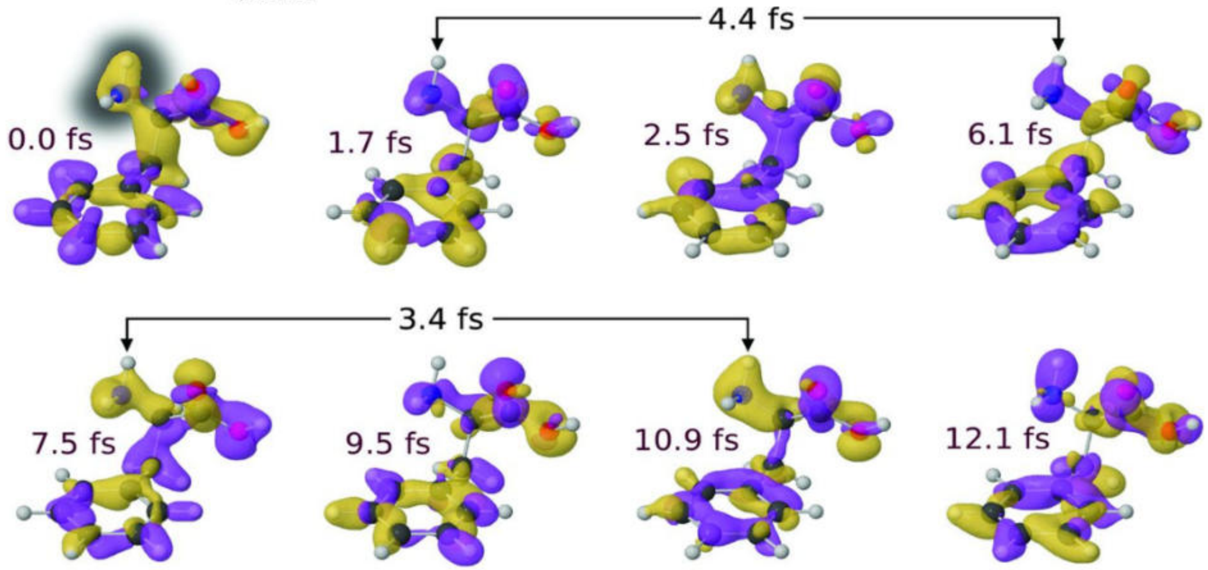
Applications of attosecond pulses

- Resolving the ultrafast dynamics of electrons in molecules





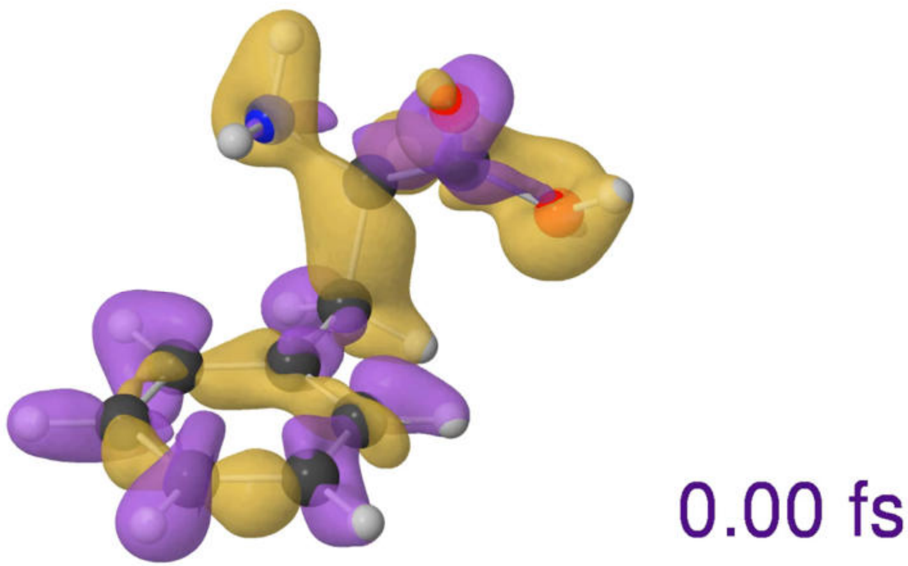




F. Calegari *et al.* Ultrafast electron dynamics in phenylalanine initiated by attosecond pulses *Science* **346**, 336 (2014)

Applications of attosecond pulses

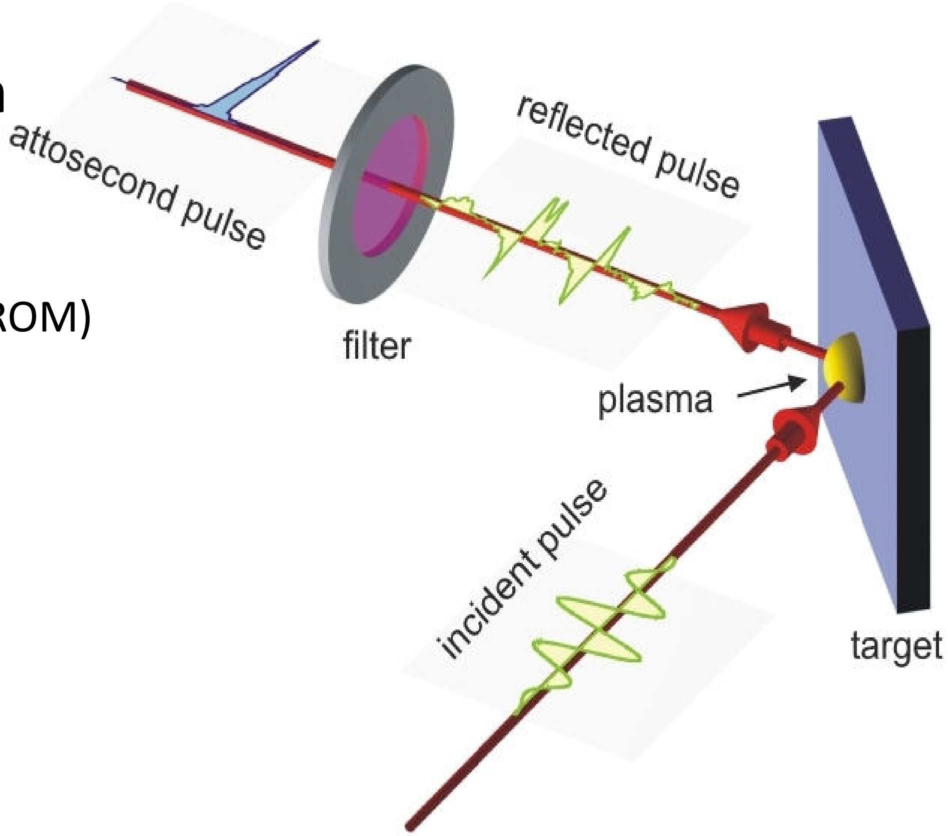
- Resolving the ultrafast dynamics of electrons in molecules



F. Calegari *et al.* Ultrafast electron dynamics in phenylalanine initiated by attosecond pulses *Science* **346**, 336(2014)

to conclude…

- HHG and SAP generation from gas has proven to be an excellent tool for ultrafast science
- There are other mechanisms of generation
  - E.g. During interaction of intense laser ($I > 10^{18}$ W/cm^2) with solid surface
    - Reflection from Relativistically Oscillating Mirror (ROM)

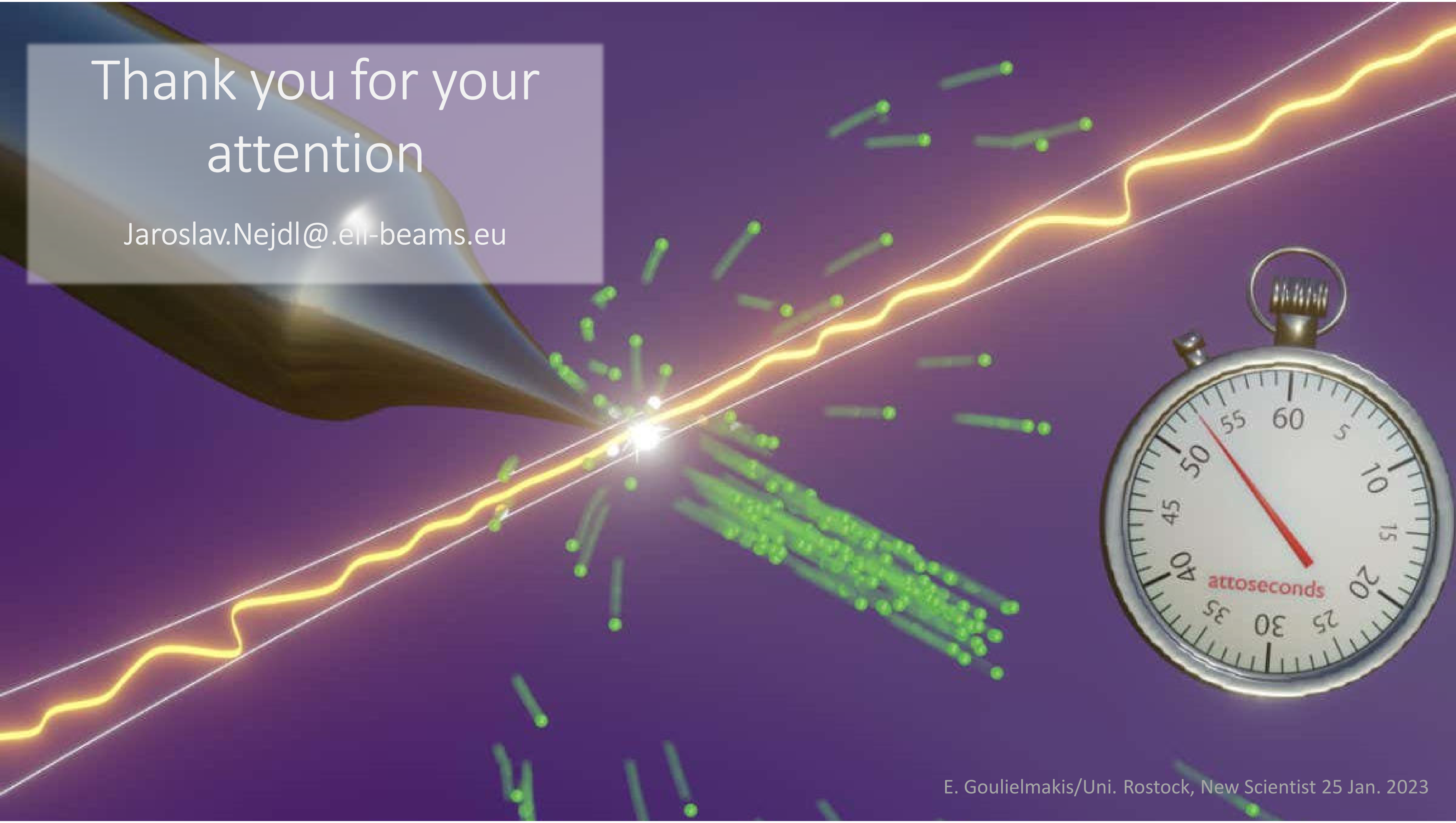


U. Teubner and P. Gibbon, Rev. Mod. Phys. **81**, p. 445 (2009).

but that is another story…

Thank you for your attention

Jaroslav.Nejdl@.eli-beams.eu

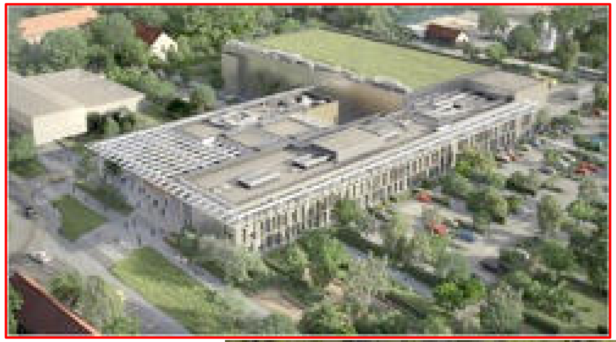


E. Goulielmakis/Uni. Rostock, New Scientist 25 Jan. 2023

Extreme Light Infrastructure (ELI)

ELI comprises three branches:

- **Attosecond Laser Science**
  new regimes of time resolution
  (ELI ALPS, Szeged, HU)



- **High-Energy Beam Facility**
  ultra-short pulses of high-energy particles and radiation
  (ELI Beamlines, Dolní Břežany, CZ)

- **Nuclear Physics Facility**
  brilliant gamma beams (up to 19 MeV) and brilliant neutron beam
  (ELI NP, Magurele, RO)