The route to attosecond pulses

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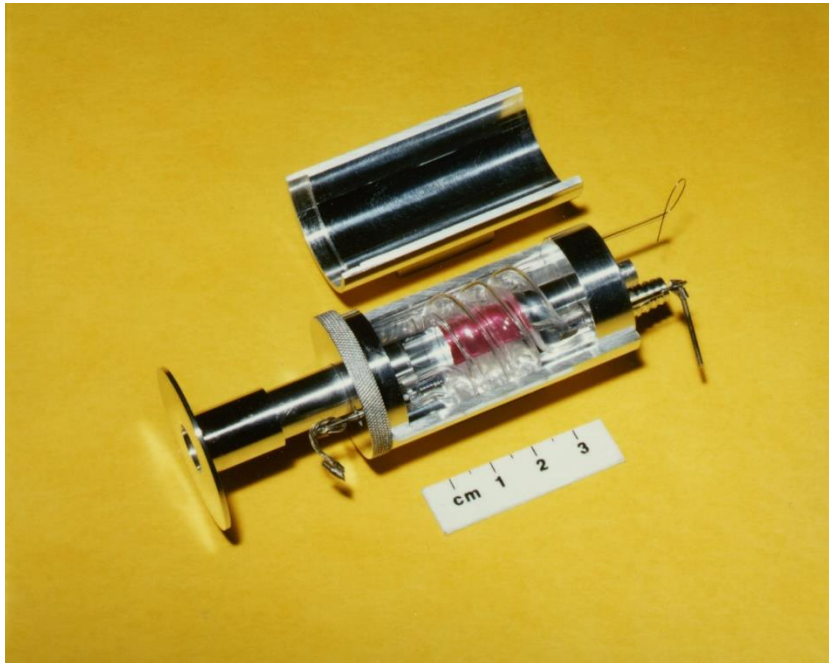
Outline

- **High-order harmonic generation**
- **Attosecond light pulses**
- **Attosecond physics**

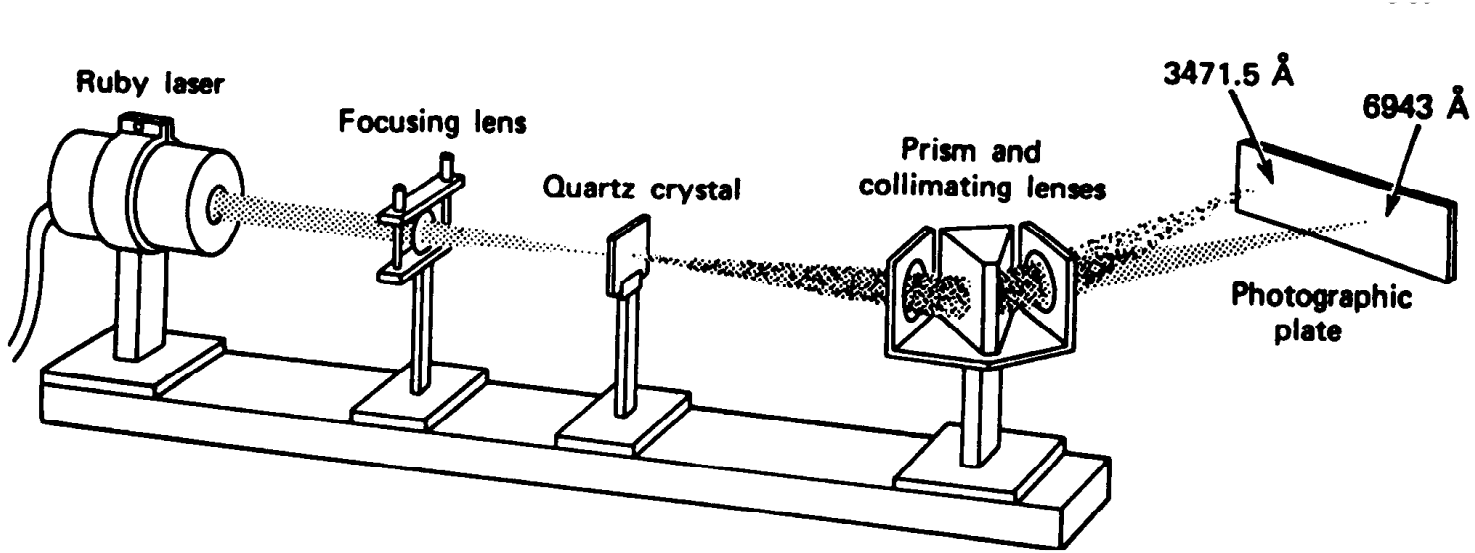
The invention of the laser



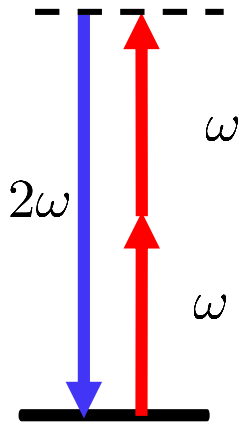
Lejournal.cnrs.com

Maiman, Nature, **187**, 493 (1960)

Nonlinear optics



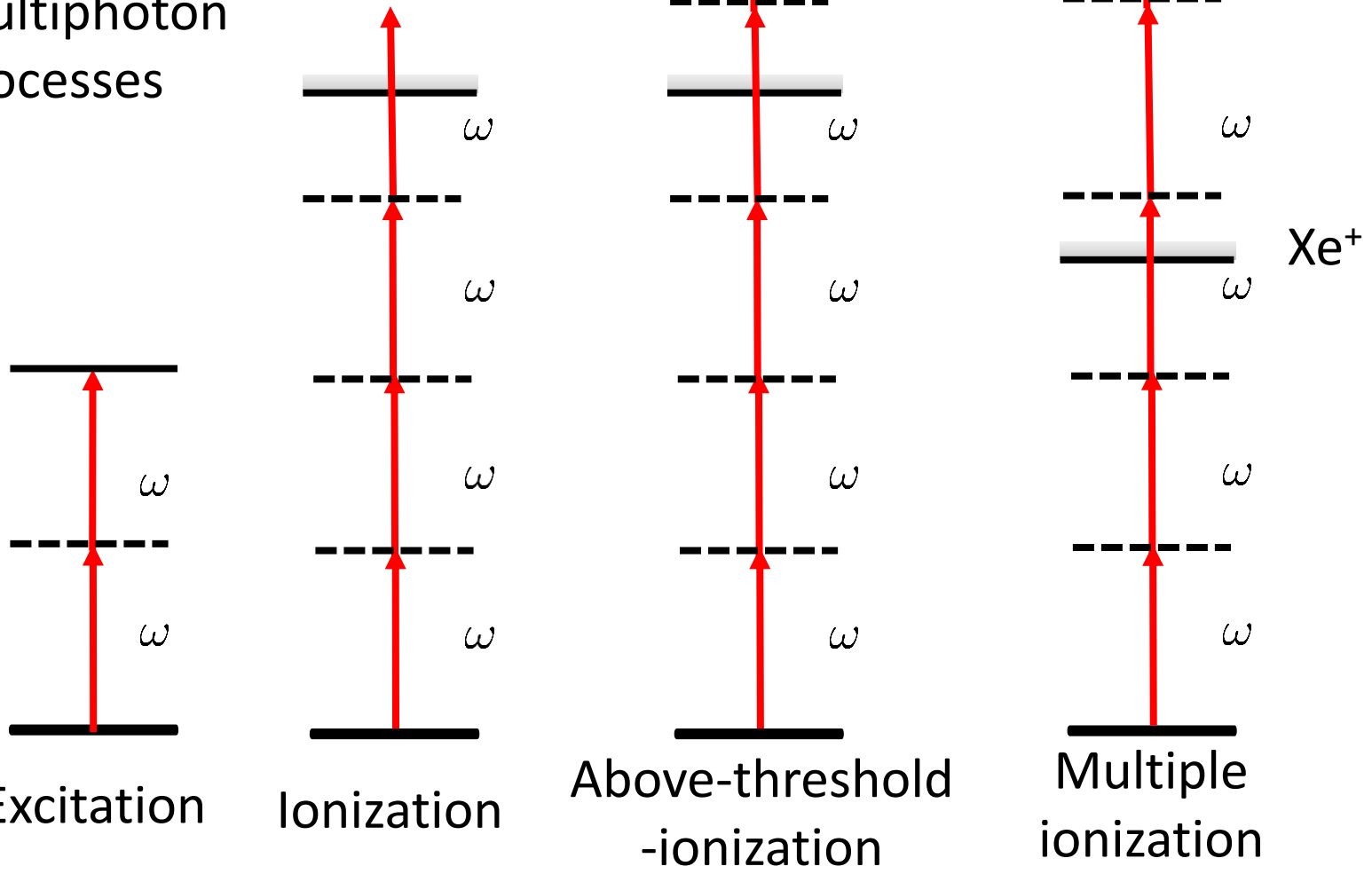
New frequencies



Franken et al, Phys. Rev. Lett. **7**, 118 (1961)

Atoms in strong laser fields

Multiphoton processes



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Predicted in 1931 by

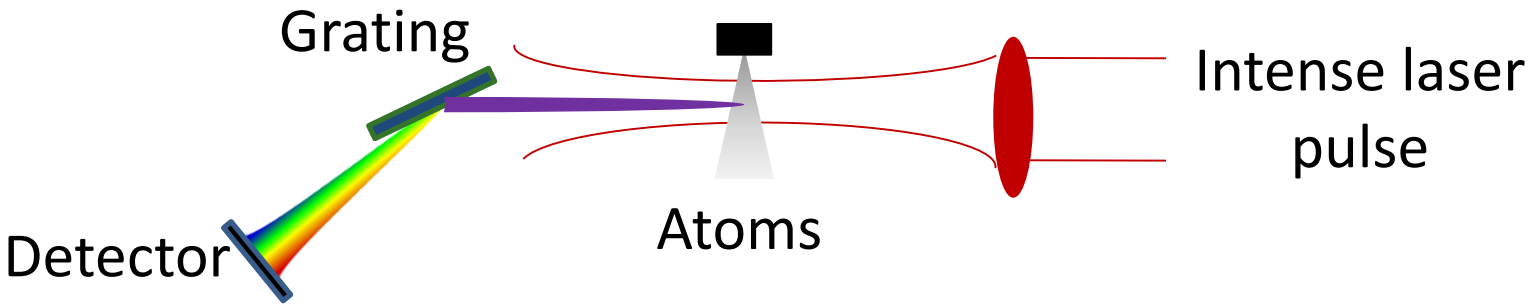
Maria Goeppert-Mayer

Voronov et al. Sov. Phys. JETP (1966)

Agostini et al. Phys. Rev. Lett. **42**, 1127 (1979)

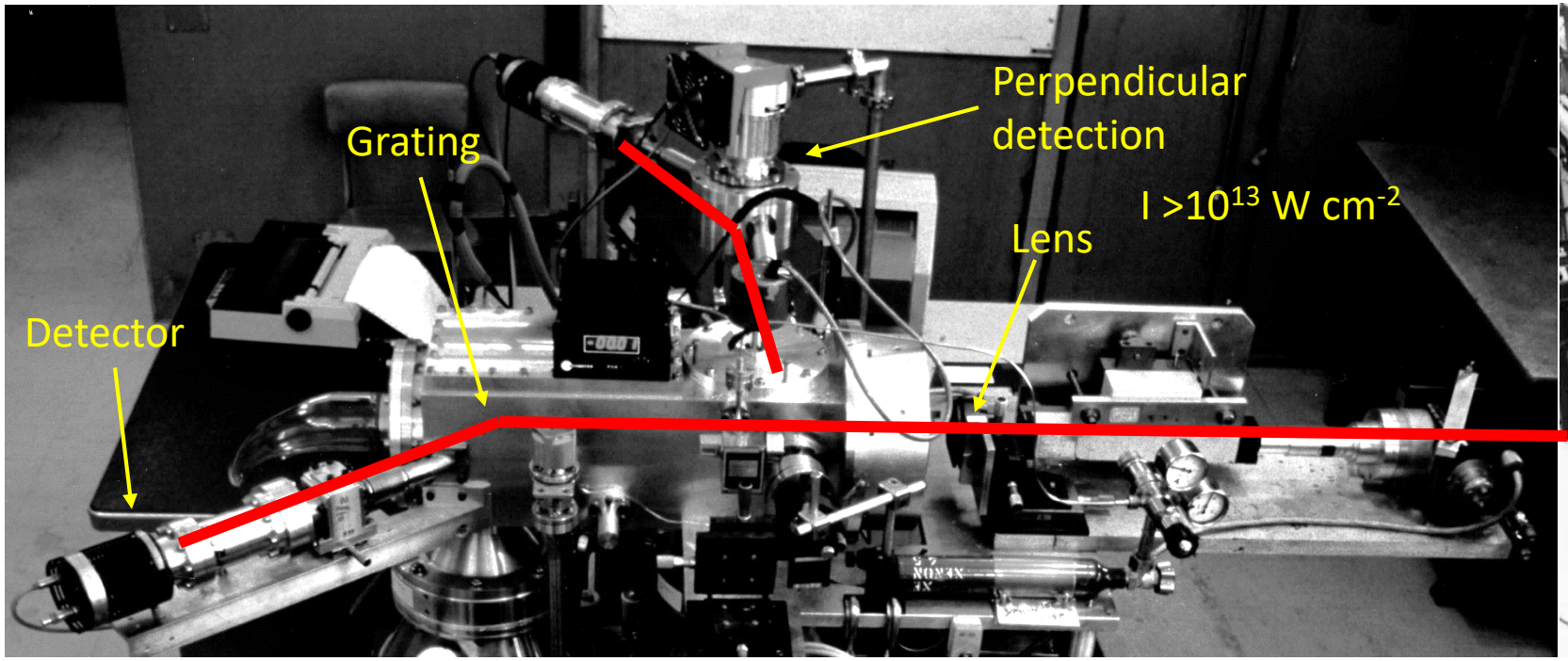
L'Huillier et al. Phys. Rev. A **27**, 2503 (1983)

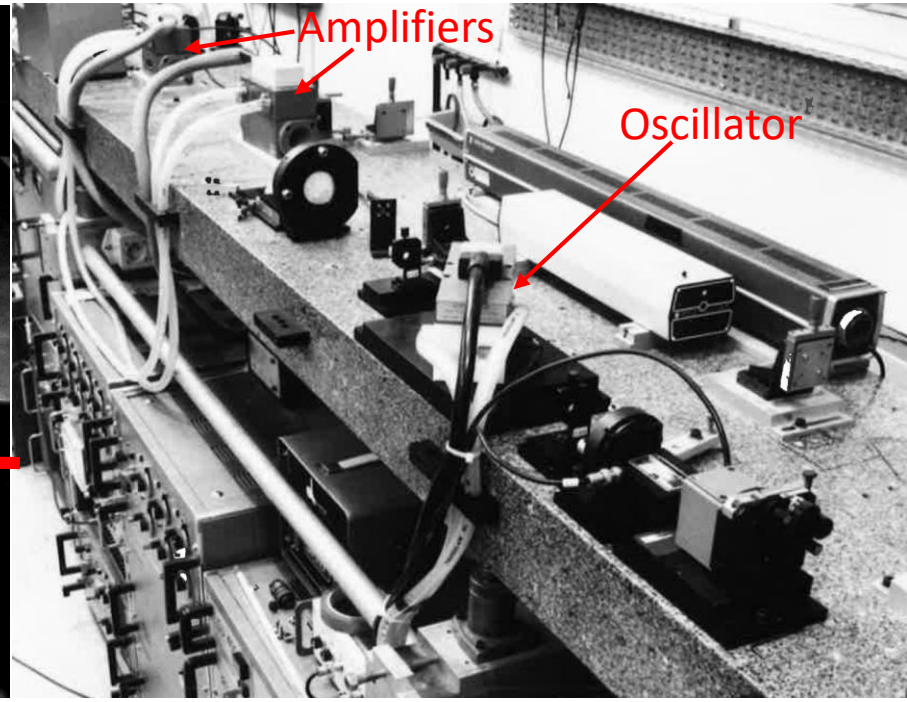
Fluorescence?



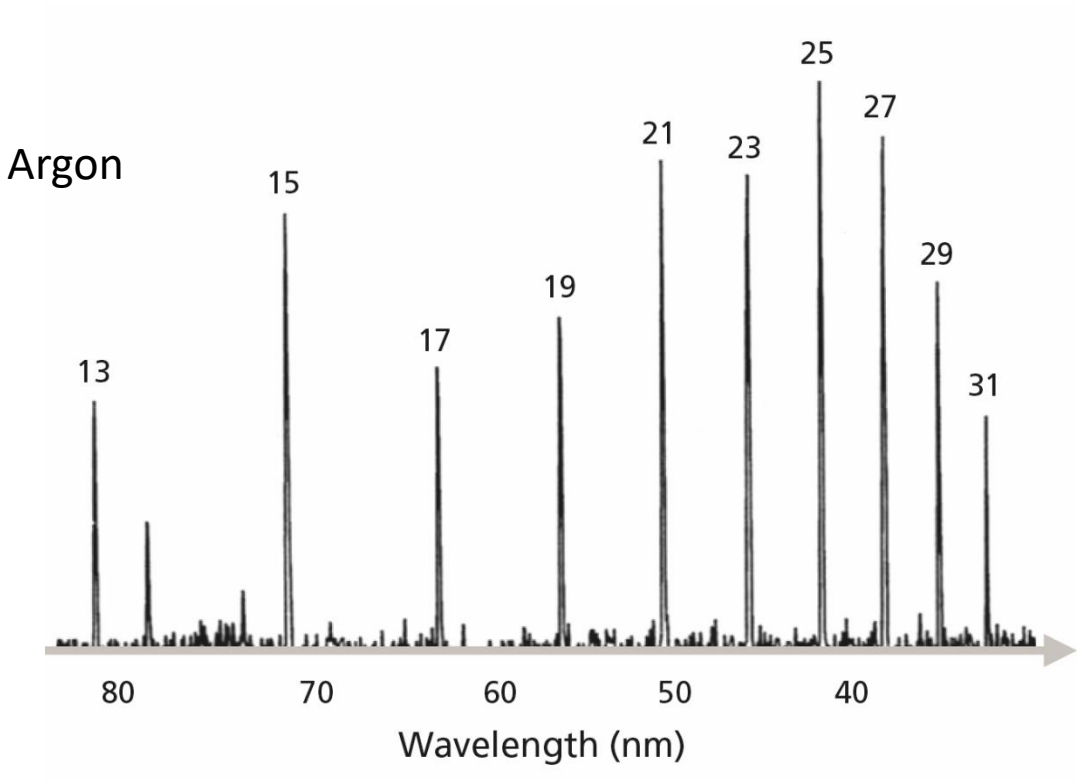
G. Mainfray
C. Manus

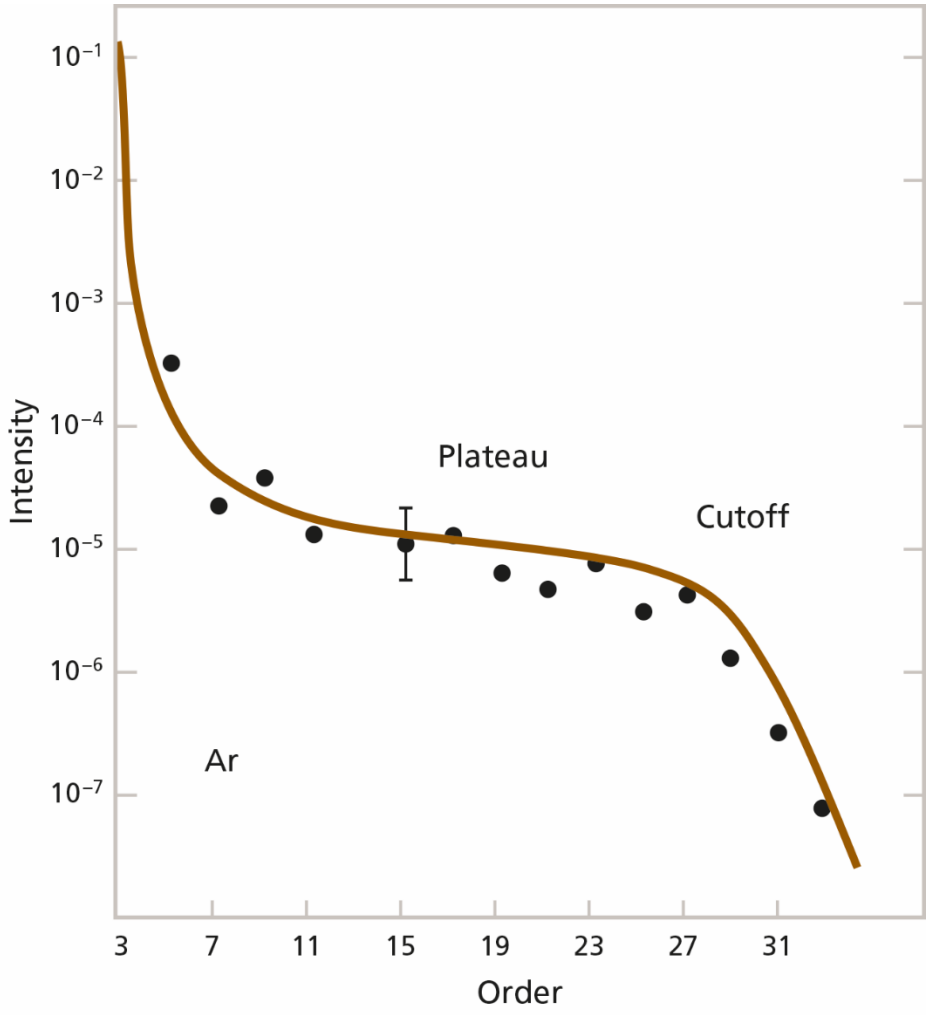
Nd-YAG 1 μm 40 ps





High-order harmonic generation (HHG)





$$\mathcal{P} = a_1\mathcal{E} + a_3\mathcal{E}^3 + a_5\mathcal{E}^5 + \cdots$$

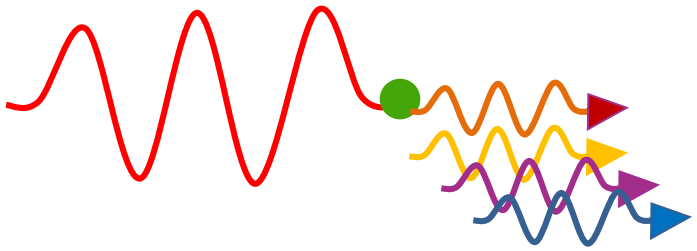
$$a_1\mathcal{E} \gg a_3\mathcal{E}^3 \gg a_5\mathcal{E}^5 \gg \cdots$$

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

McPherson et al. JOSA B **4**, 595 (1987)

Atomic physics and nonlinear optics

Single-atom response



Schrödinger equation

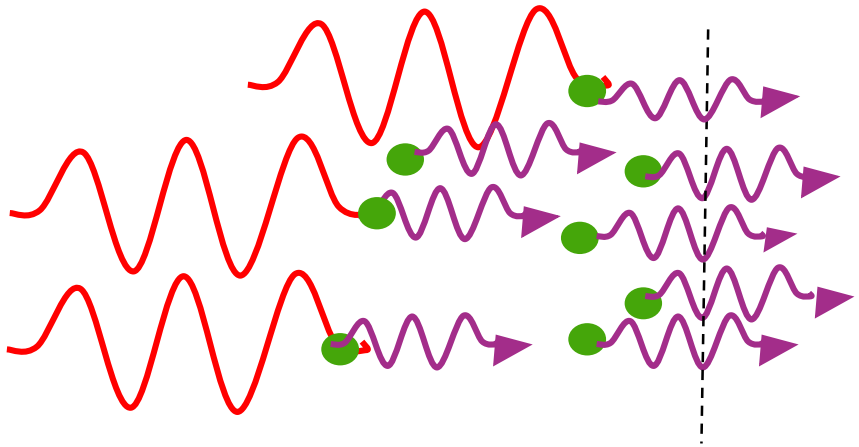
$$i\hbar\frac{\partial\Psi}{\partial t} = -\frac{\hbar^2}{2m}\nabla^2\Psi + \left[V(r) + e\boldsymbol{E}(t)\cdot\boldsymbol{r}\right]\Psi$$

Atomic potential

Laser-atom interaction

Kulander and Shore, Phys. Rev. Lett. **62**, 524 (1989)

Many-atom response



Maxwell equations → Wave equation

$$\nabla^2\mathcal{E} - \frac{1}{c^2}\frac{\partial^2\mathcal{E}}{\partial t^2} = \frac{1}{\epsilon_0 c^2}\frac{\partial^2\mathcal{P}}{\partial t^2}$$

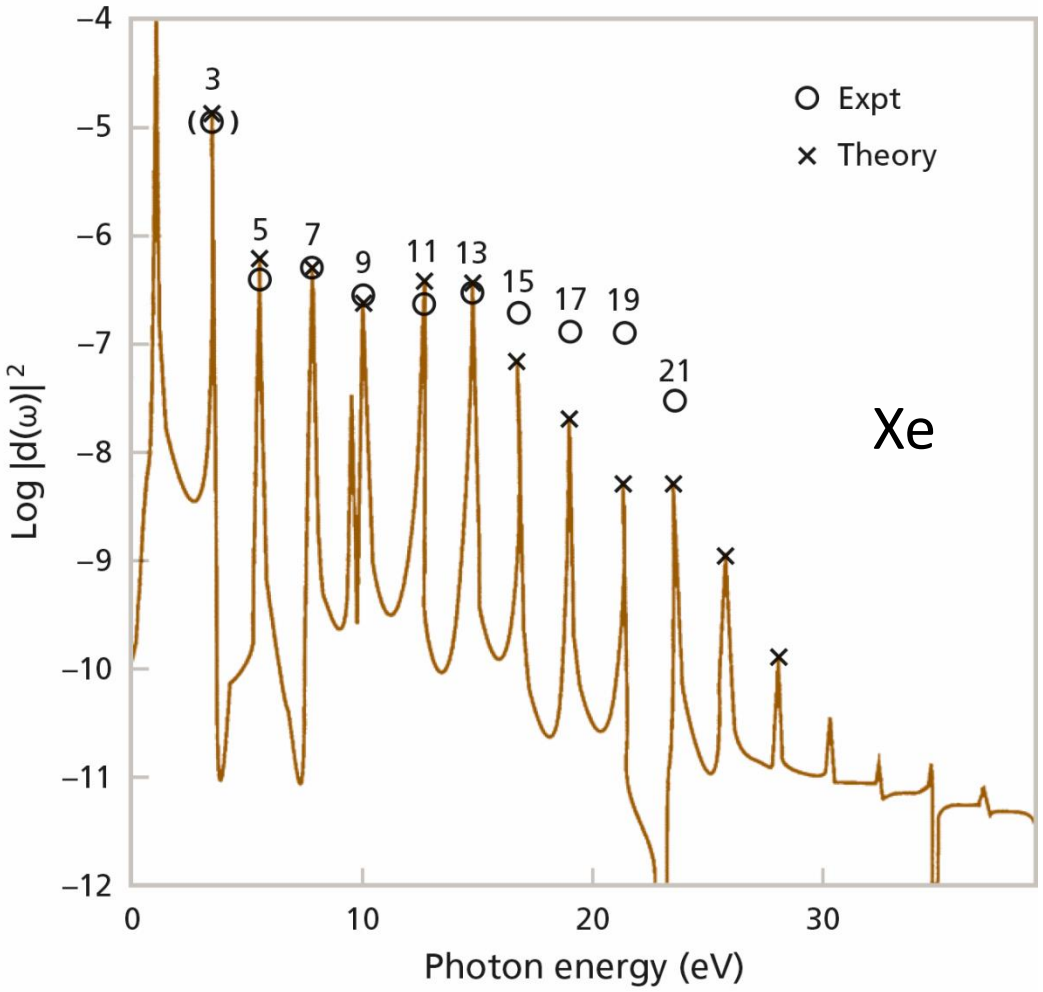
Generated field

Medium Polarization

L'Huillier, Schafer and Kulander, Phys. Rev. Lett. **66**, 2200 (1991)

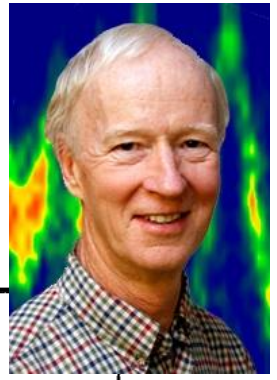
Numerical simulations

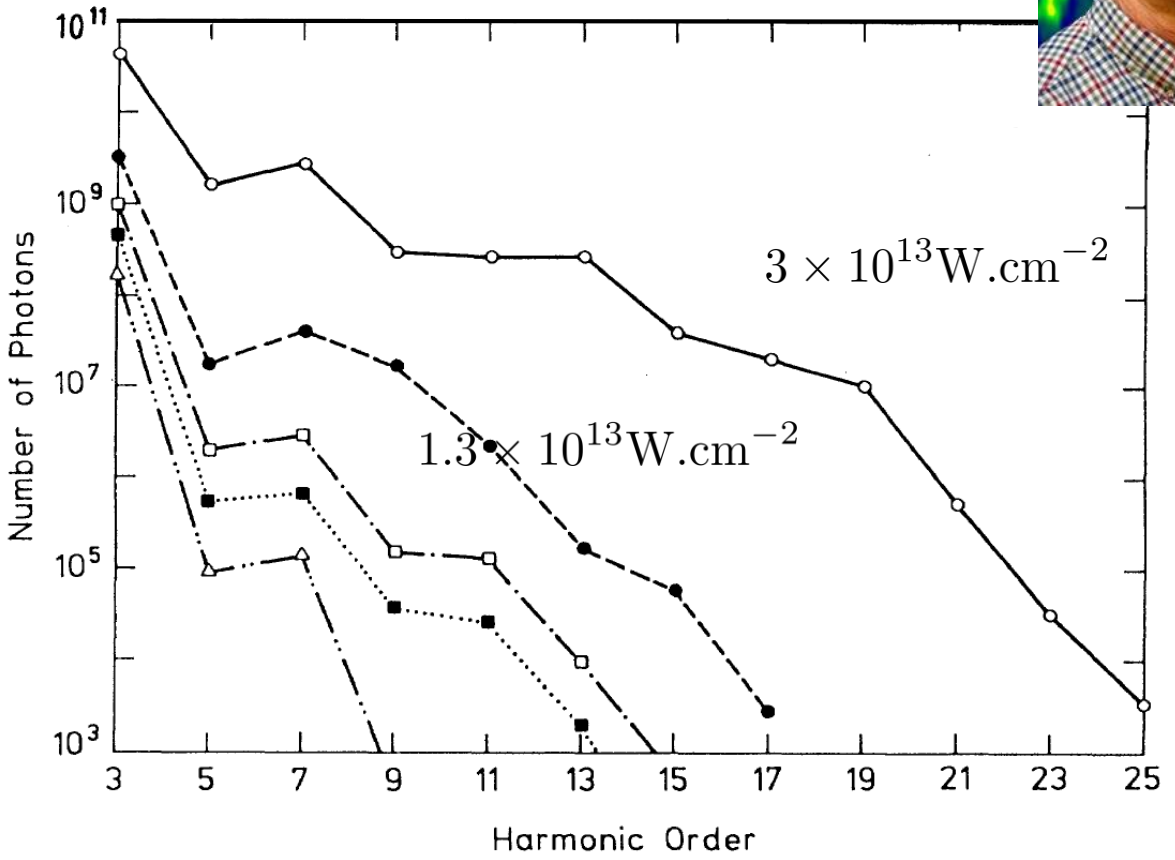
Strong field atomic physics



Kulander and Shore, Phys. Rev. Lett. **62**, 524 (1989)

Strong field nonlinear optics





L'Huillier, Schafer and Kulander, Phys. Rev. Lett. **66**, 2200 (1991)

Progress in laser technology (I)

- Chirped Pulse Amplification

Strickland, Mourou, Opt. Comm. **55**, 447 (1985)

- Titanium sapphire (Ti:S)

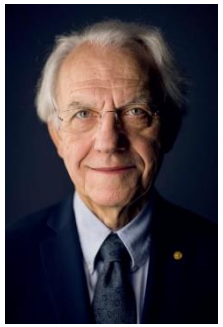
Moulton, J. Opt. Soc. Am. B **3**, 125 (1986)

- Kerr-lens mode-locking

Spence, Kean and Sibbett, Opt. Lett. **16**, 42 (1991)

Sune Svanberg
Anders Persson
Claes-Göran Wahlström

Terawatt Ti:S laser @ Lund Laser Centre



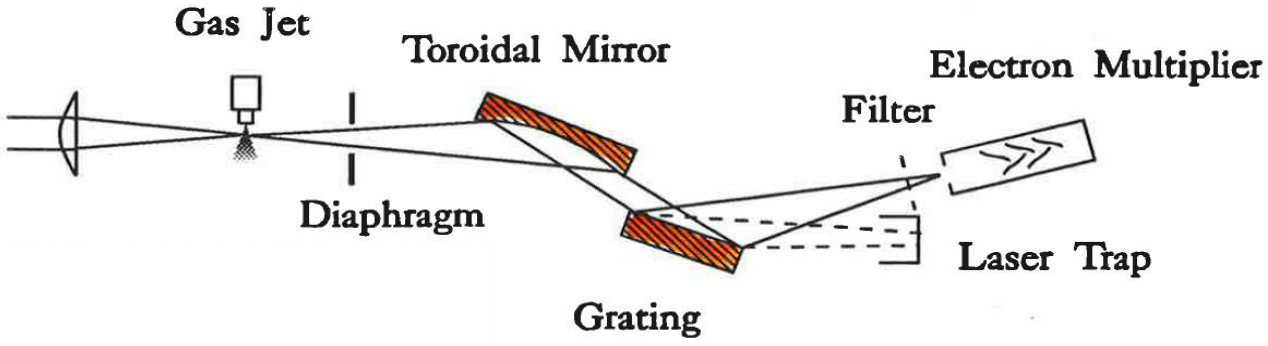
G. Mourou

Donna Strickland

2018

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

Progress in instrumentation

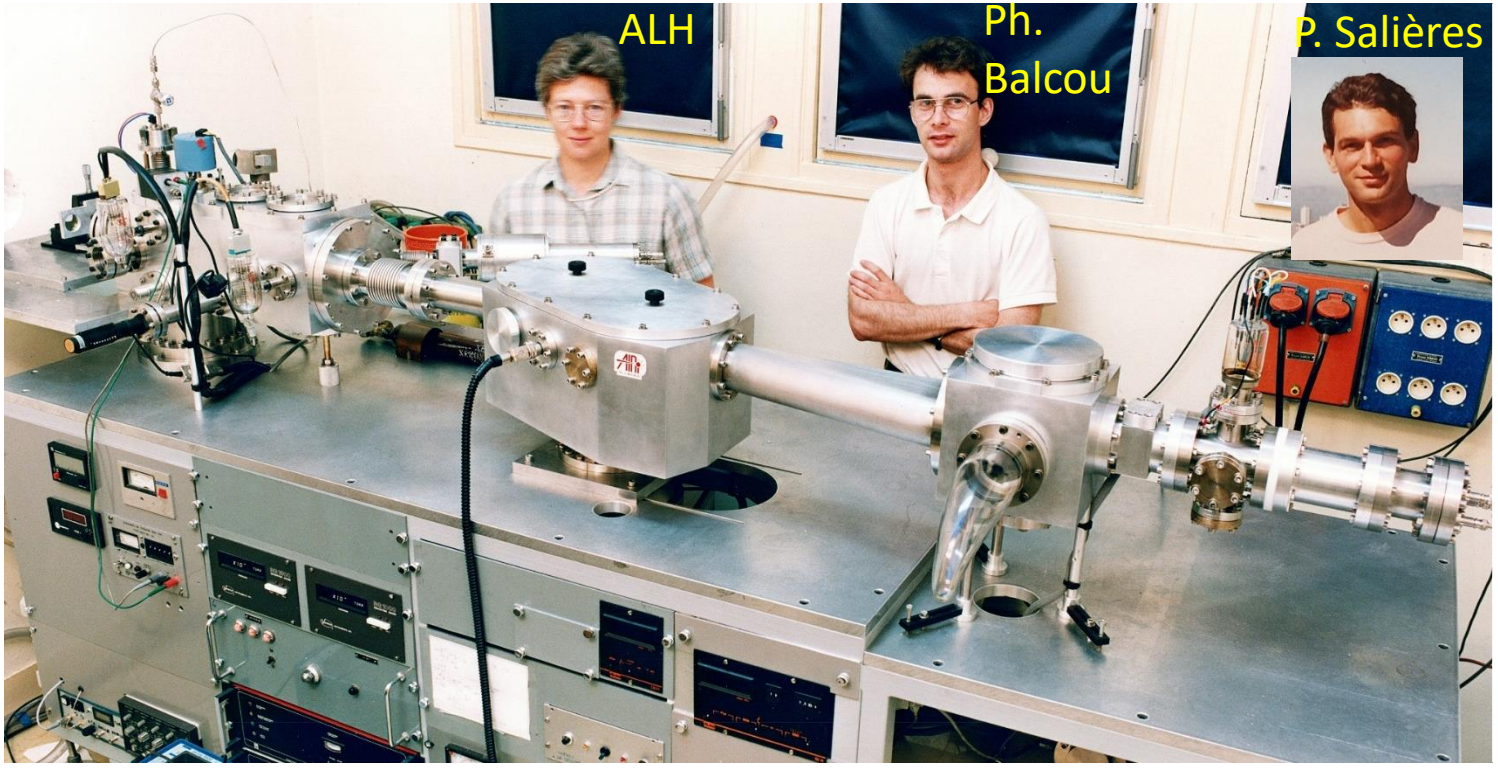


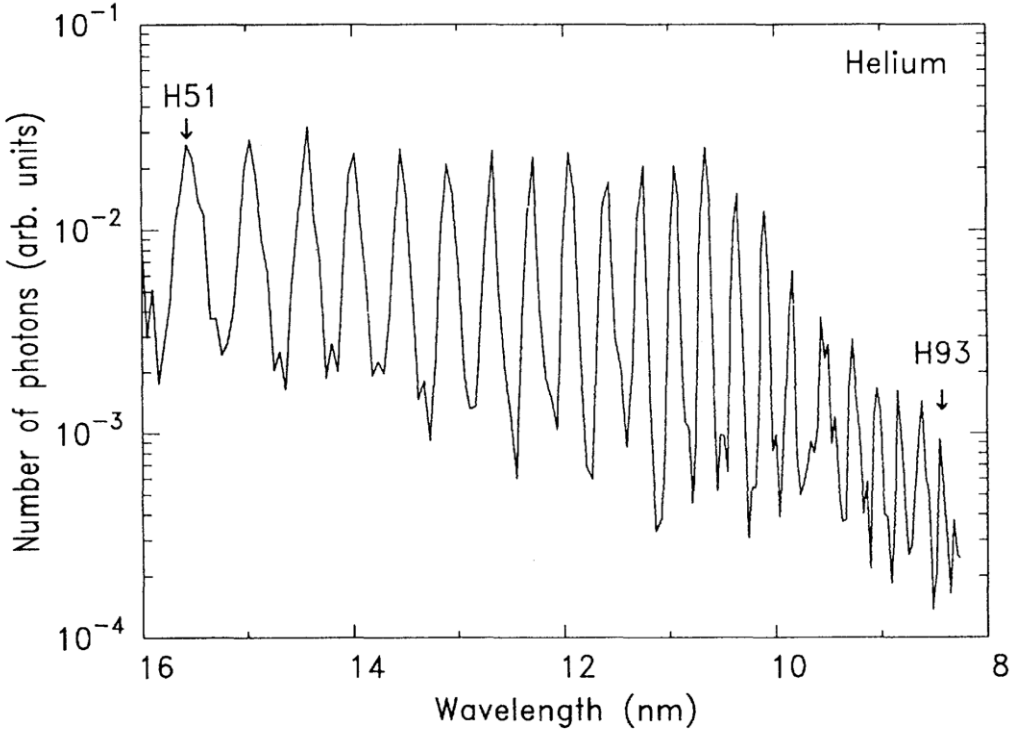
Nd:Glass laser 1 ps 1 shot/minute

L'Huillier and Balcou, Phys. Rev. Lett. **70**, 774 (1993)

Ti: Sapphire 200 fs 10 Hz

Wahlström et al., Phys. Rev. A **48**, 4709 (1993)

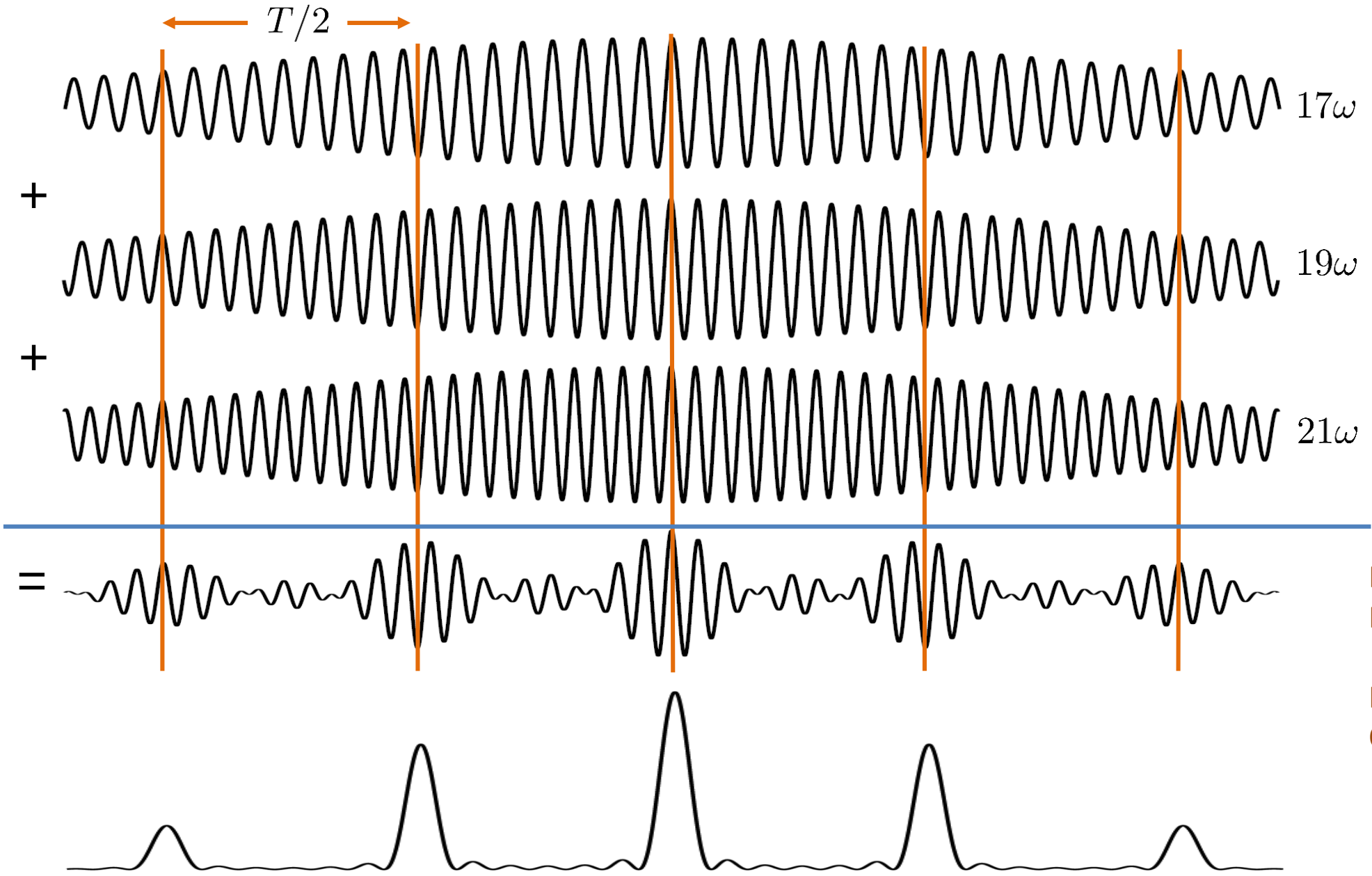




Outline

- High-order harmonic generation
- **Attosecond light pulses**
- Applications

Attosecond pulses?



High frequency

$$T = \frac{2\pi}{\omega}$$

Broad bandwidth

$$\tau \propto \frac{1}{\Delta\omega}$$

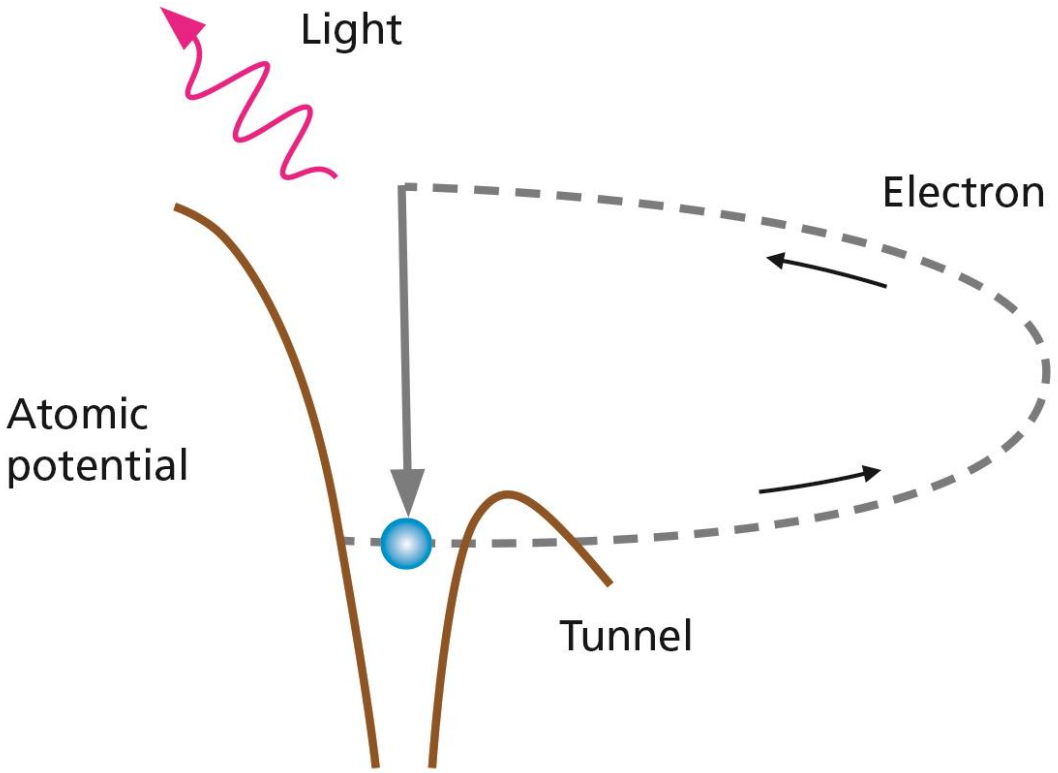
Are the harmonics phase-locked?

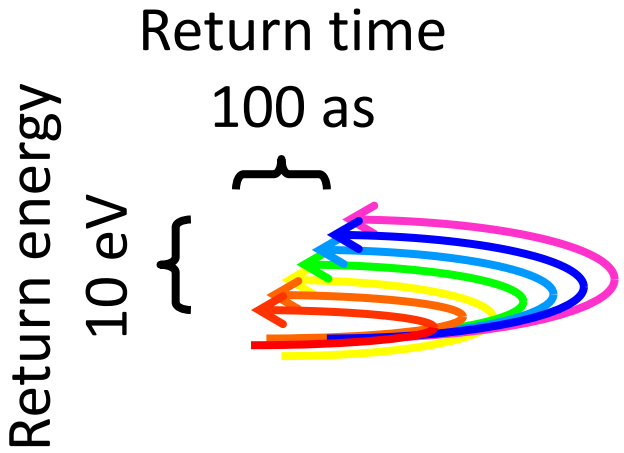
Farkas and Toth,
Phys. Lett. A **168**, 447 (1992)

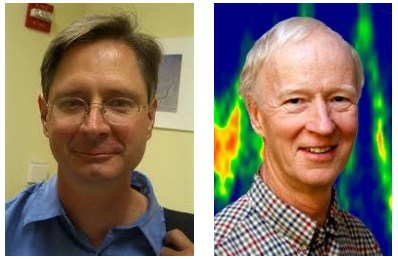
Harris, Macklin and Hänsch,
Opt. Comm. **100**, 487 (1993)

Progress in understanding: Strong field atomic physics

The three-step model







Kulander et al. Proc. SILAP, Han-sur-Lesse (1993)

Corkum Phys. Rev. Lett. **71**, 1994 (1993)

Lewenstein et al. Phys. Rev. A **49**, 2117 (1994)

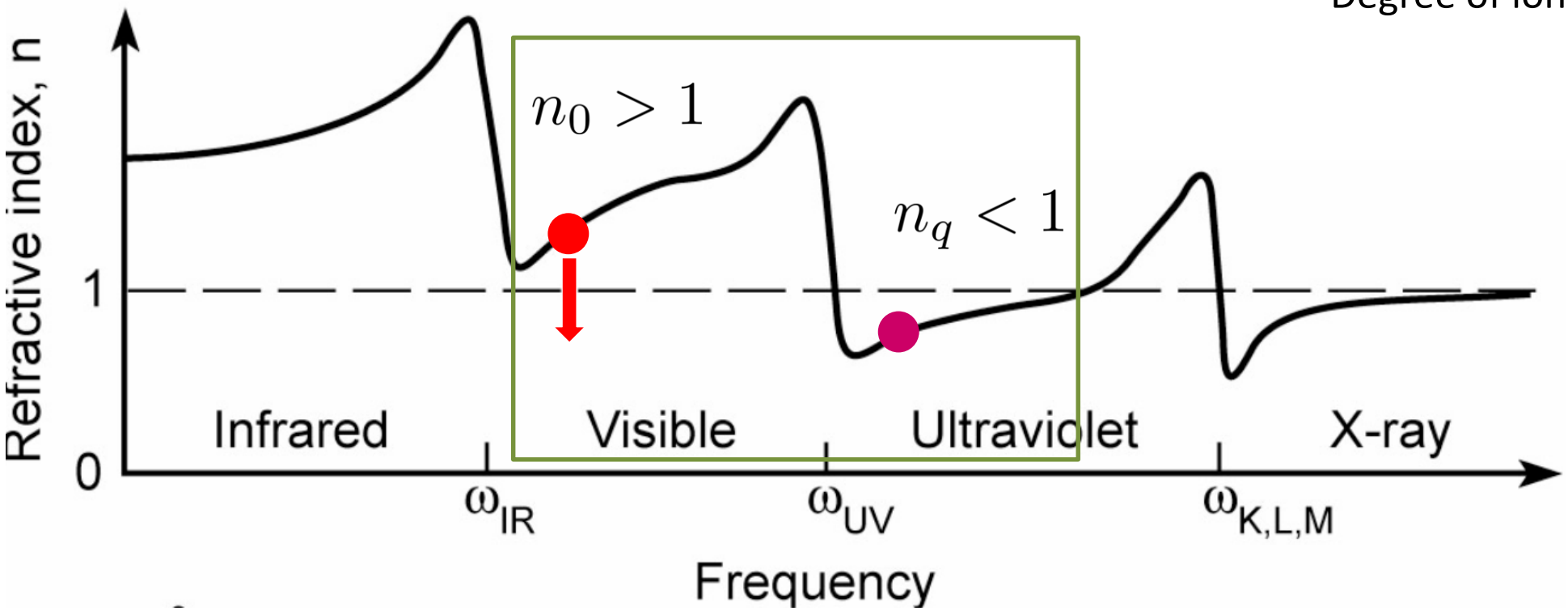
Progress in understanding: Phase matching

Phase velocity of the fundamental =
Phase velocity of the harmonic fields

$$\frac{c}{n_0} = \frac{c}{n_q}$$

$$n_0 = 1 + \frac{\mathcal{N}_n \alpha}{2\epsilon_0}$$

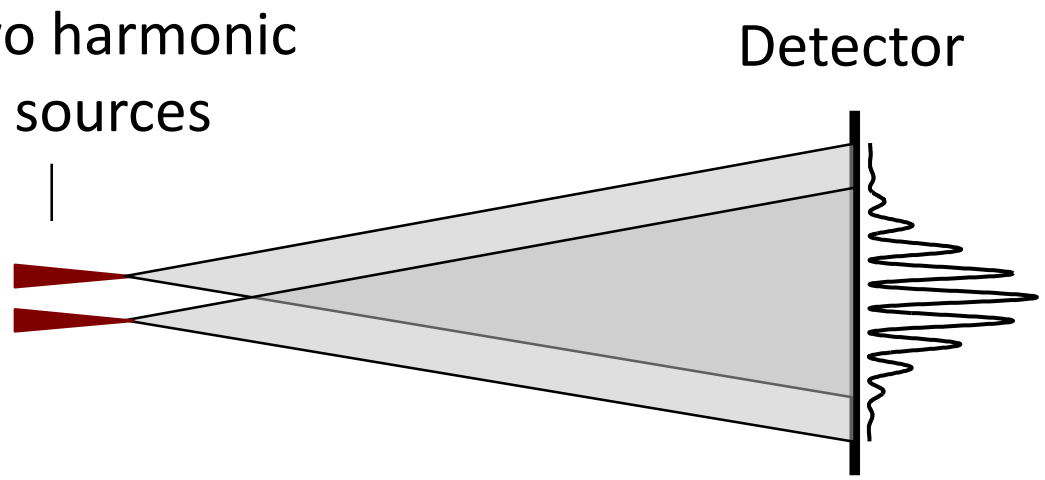
Degree of ionization = a few %



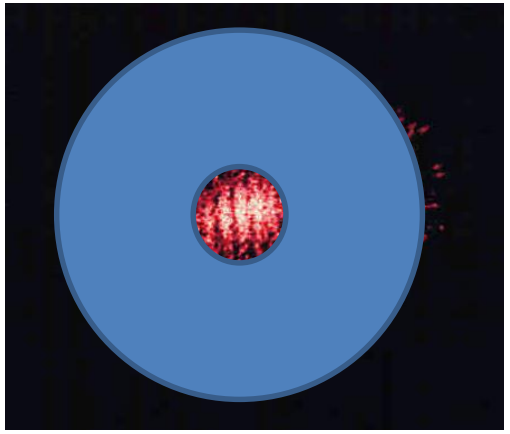
Influence of focusing

Attwood, ”X-ray and extreme ultraviolet radiation”, 2017

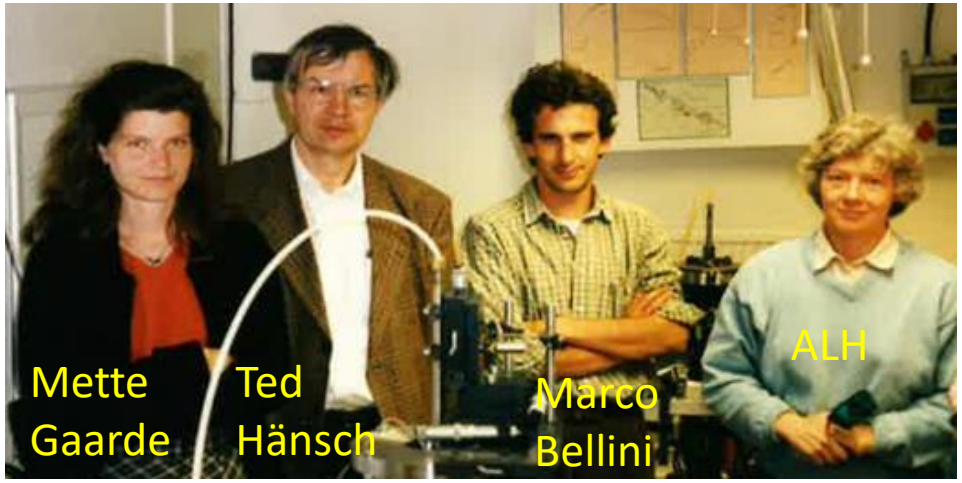
Progress in experiment: Short and long trajectories



Harmonic 15



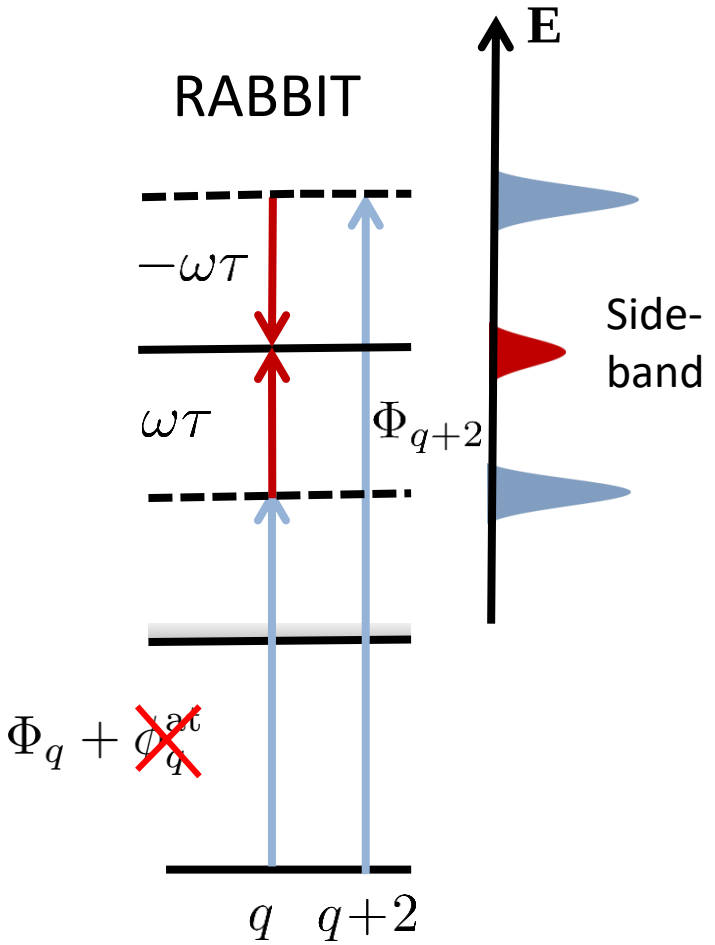
Short and long trajectories

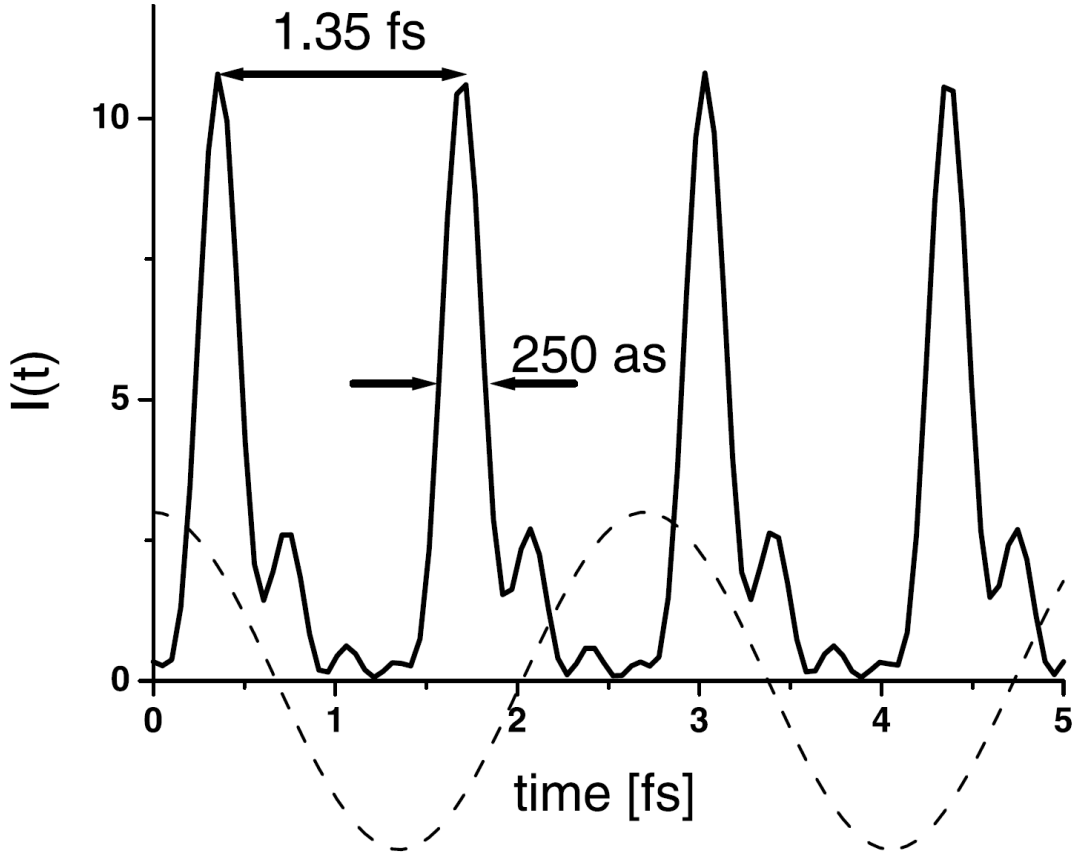


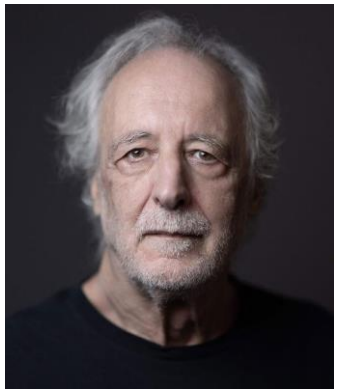
Claire Lyngå, Claes-Göran Wahlström

Bellini et al. Phys. Rev. Lett. **81**, 297 (1998)

First measurement of attosecond pulses in a train







Pierre Agostini

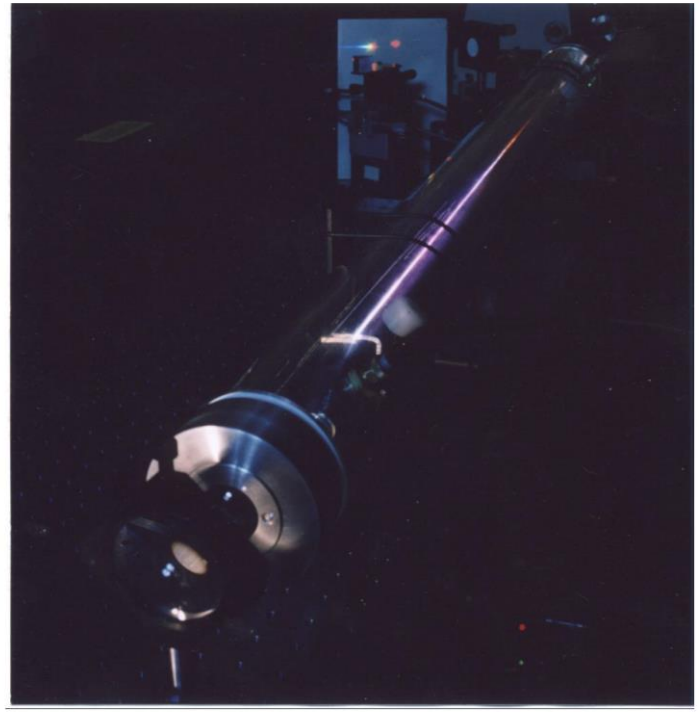
$$S \propto a + b\cos(2\omega\tau + \Phi_q - \Phi_{q+2})$$

Véniard et al., Phys. Rev. A **54**, 721 (1996)

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

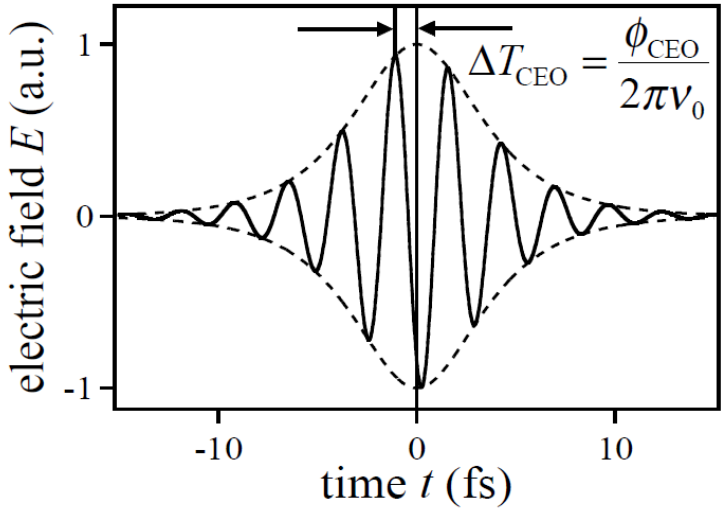
Progress in laser technology (II)

Few-cycle laser pulses (< 10 fs)
Hollow fiber compression
Chirped mirrors



Nisoli et al. Opt. Lett. **22**, 522 (1997)
Kärtner et al., Opt. Lett. **22**, 831 (1997)

Measurement and control of the Carrier-Envelope Phase

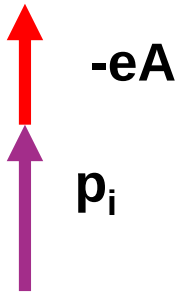


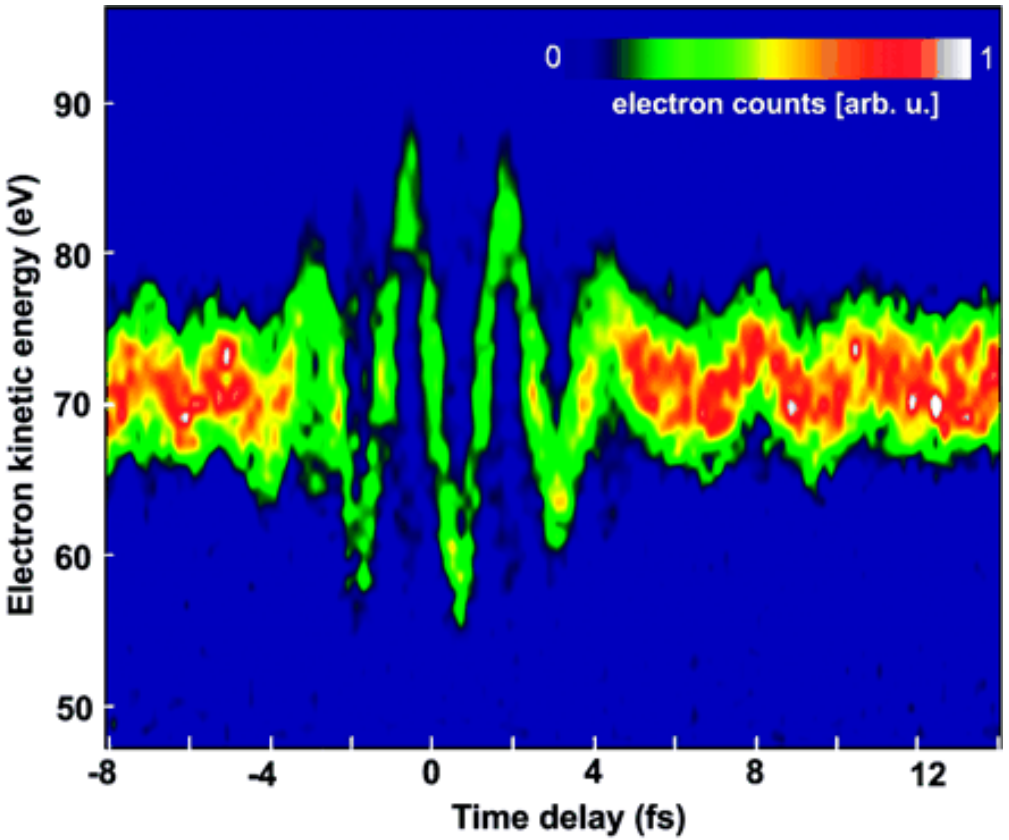
Telle et al. Appl. Phys. B **69**, 327 (1999)

First measurement of isolated attosecond pulses

Ferenc Krausz

Streaking camera





τ=250 as

Henstchel et al., Nature **414**, 509 (2001)

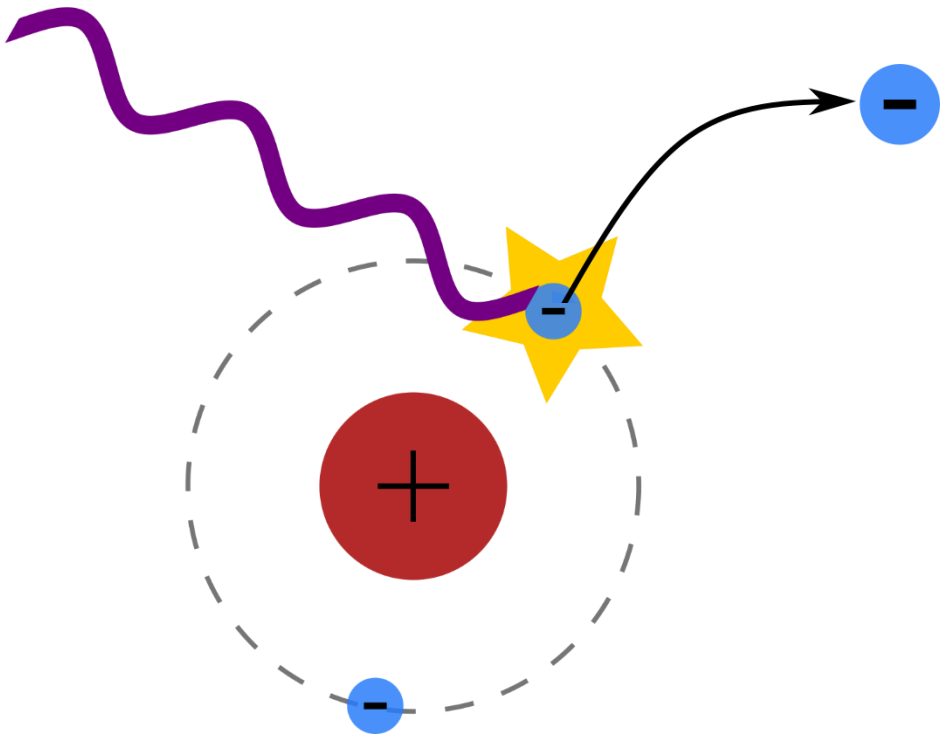
Goulielmakis et al., Science **305**, 1267 (2004)

Outline

- High-order harmonic generation
- Attosecond light pulses
- **Applications**

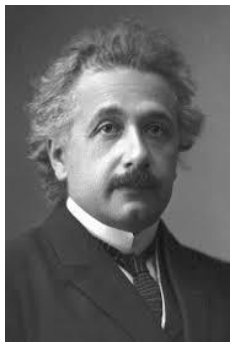
  - Atomic Spectroscopy
  - Industrial application

Temporal dynamics of photoionization



Photoelectric effect
Hertz 1887
Einstein 1905

What are the
wave/quantum properties
of the photoelectron?



A. Einstein

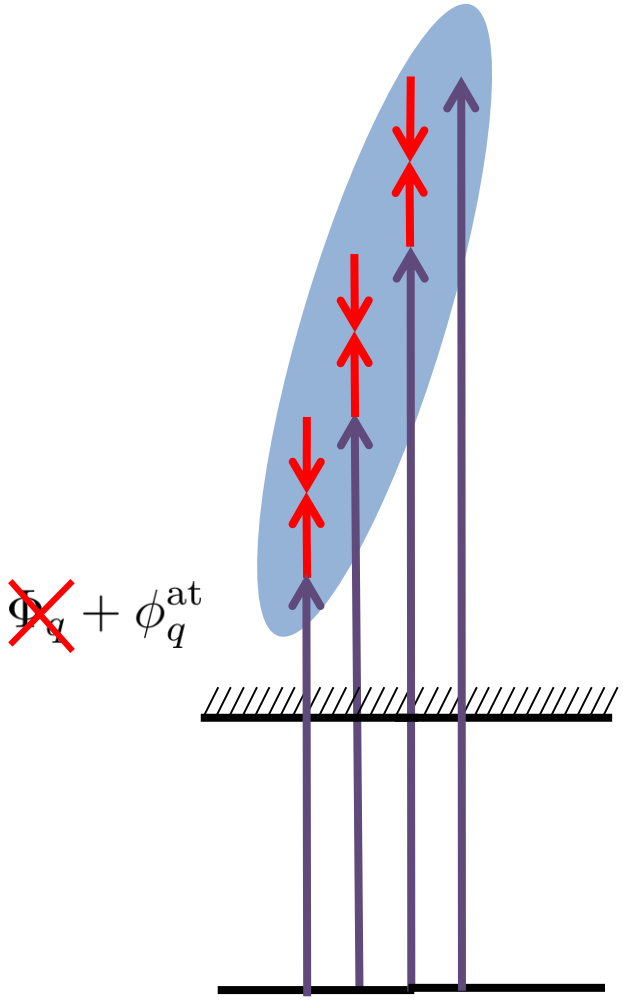
"for his services to Theoretical Physics, and especially for his discovery of the law of the photoelectric effect"

1921

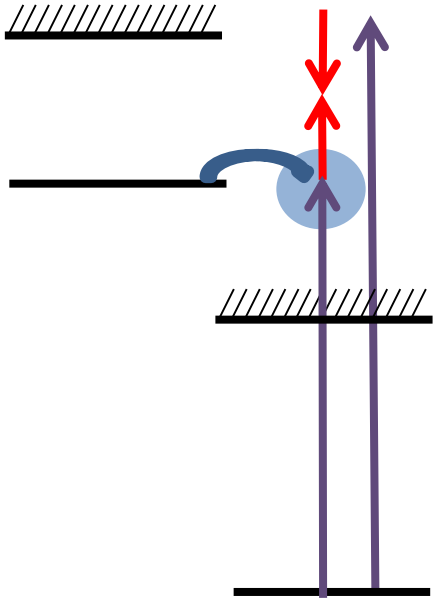
@NobelPrize.org

Measuring the phase of a photoelectron

Non-resonant attosecond excitation



Discrete state



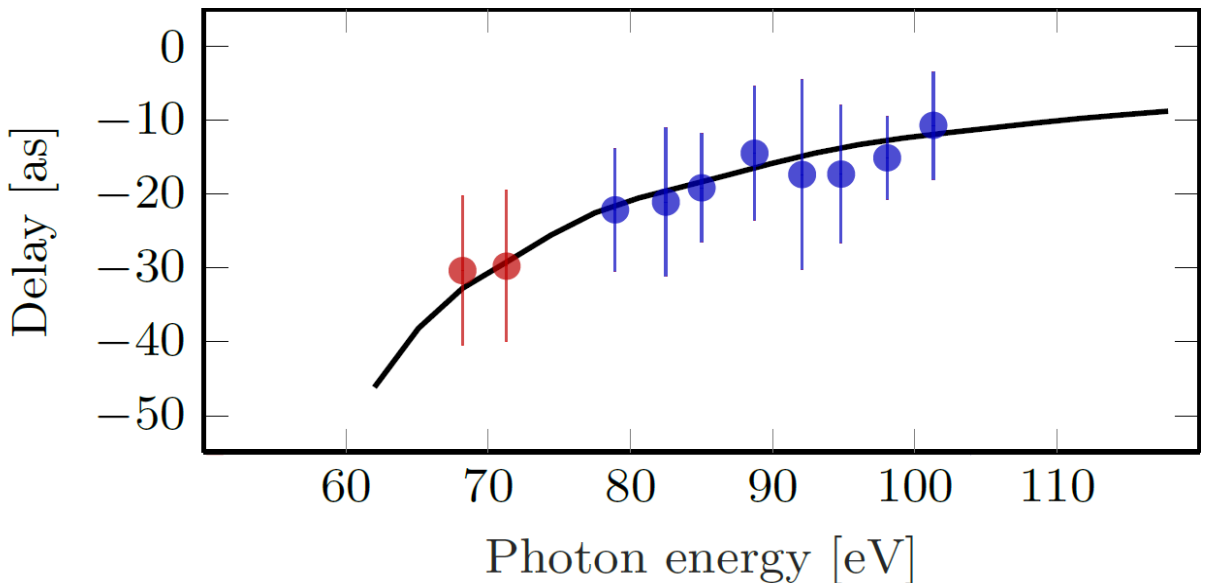
Resonant harmonic excitation

Cavalieri et al., Nature **449**, 1029 (2007)
Schultze et al., Science **328**, 1658 (2010)
Klünder et al., Phys. Rev. Lett. **106**, 143002 (2011)

Haessler et al. Phys. Rev. A **80**, 011404 (2009)
Kotur et al., Nat. Comm. **7**, 10566 (2016)
Gruson et al., Science **354**, 734, (2016)

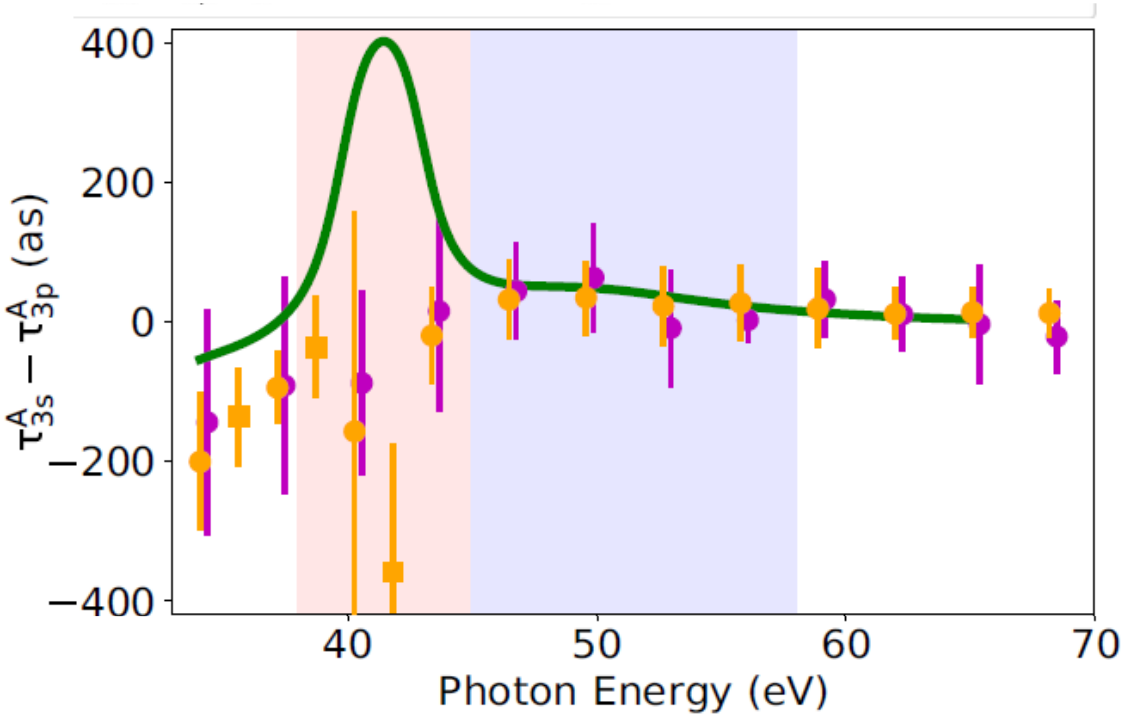
Differences in photoionization time delays

Ne 2s and 2p ionization



Schultze et al., Science **328**, 1658 (2010)
Isinger et al., Science **358**, 893 (2017)

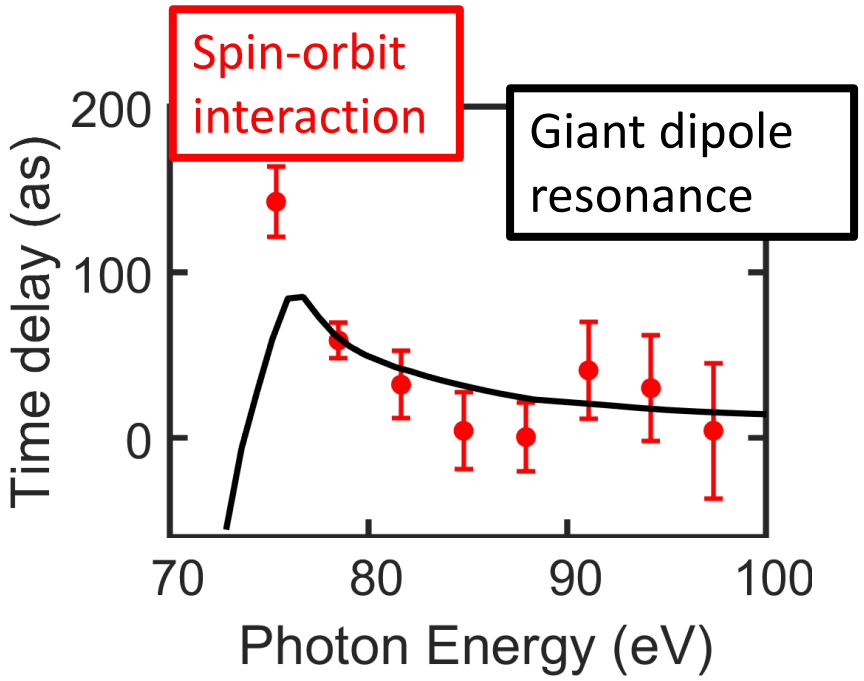
Ar 3s and 3p ionization



Klünder et al., Phys. Rev. Lett. **106**, 143002 (2011)
Alexandridi et al., Phys. Rev. Res. **3**, L012012 (2021)

Photoionization time delays

Xe 4d shell ionization



Zhong et al., Nat Comm **11**, 5042 (2020)

→ Angular distributions

Heuser et al., Phys. Rev. A **94**, 063409 (2016)
Fuchs et al., Optica **7,** 154 (2020)
Peschel et al., Nat. Comm. **13**, 5205 (2022)

→ Measurement of the density matrix of a photoelectron

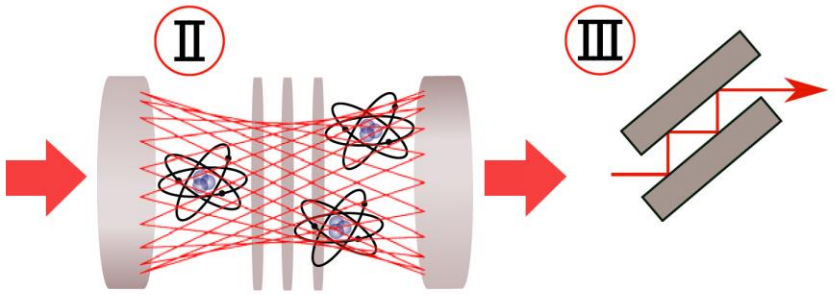
Bourasssin-Bouchet et al., Phys. Rev. X **10**, 031048 (2020)
Laurell et al., Phys. Rev. Res. **4**, 033220 (2022)
Laurell et al., arxiv.org/abs/2309.13945

Progress in laser technology (III)

Ytterbium laser @1030 nm
10 -100 W, 200 fs

High repetition rate > 100 kHz
High average power

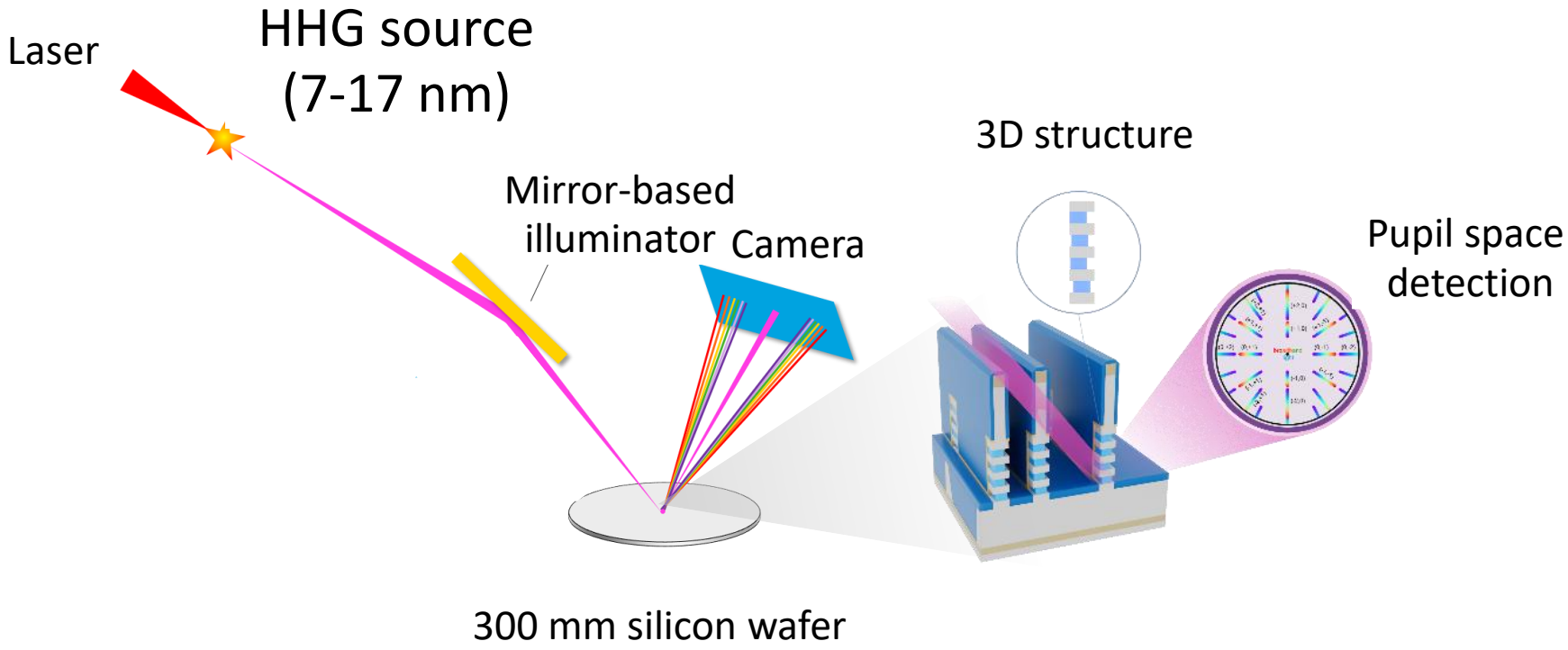
Post-compression in a multipass cell



Hönninger et al, Appl. Phys. B **69**, 3 (1999)

Schulte et al., Opt. Lett. **41**, 4511 (2016)
Viotti et al., Optica **9**, 197 (2022)

Application in semiconductor industry



Metrology of chips based on soft X-ray scatterometry

Courtesy

ASML

Supplier of lithography and metrology machines to the semiconductor industry

Porter et al., Proc. SPIE 12496, Metrology, Inspection and Process Control, XXXVII, 1249611 (2023)

Next chapter in attosecond science

Applications in many fields

- Atomic spectroscopy
- Attosecond chemistry
- Condensed matter physics
- Industrial applications
- Ultrafast quantum dynamics

Attosecond sources via HHG: XUV range, low energy/pulse (nJ), often in a train

Attosecond sources via FEL: X-ray range, high energy/pulse (μJ), isolated

The prize ceremony

The Nobel banquet

Thank you!

Johan Mauritsson
Per Eng-Johnsson
Cord Arnold
Mathieu Gisselbrecht
Anne-Lise Viotti
David Busto

Thank you!

swiss physical society

Vetenskapsrådet

erc

Knut och Alice Wallenbergs Stiftelse

Swiss Society for Photon Science