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The route to attosecond pulses

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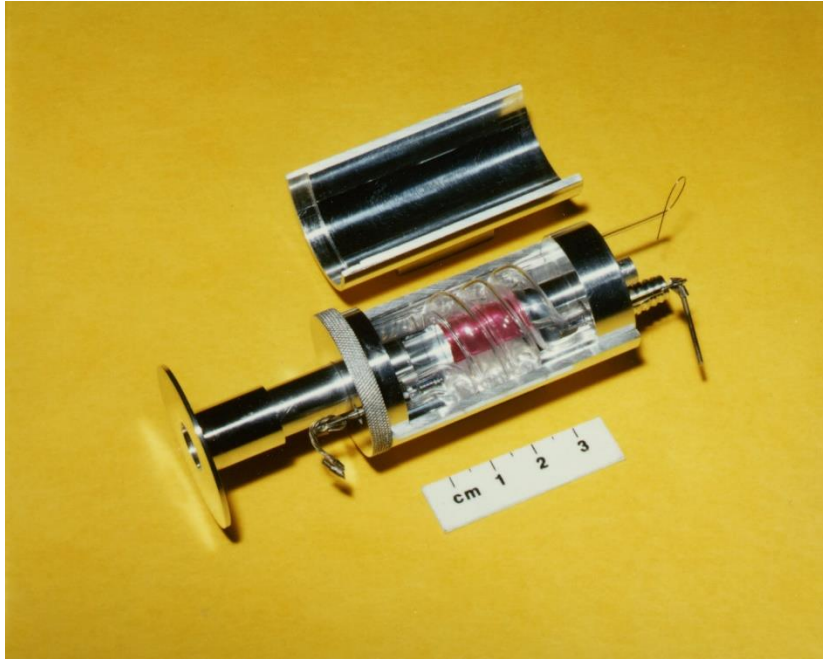


Outline

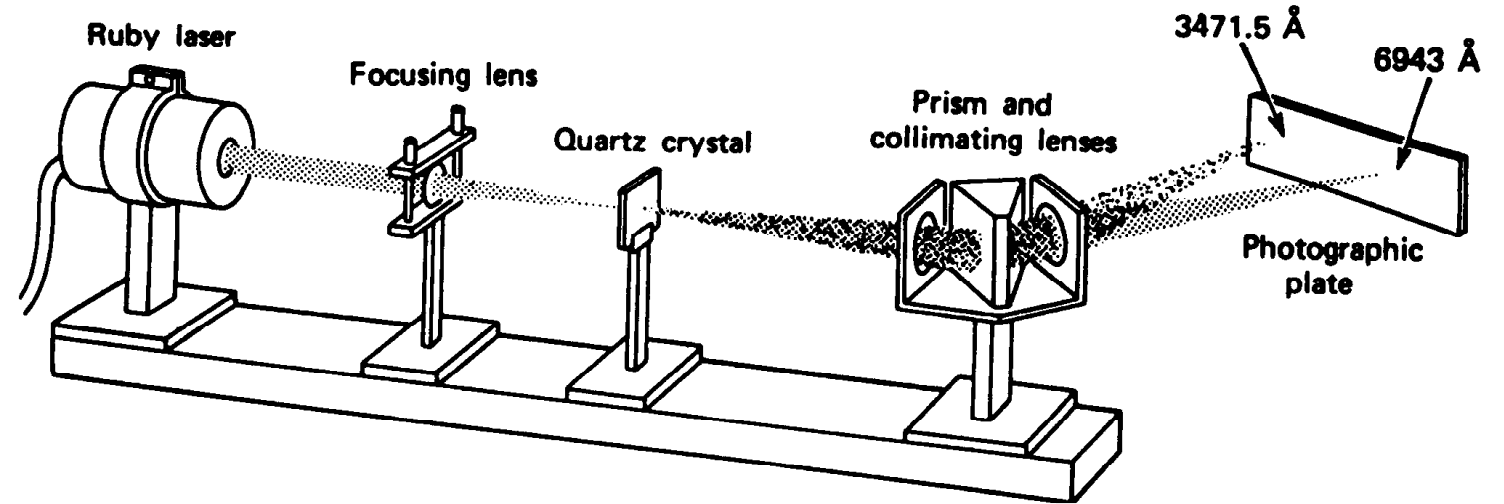
- **High-order harmonic generation**
- **Attosecond light pulses**
- **Attosecond physics**

The invention of the laser

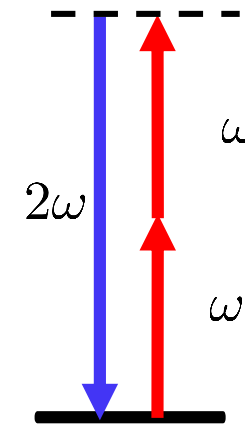
Nonlinear optics



Lejournal.cnrs.com

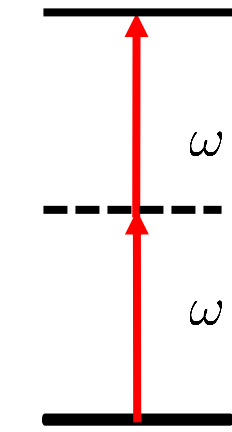


New frequencies

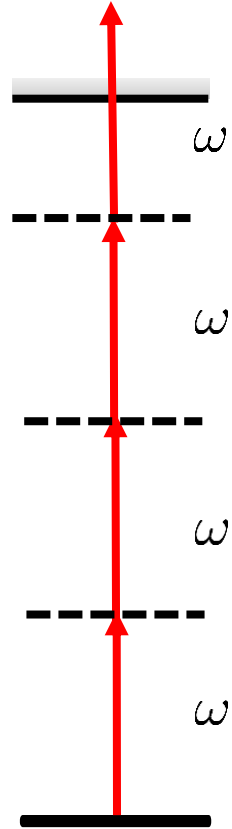


Atoms in strong laser fields

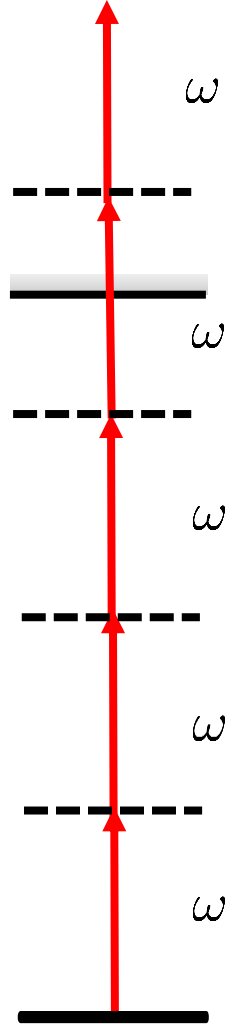
Multiphoton processes



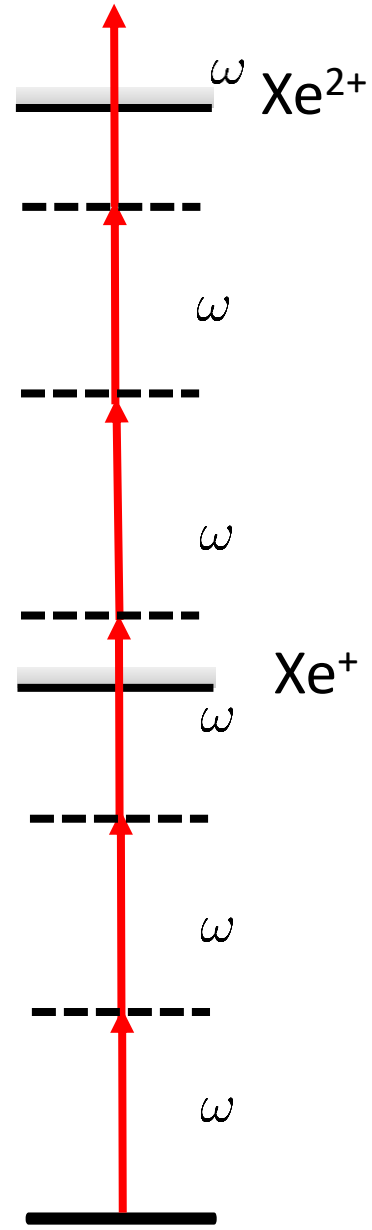
Excitation



Ionization



Above-threshold
-ionization



Multiple
ionization



@NobelPrize.org

Predicted in
1931 by

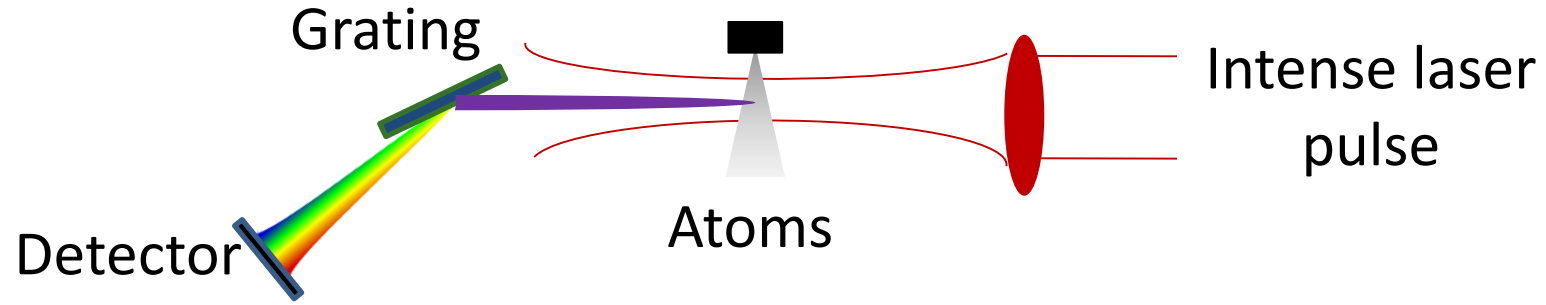
Maria Goeppert-Mayer

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

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

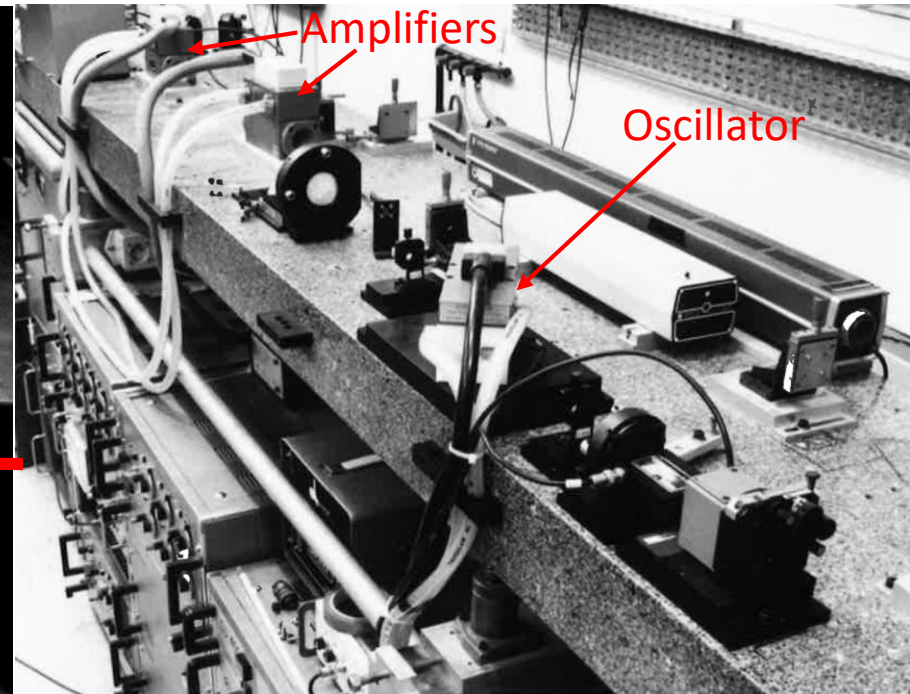
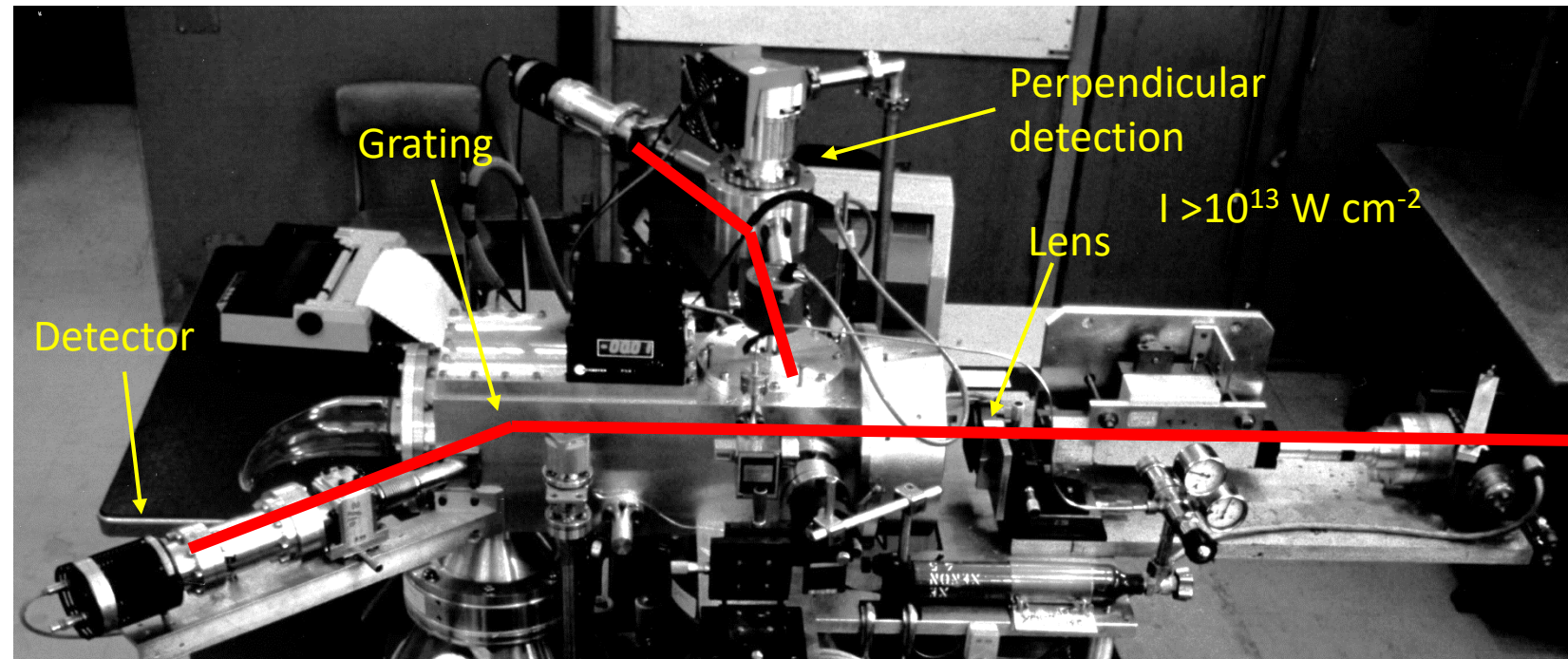
L'Huillier et al. Phys. Rev. A **27**,
2503 (1983)

Fluorescence?

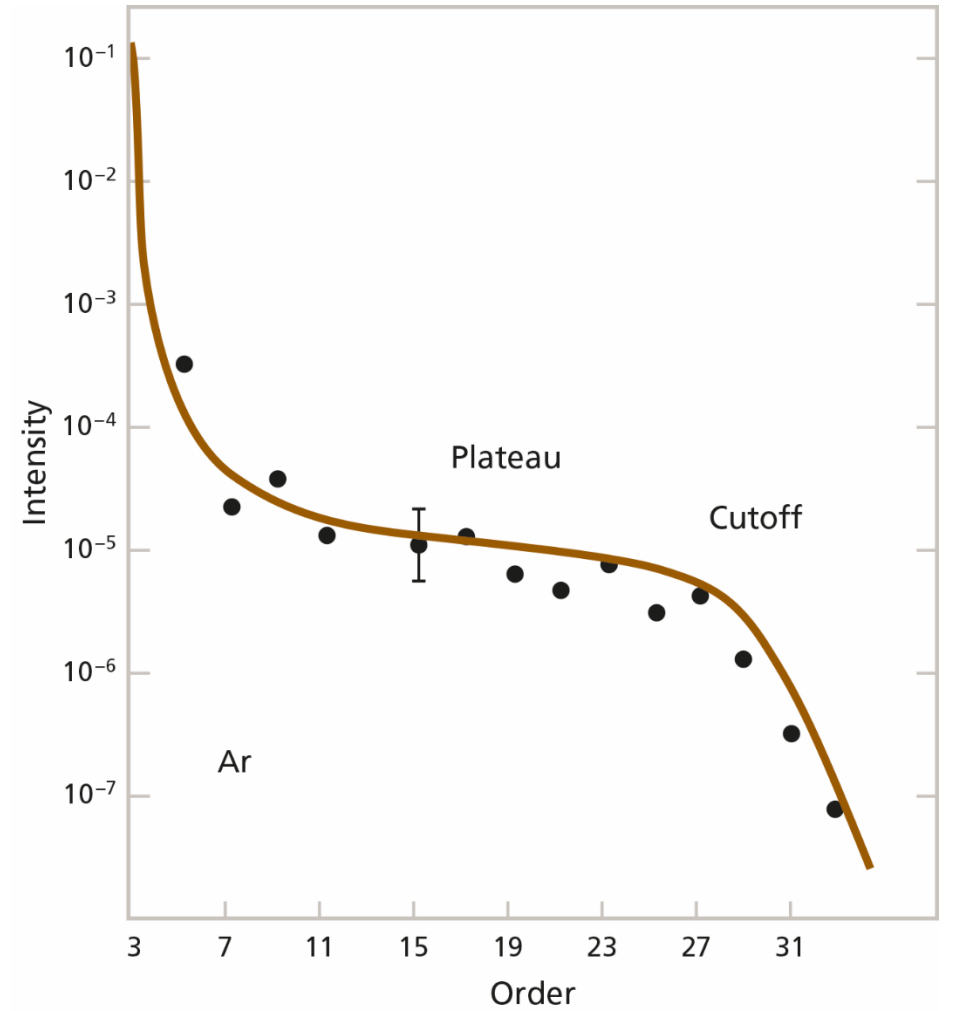
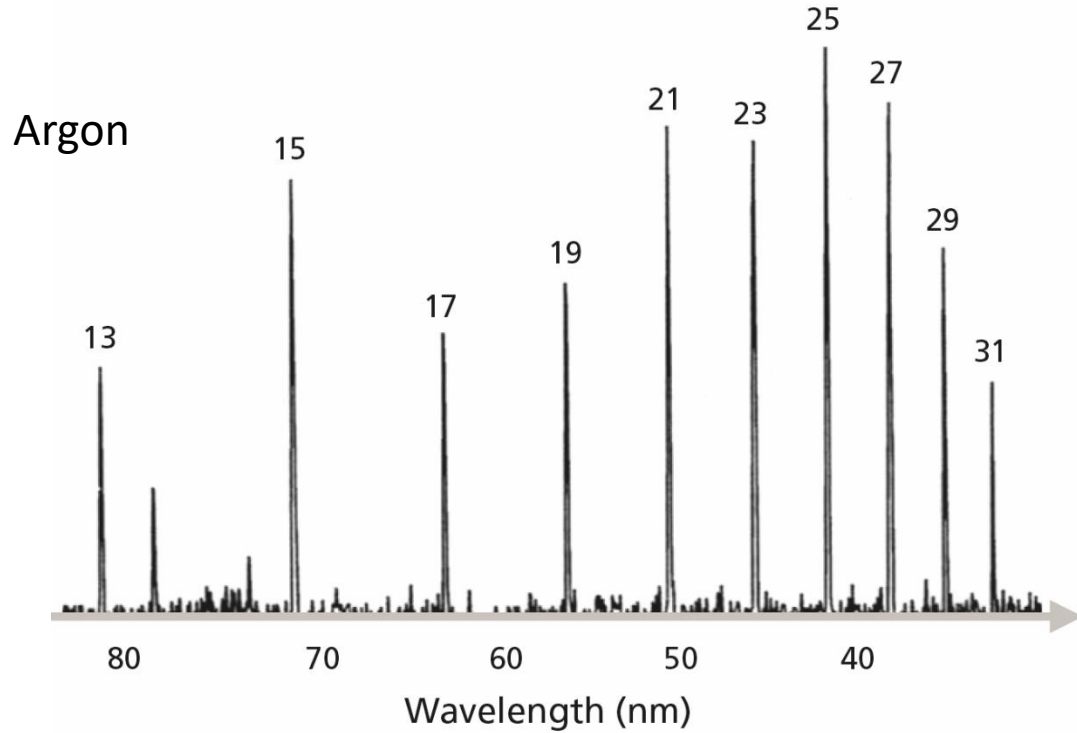


G. Mainfray
C. Manus

Nd-YAG 1 μm 40 ps



High-order harmonic generation (HHG)



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

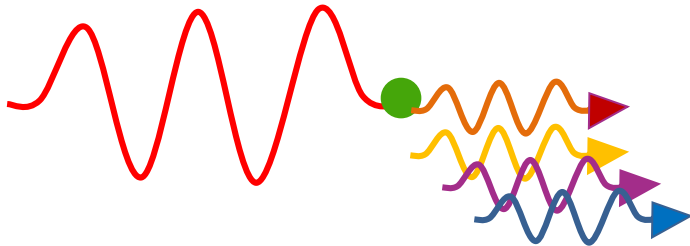
$$a_1 \mathcal{E} \gg a_3 \mathcal{E}^3 \gg a_5 \mathcal{E}^5 \gg \dots$$

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

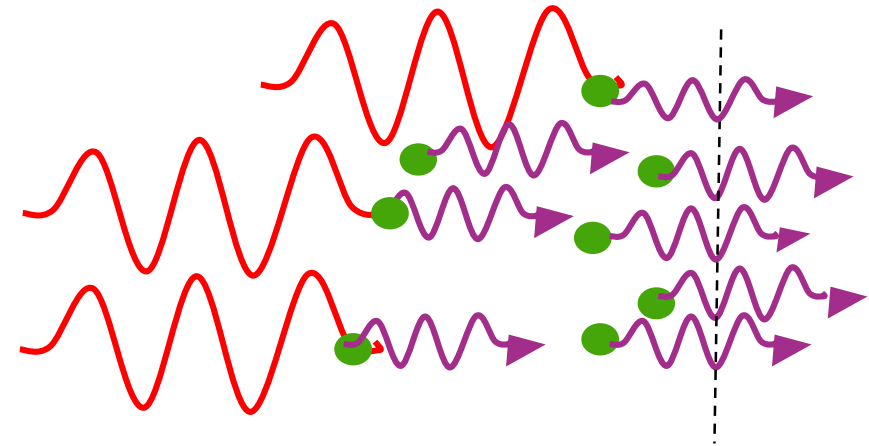
$$i\hbar \frac{\partial \Psi}{\partial t} = -\frac{\hbar^2}{2m} \nabla^2 \Psi + [V(r) + e\mathbf{E}(t) \cdot \mathbf{r}] \Psi$$

Atomic potential

Laser-atom interaction

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

Many-atom response



Maxwell equations → Wave equation

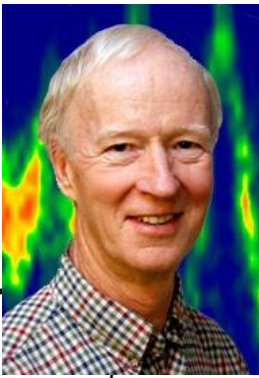
$$\nabla^2 \mathcal{E} - \frac{1}{c^2} \frac{\partial^2 \mathcal{E}}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \mathcal{P}}{\partial t^2}$$

Generated field

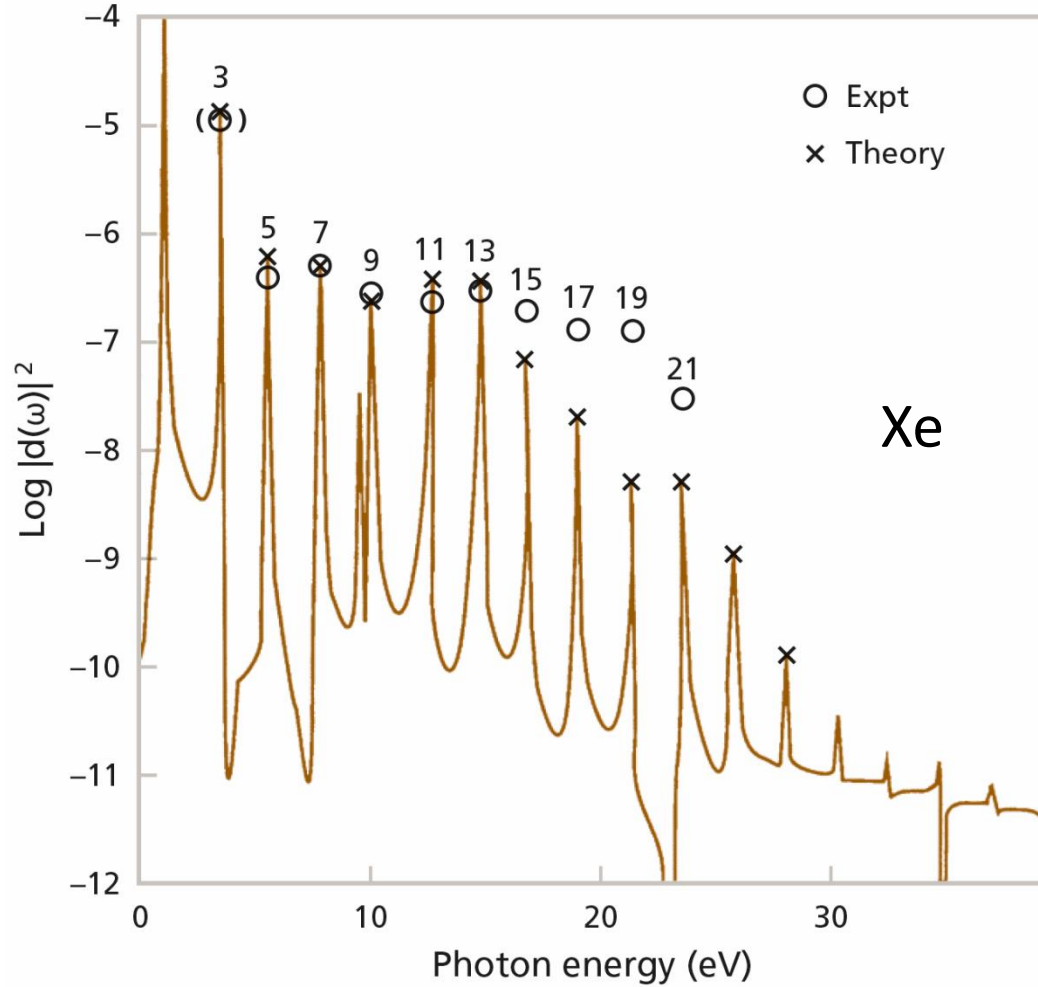
Medium Polarization

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

Numerical simulations

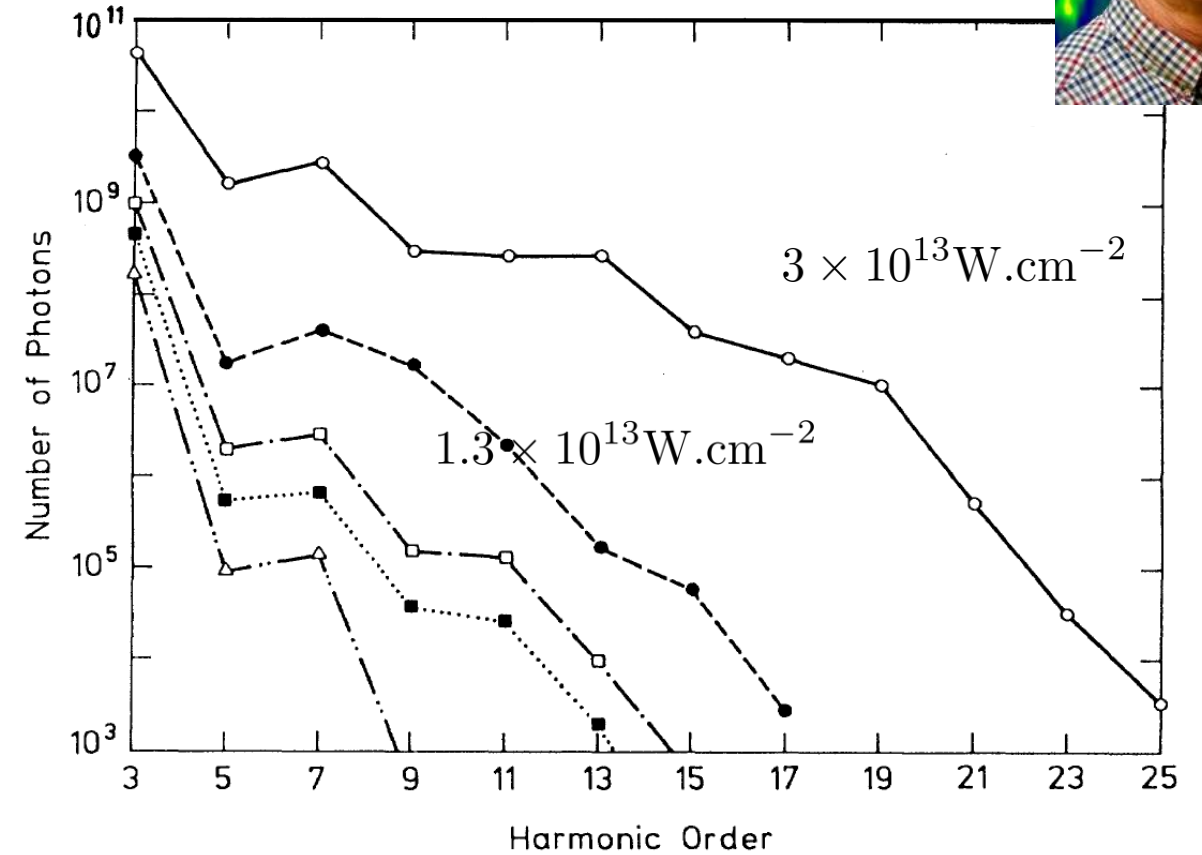


Strong field atomic physics



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)

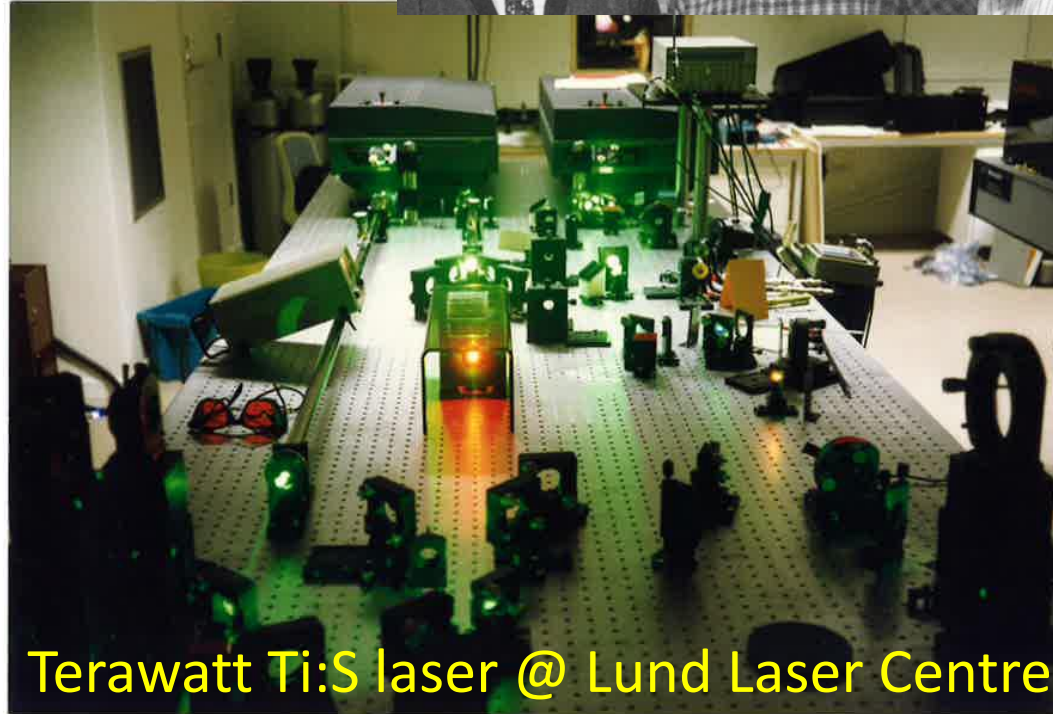
➤ Titanium sapphire (Ti:S)

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



G. Mourou

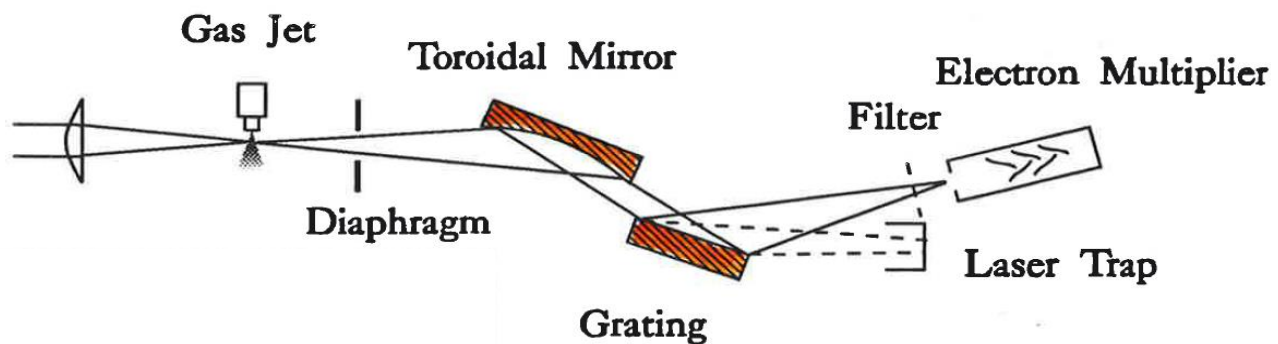
Donna
Strickland



2018

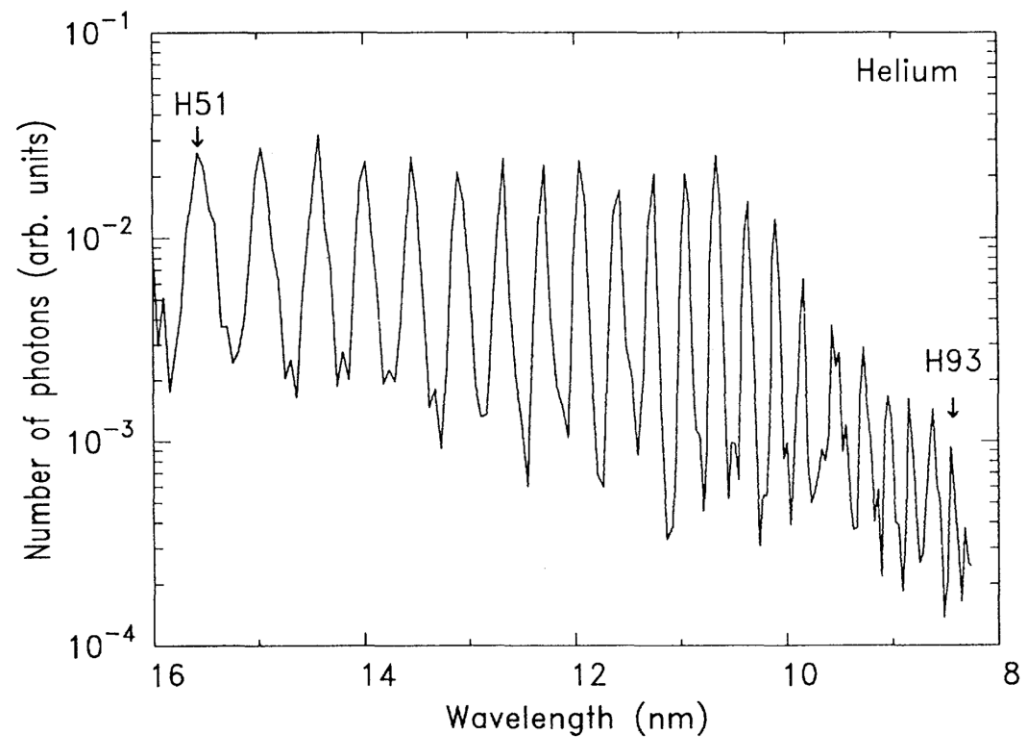
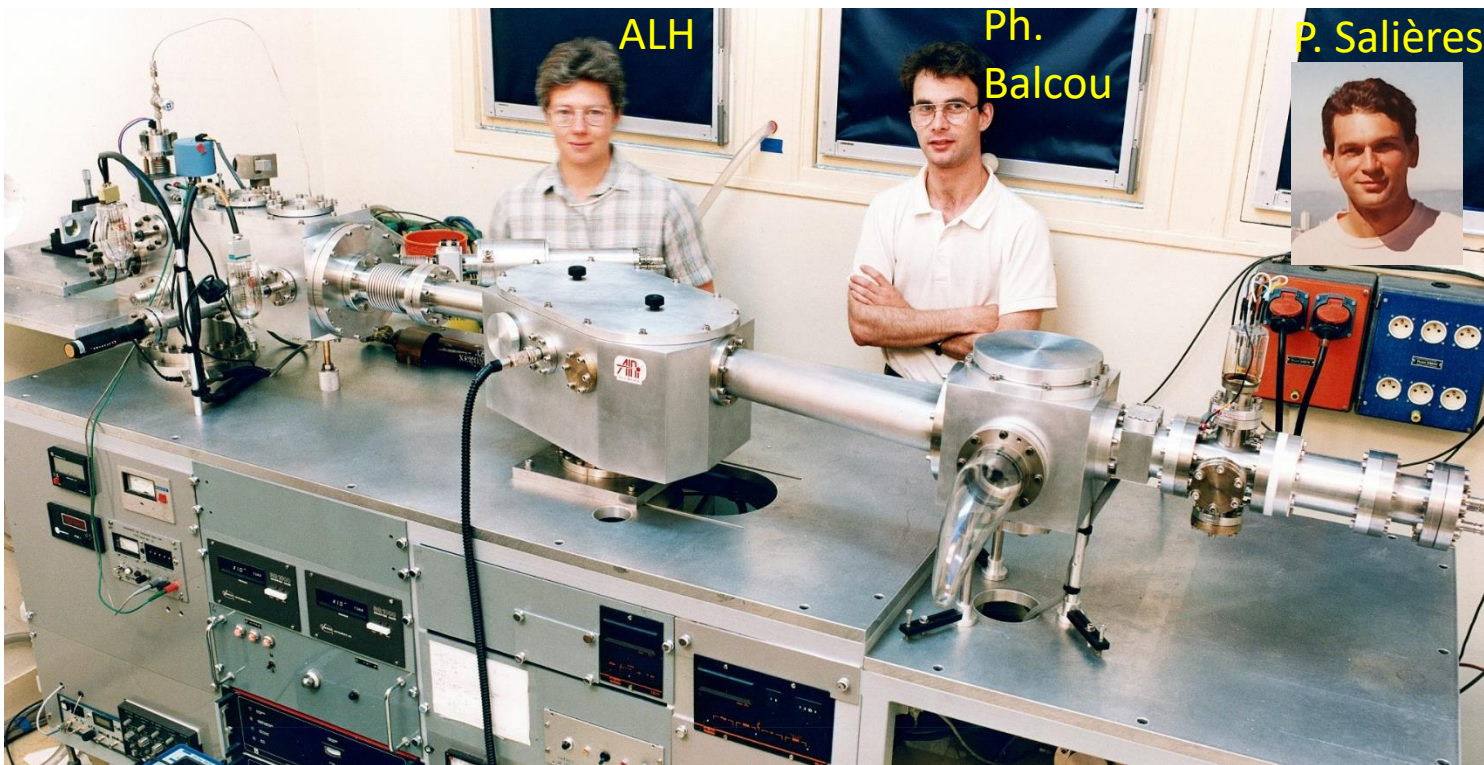
“for their method of generating high-intensity, ultra-short optical pulses”

Progress in instrumentation



Nd:Glass laser 1 ps 1 shot/minute
L'Huillier and Balcou, Phys. Rev. Lett. **70**, 774 (1993)

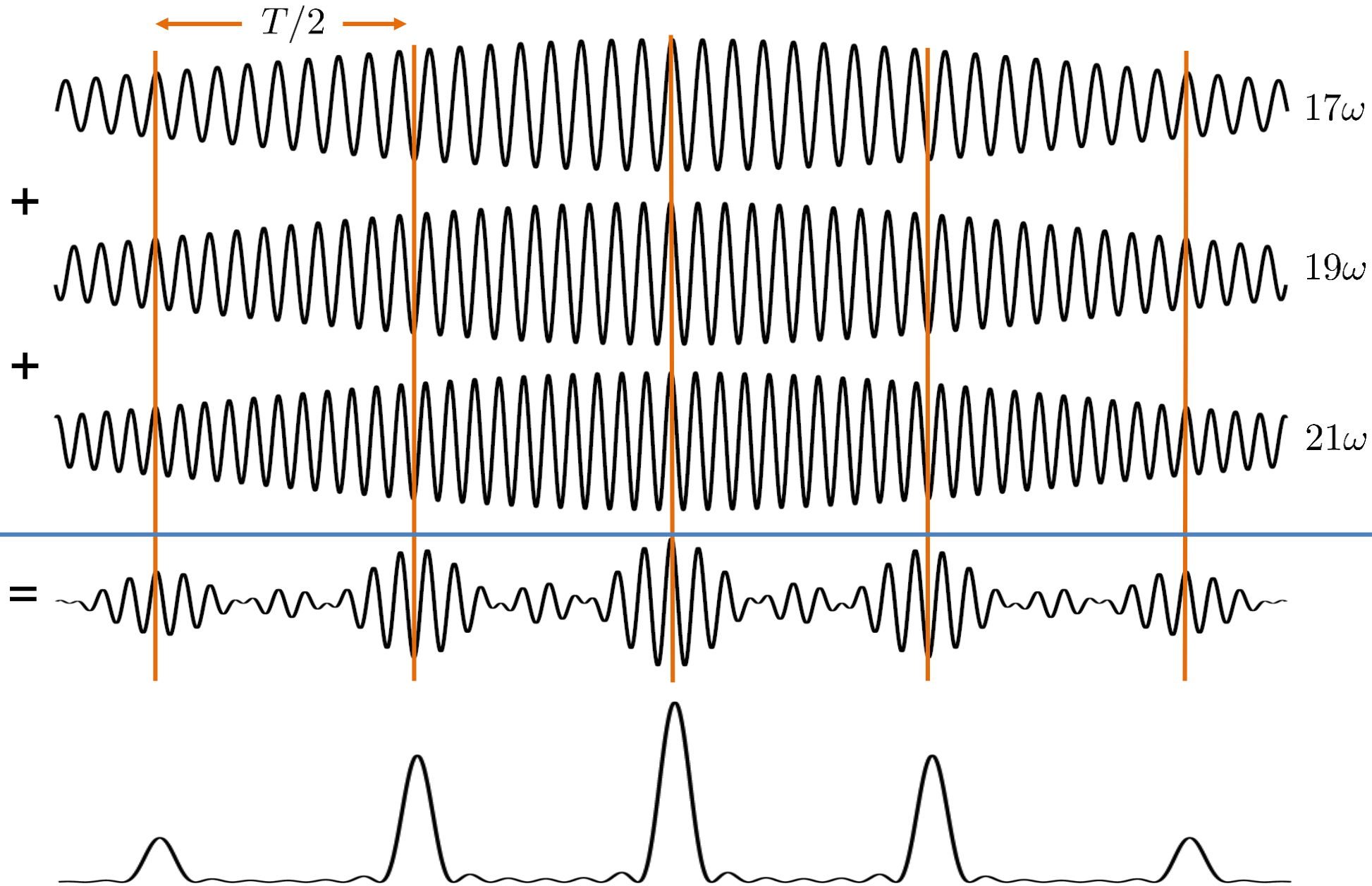
Ti: Sapphire 200 fs 10 Hz
Wahlström et al., Phys. Rev. A **48**, 4709 (1993)



Outline

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

Attosecond pulses?



High frequency

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

Broad bandwidth

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

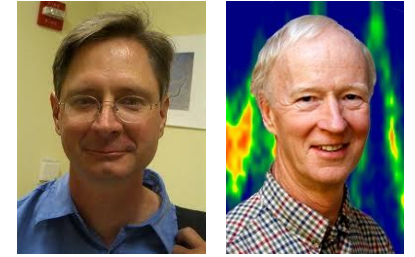
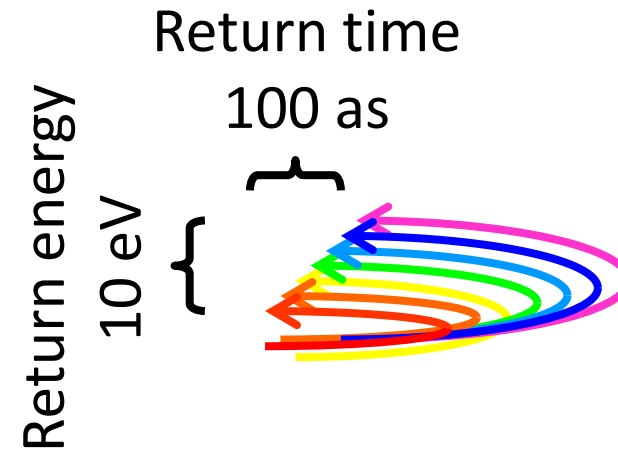
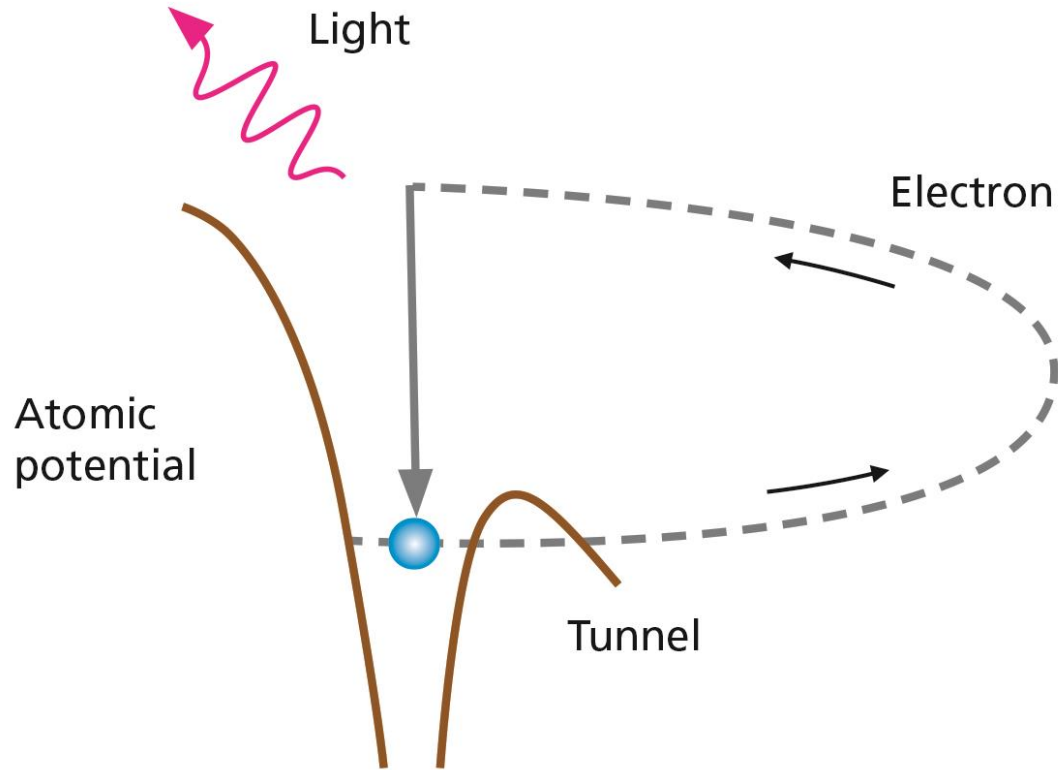
Are the
harmonics
phase-locked?

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)



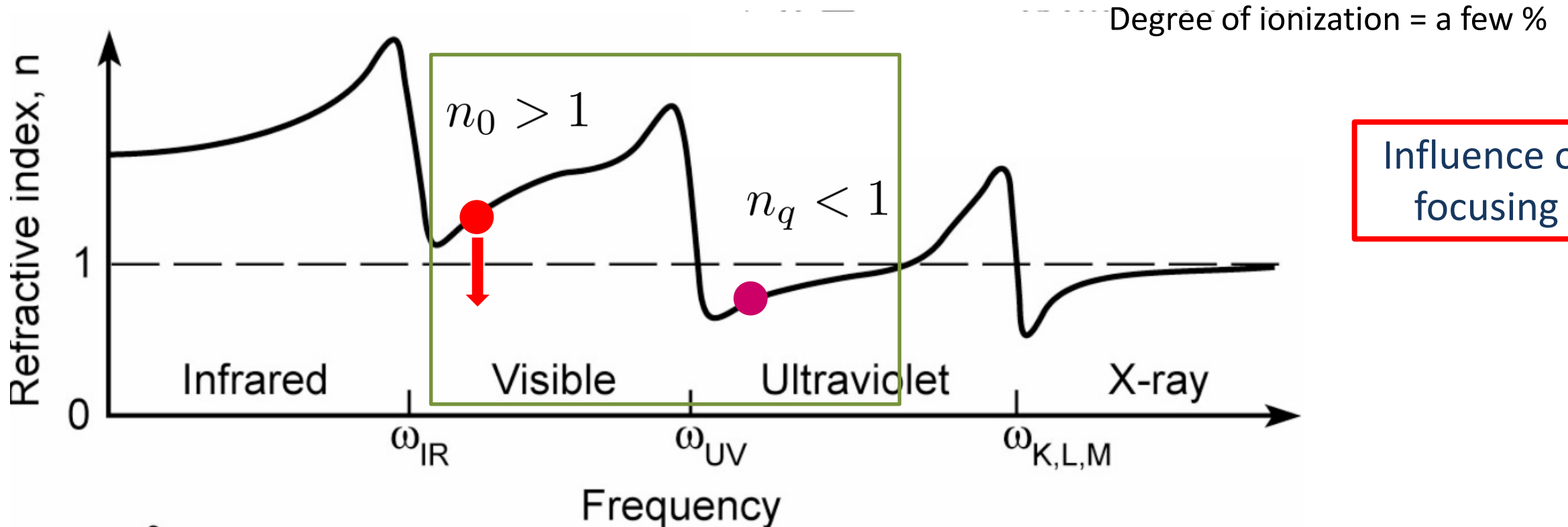
Lewenstein et al. Phys. Rev. A **49**, 2117 (1994)

Progress in understanding: Phase matching

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

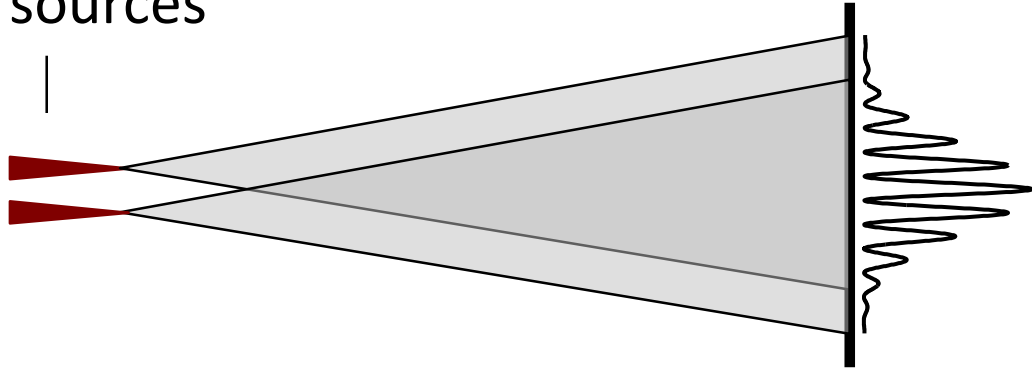
$$\frac{c}{n_0} = \frac{c}{n_q}$$

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

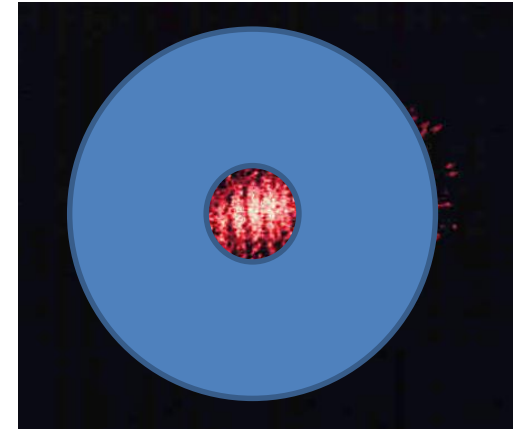


Progress in experiment: Short and long trajectories

Two harmonic sources



Harmonic 15



Short and long trajectories



Mette Gaarde

Ted Hänsch

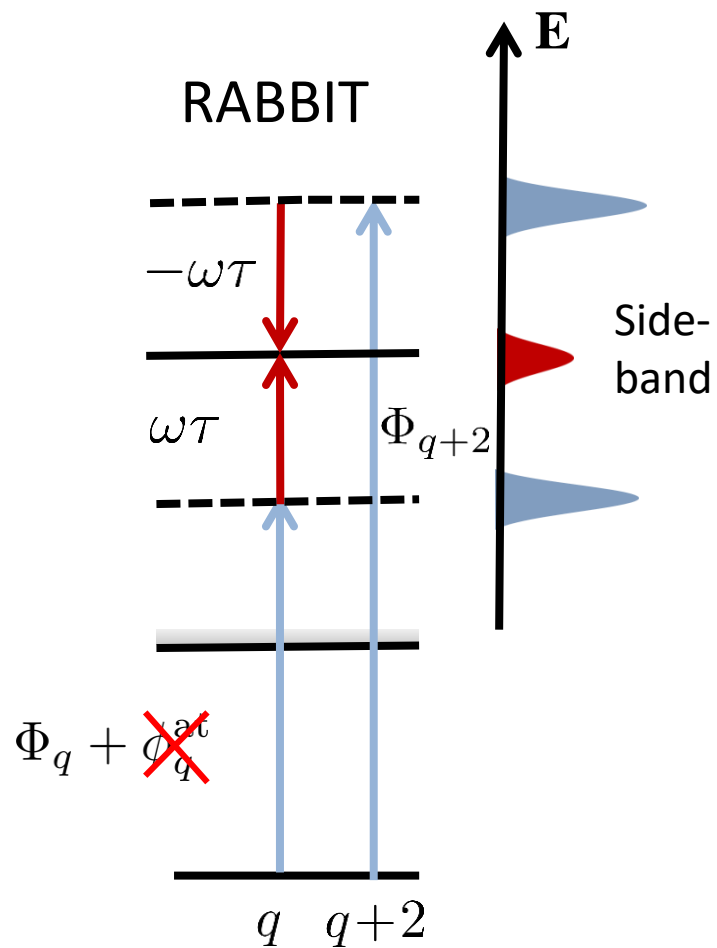
Marco Bellini

ALH

Claire Lyngå,
Claes-Göran
Wahlström

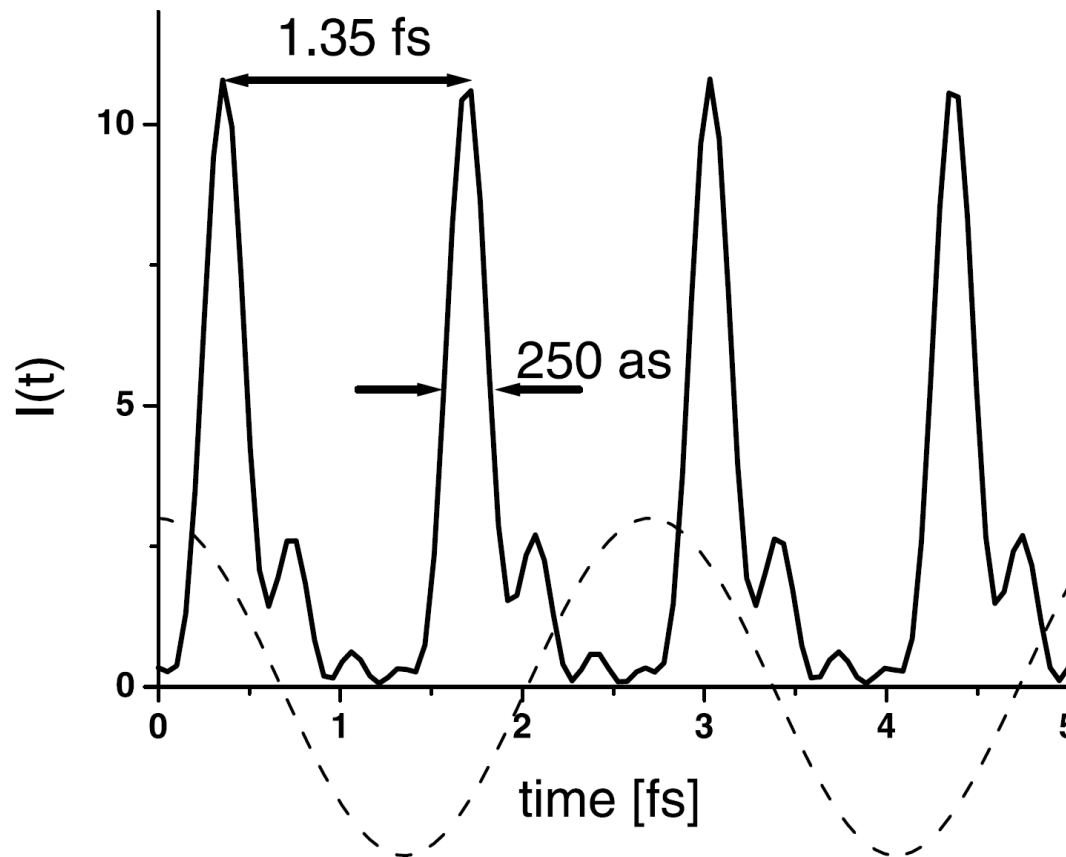
Bellini et al. Phys. Rev. Lett. **81**, 297 (1998)

First measurement of attosecond pulses in a train



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

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



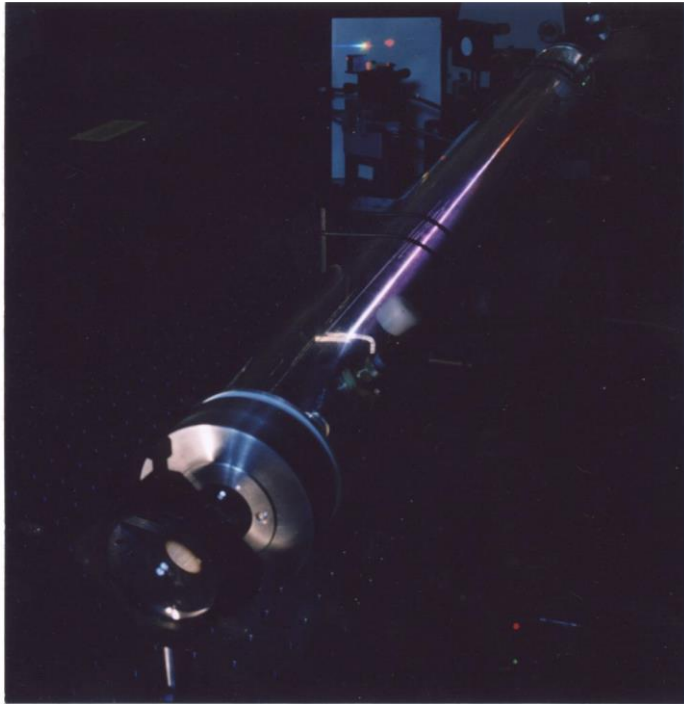
Pierre
Agostini



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

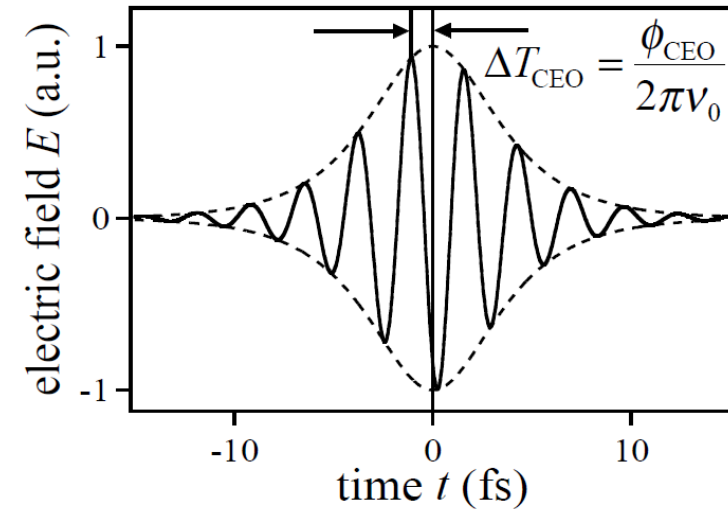
Progress in laser technology (II)

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)

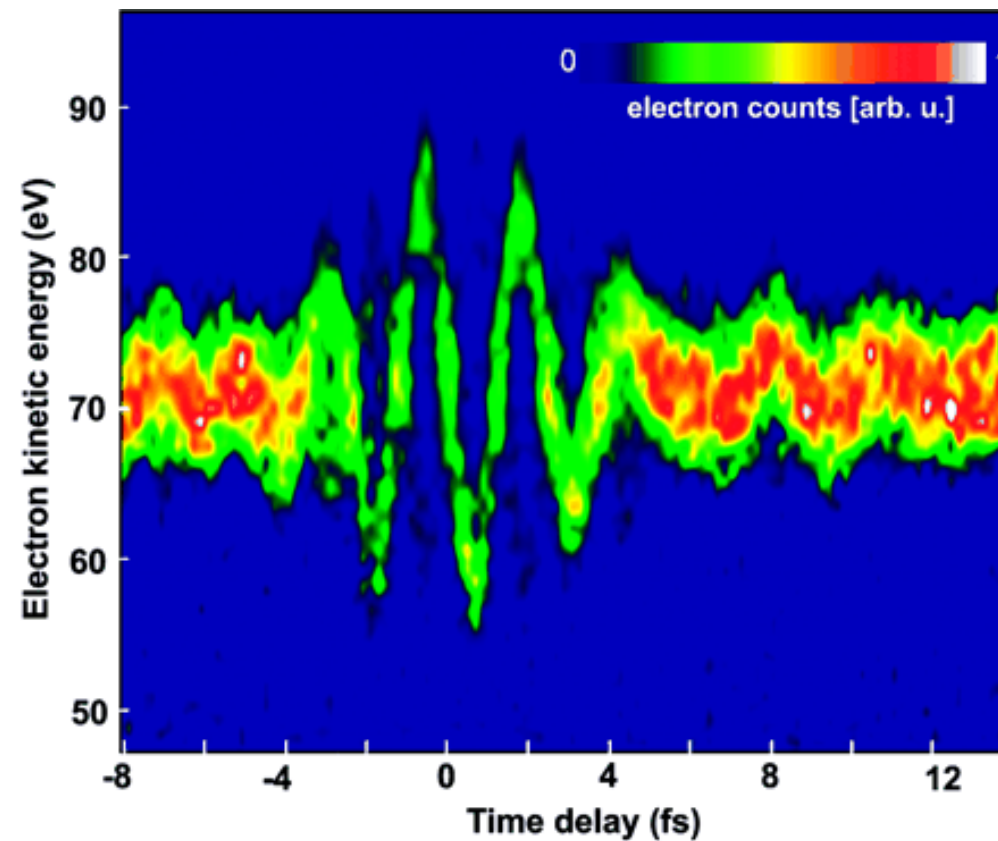
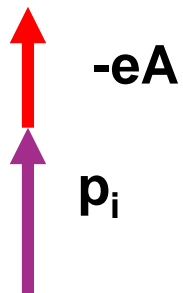
First measurement of isolated attosecond pulses



Ferenc
Krausz



Streaking
camera



$\tau=250$ as

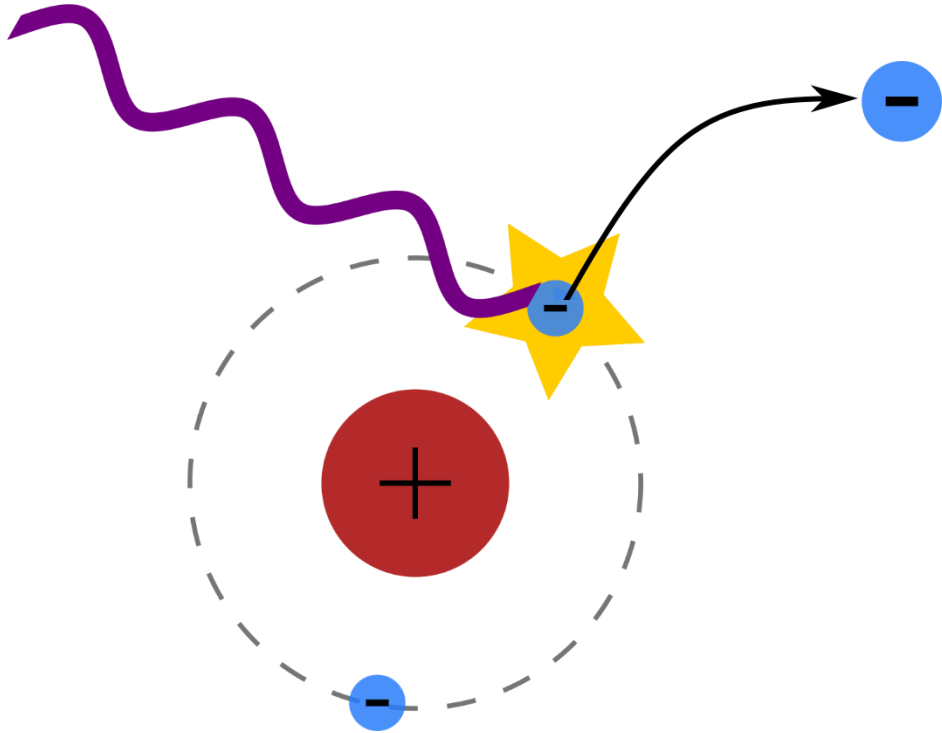
Henstchel et al., Nature **414**, 509 (2001)

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

Outline

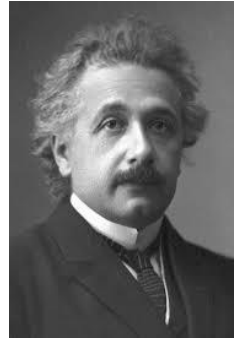
- High-order harmonic generation
- Attosecond light pulses
- **Applications**
 - Atomic Spectroscopy
 - Industrial application

Temporal dynamics of photoionization



Photoelectric effect
Hertz 1887
Einstein 1905

What are the
wave/quantum properties
of the photoelectron?



A. Einstein

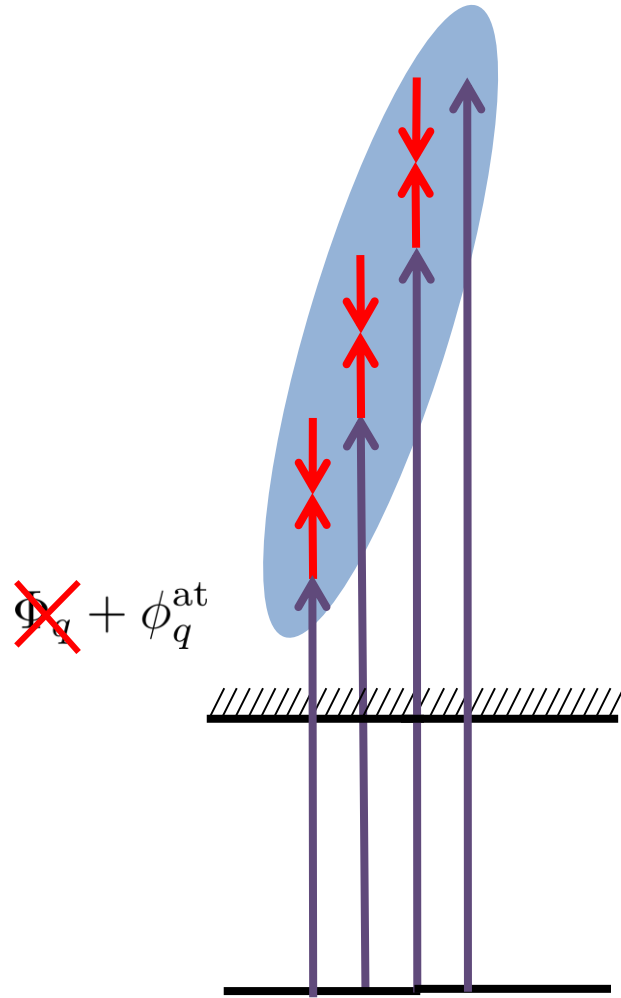
*“for his services to
Theoretical Physics,
and especially for his
discovery of the law of
the photoelectric
effect”*

1921

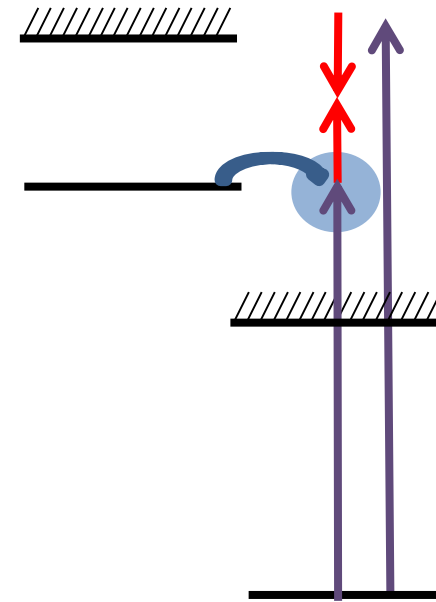


Measuring the phase of a photoelectron

Non-resonant
attosecond
excitation



Discrete
state



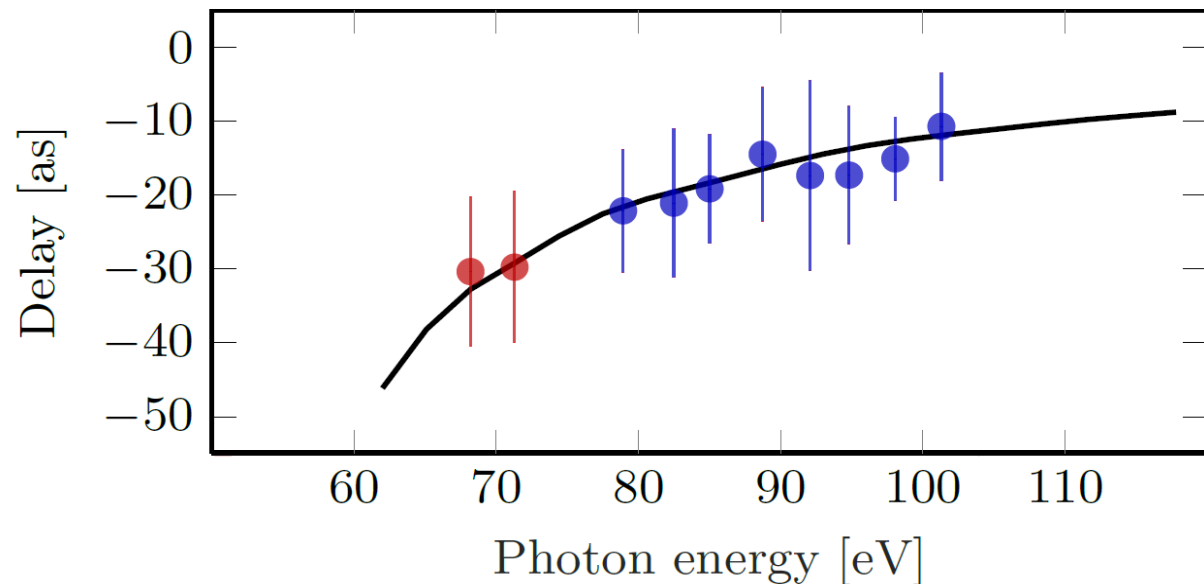
Resonant
harmonic
excitation

Cavaliere et al., Nature **449**, 1029 (2007)
Schultze et al., Science **328**, 1658 (2010)
Klunder 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)

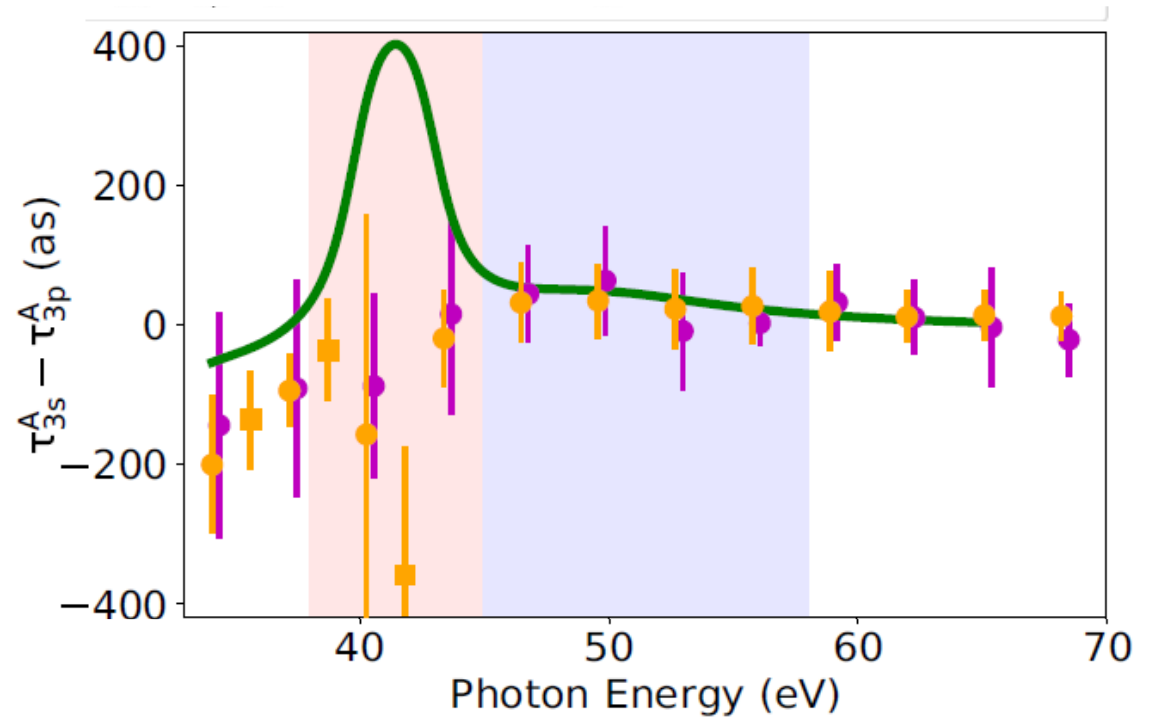
Differences in photoionization time delays

Ne 2s and 2p ionization



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

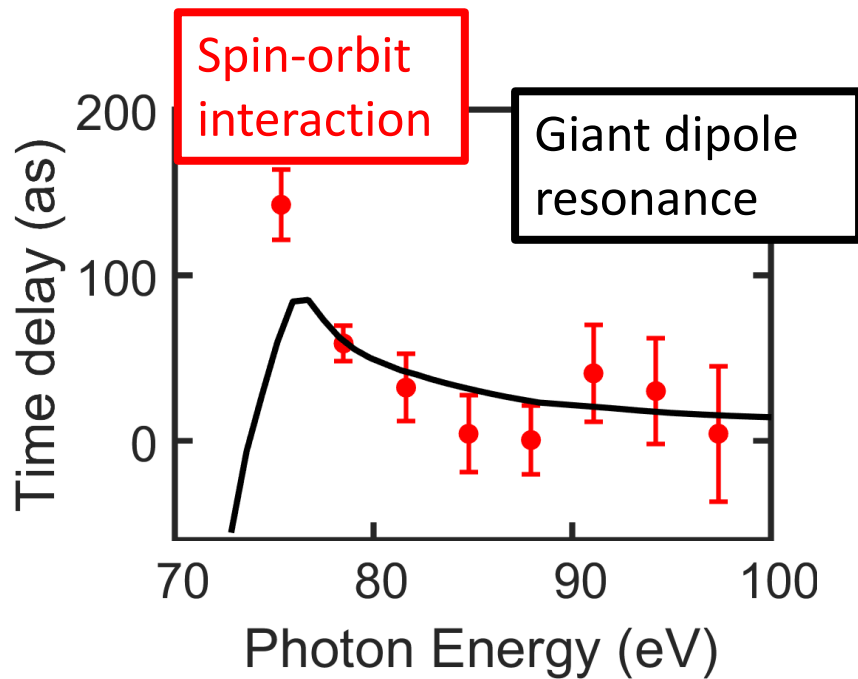
Ar 3s and 3p ionization



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

Photoionization time delays

Xe 4d shell ionization



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

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

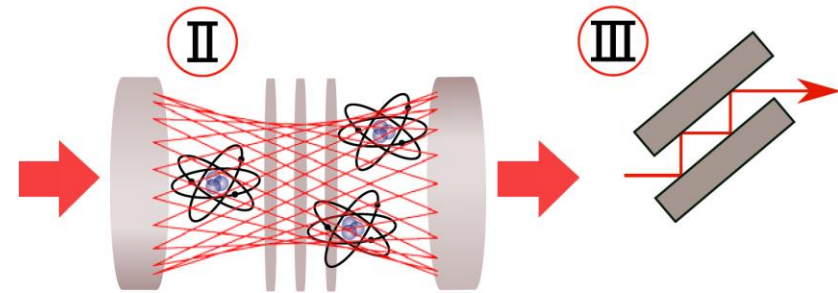
Progress in laser technology (III)

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

High repetition rate > 100 kHz
High average power

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

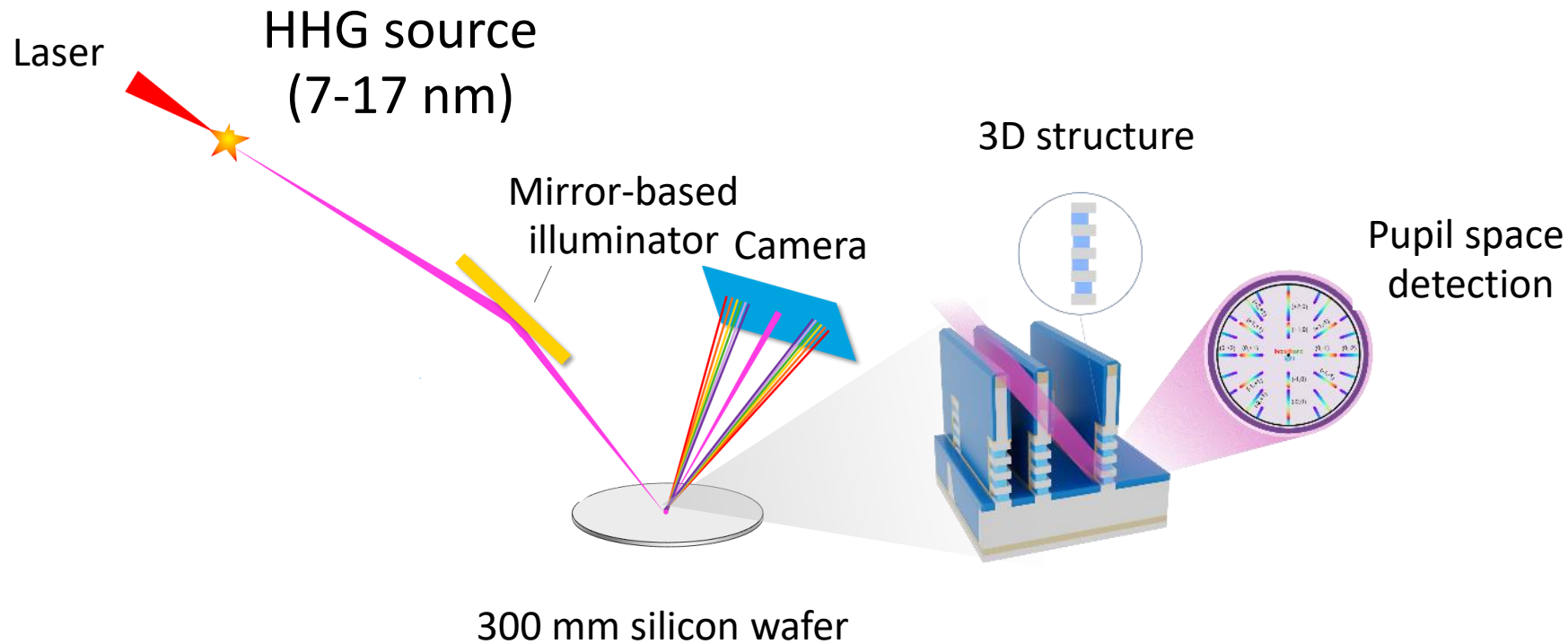
Post-compression in a multipass cell



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!

