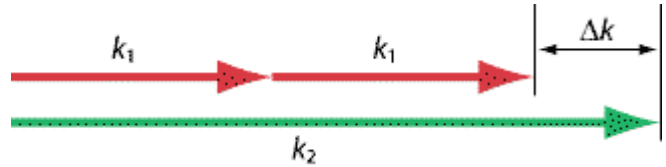
Soft X-Rays and Extreme Ultraviolet Radiation

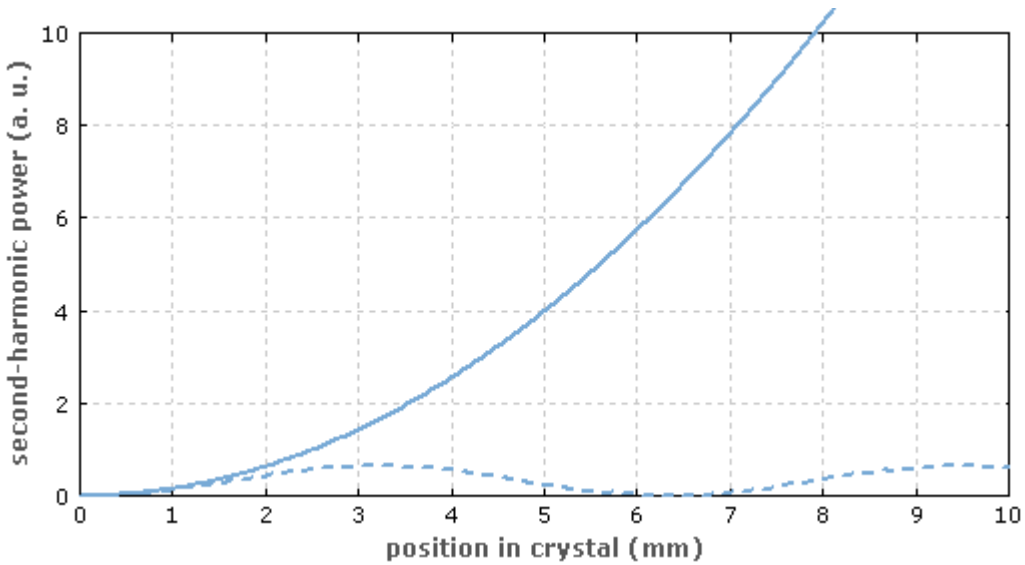
High-Harmonic Generation II

- Phasematching techniques
- Attosecond pulse generation
- Applications
- Specialized optics for HHG sources

Dr. Yanwei Liu, University of California, Berkeley
and Lawrence Berkeley National Laboratory

Phase-matching of nonlinear process

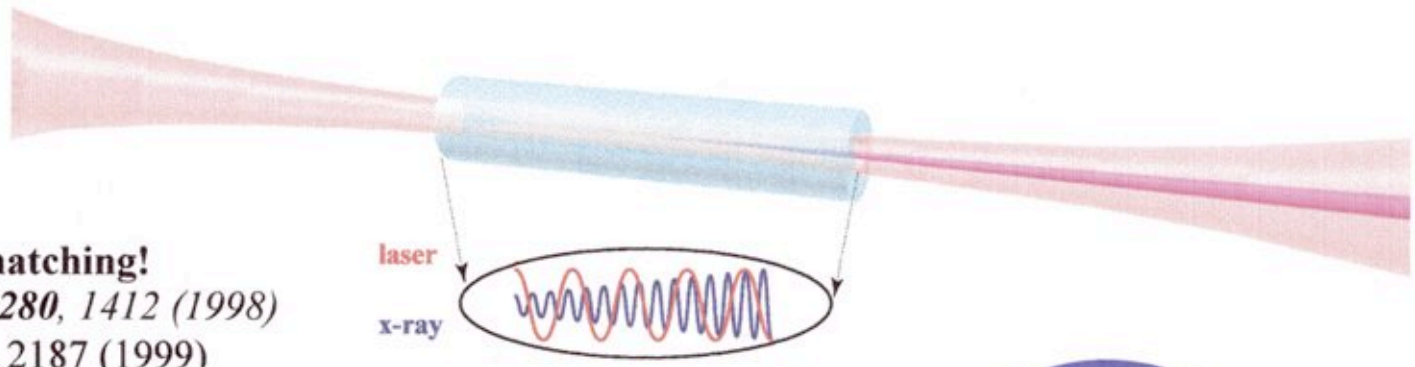




Growth of second-harmonic power in a crystal along the propagation direction, assuming a constant pump intensity. Solid curve: phase-matched case, with the power growing in proportion to the square of the propagation distance. Dashed curve: non phase-matched case, with the second-harmonic power oscillating between zero and a small value.

Adopted from Encyclopedia of Laser Physics and Technology

HHG in Hollow Fibers



Phase matching!
Science ***280****, 1412 (1998)*
PRL **83**, 2187 (1999)

$$k = \frac{2\pi}{\lambda}\left(1 + P\delta(\lambda) - \frac{1}{2}\left[\frac{u\lambda}{2\pi a}\right]^2 - \frac{1}{2}\frac{N_e r_e \lambda^2}{\pi}\right)$$

vacuum **gas** **waveguide** **ionization**

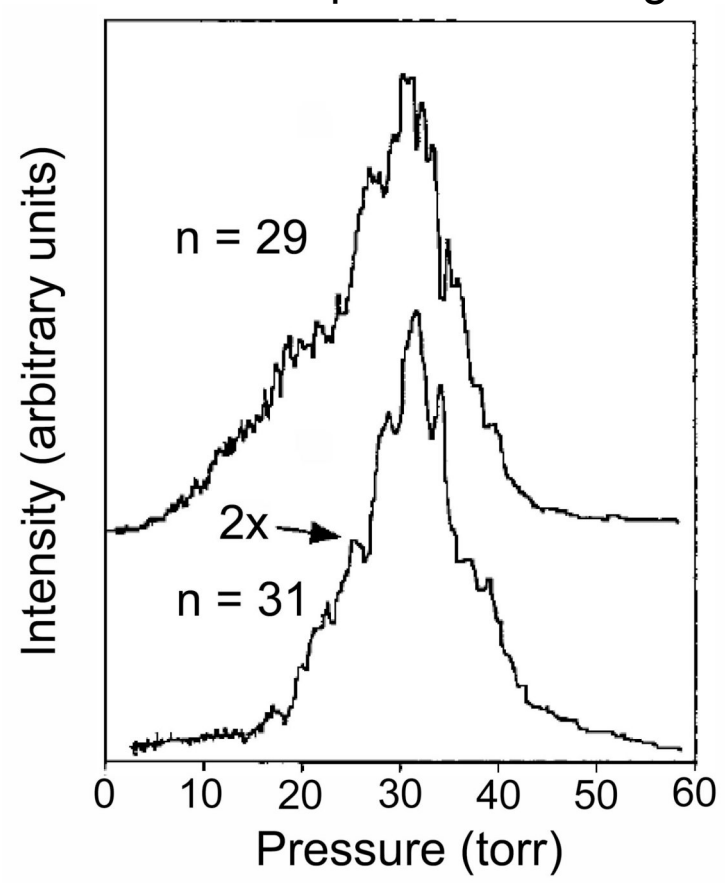
Hollow fibers allow phase matching of low-order harmonics

Higher harmonics are generated at higher laser intensities and higher levels of ionization. Since laser and EUV light cannot stay in phase for high levels of ionization, high harmonics cannot be generated efficiently.

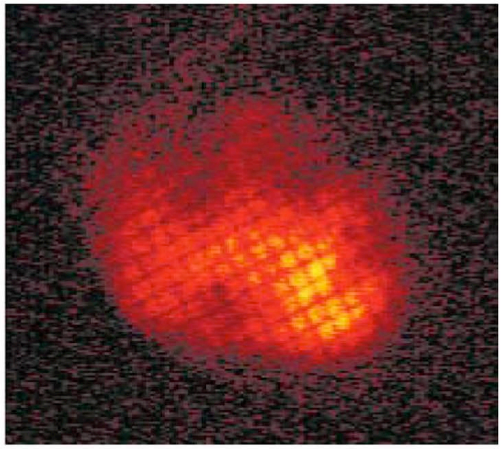
HHG in Hollow Fibers

Phase-Matched Generation of Coherent EUV Radiation
Andy Rundquist, *et al.*
Science **280**, 1412 (1998)

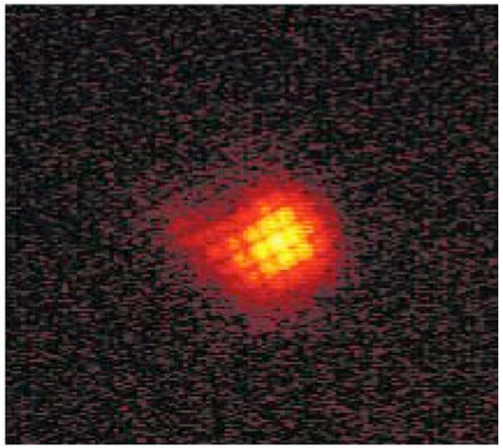
Pressure phase matching



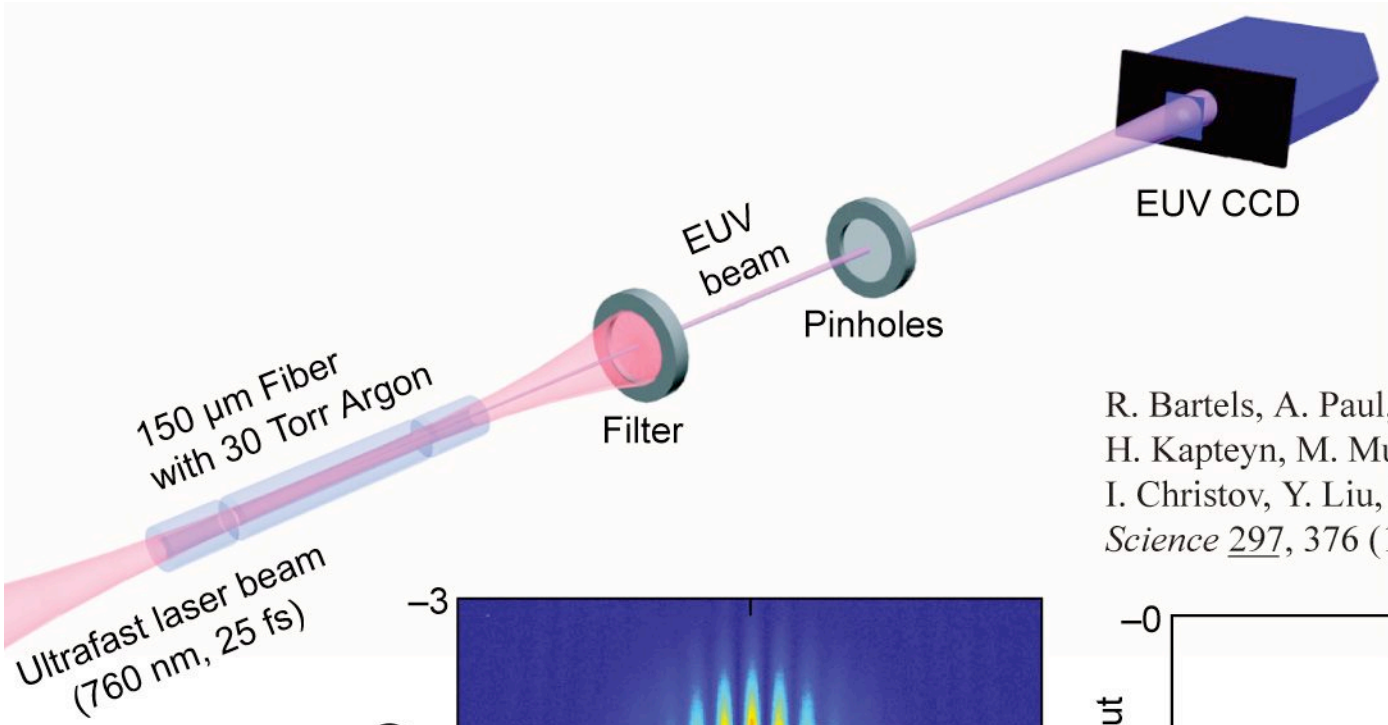
No phase-matching,
n = 23-31, EUV output beam



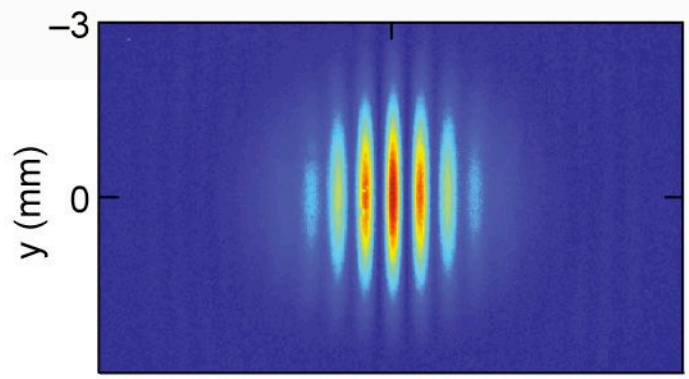
With phase-matching,
n = 23-31, EUV output beam

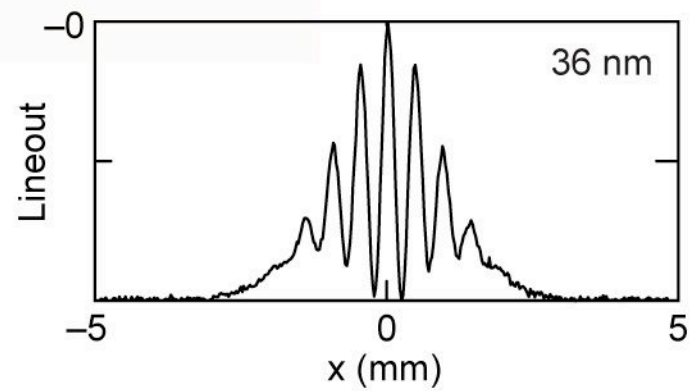


Fully coherent EUV from HHG in hollow fiber



R. Bartels, A. Paul, H. Green, H. Kapteyn, M. Murnane, S. Backus, I. Christov, Y. Liu, D. Attwood, C. Jacobsen, *Science* 297, 376 (19 July 2002).



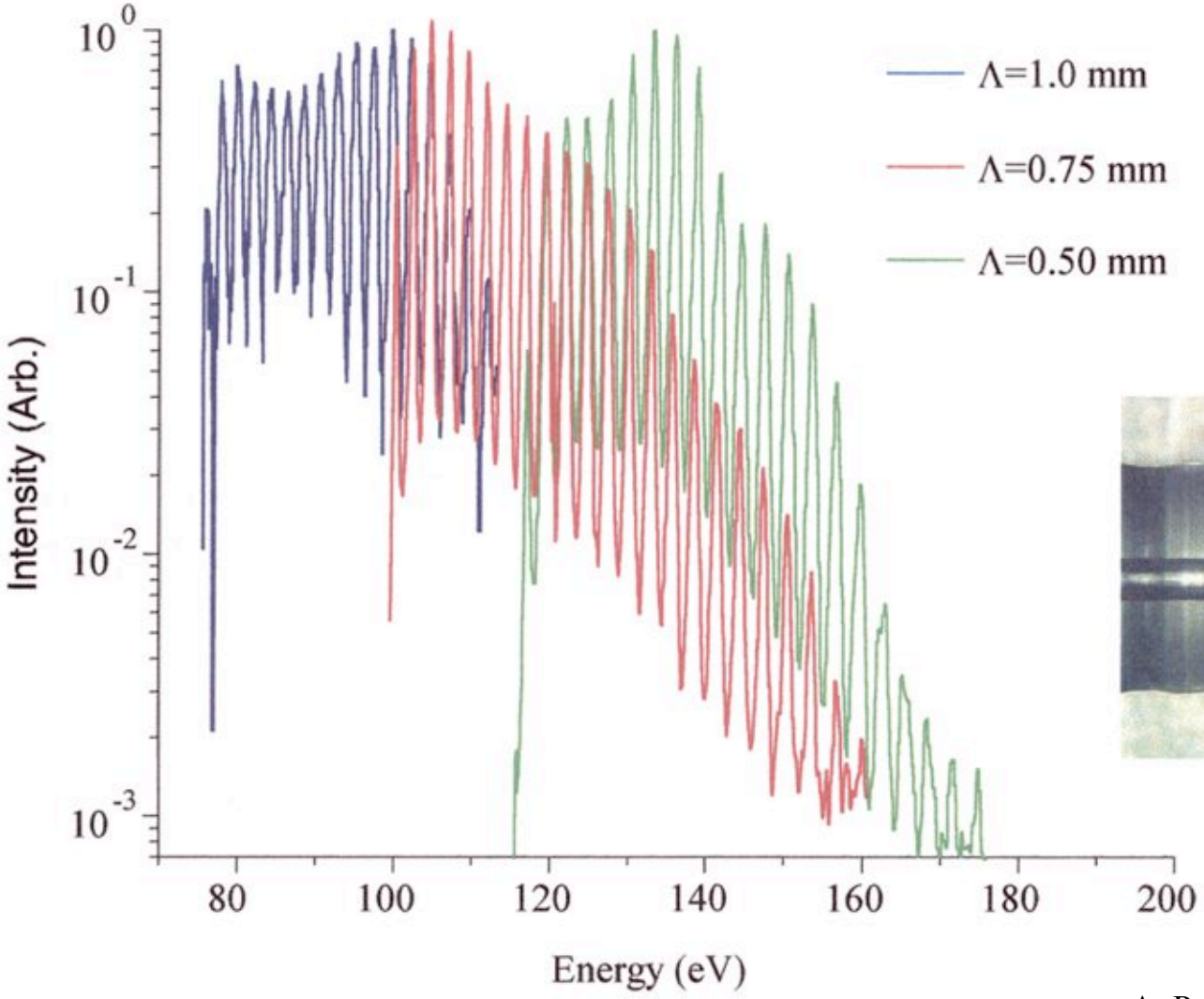


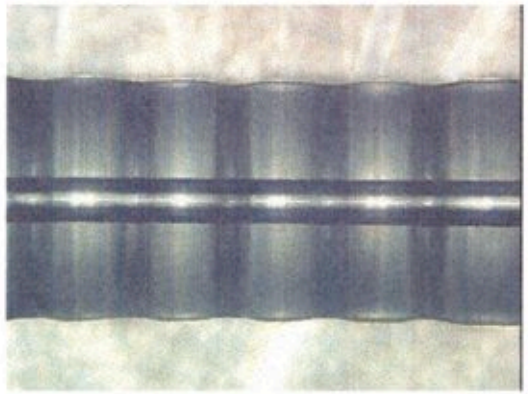
$P \simeq 10\ \mu W \rightarrow 2 \times 10^{12}$ ph/sec @ 36 nm (n = 21; 34 eV)

Courtesy of Professors Margaret Murnane and Henry Kapteyn, Univ. Colorado.

HHG_cohFempto_Oct05.ai

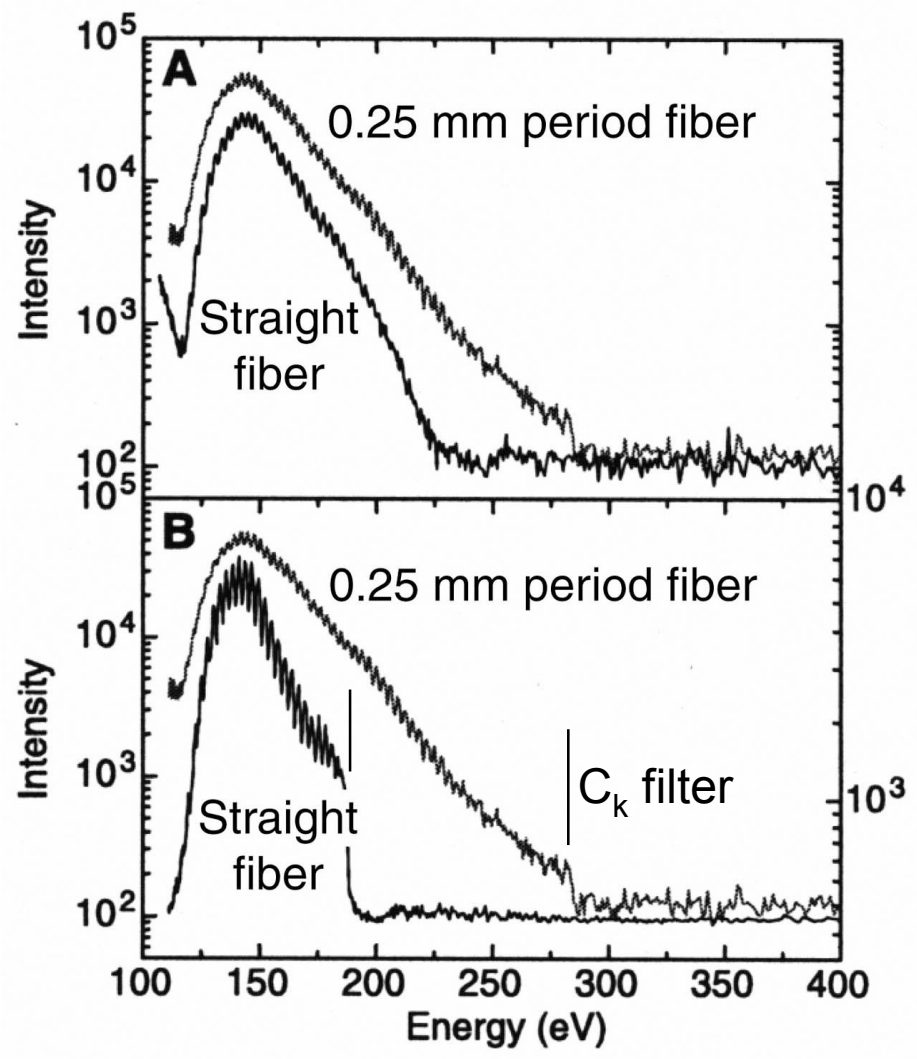
Short Modulation Period Capillaries Extend Phase Matching from 85 eV (Unmodulated Fibers) to 160 eV

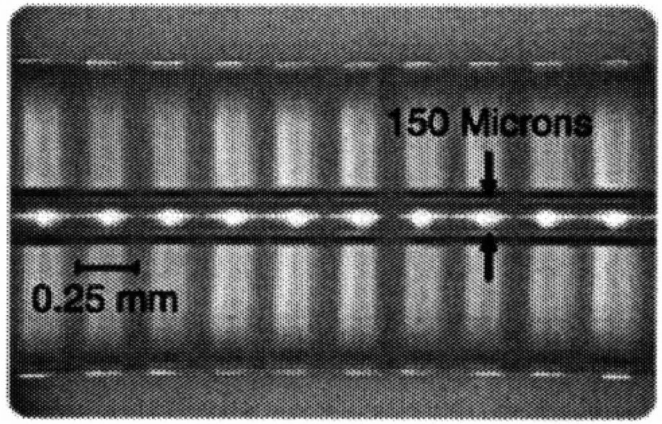




A. Paul et al., *Nature* (2 Jan 2003)
E. Gibson et al., *Science* (3 Oct 2003)

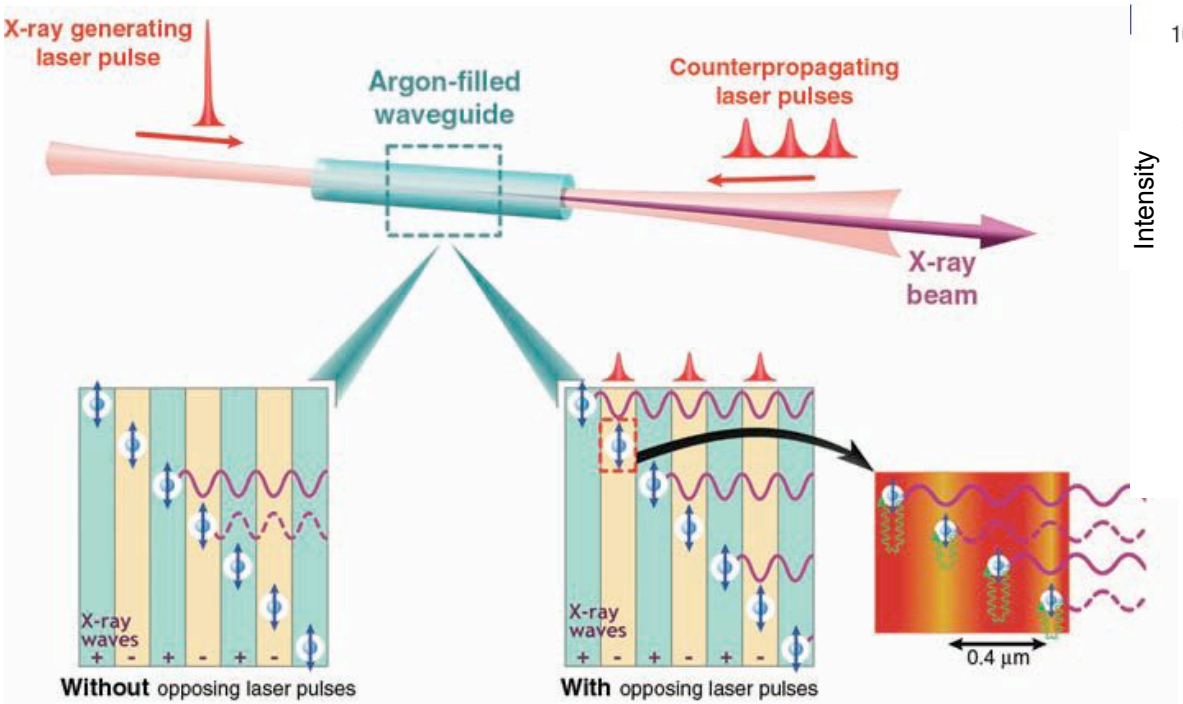
Coherent Soft X-Ray HHG with Quasi-Phase Matching

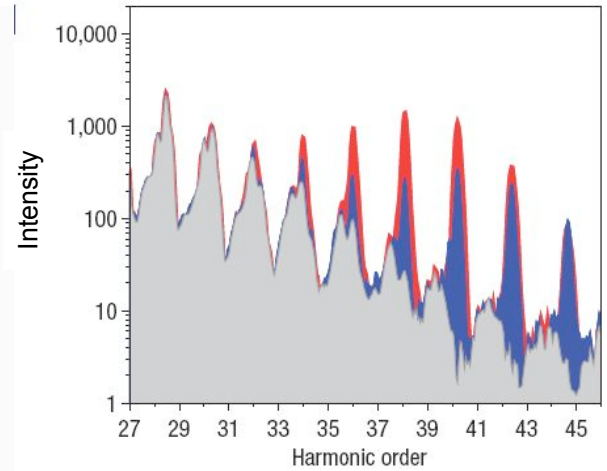




Courtesy of E. Gibson, A. Paul, H Kapteyn, M. Murnane, and colleagues
Science **302**, 96 (3 Oct 2003)

Quasi phase matching using counter-propagating pulses

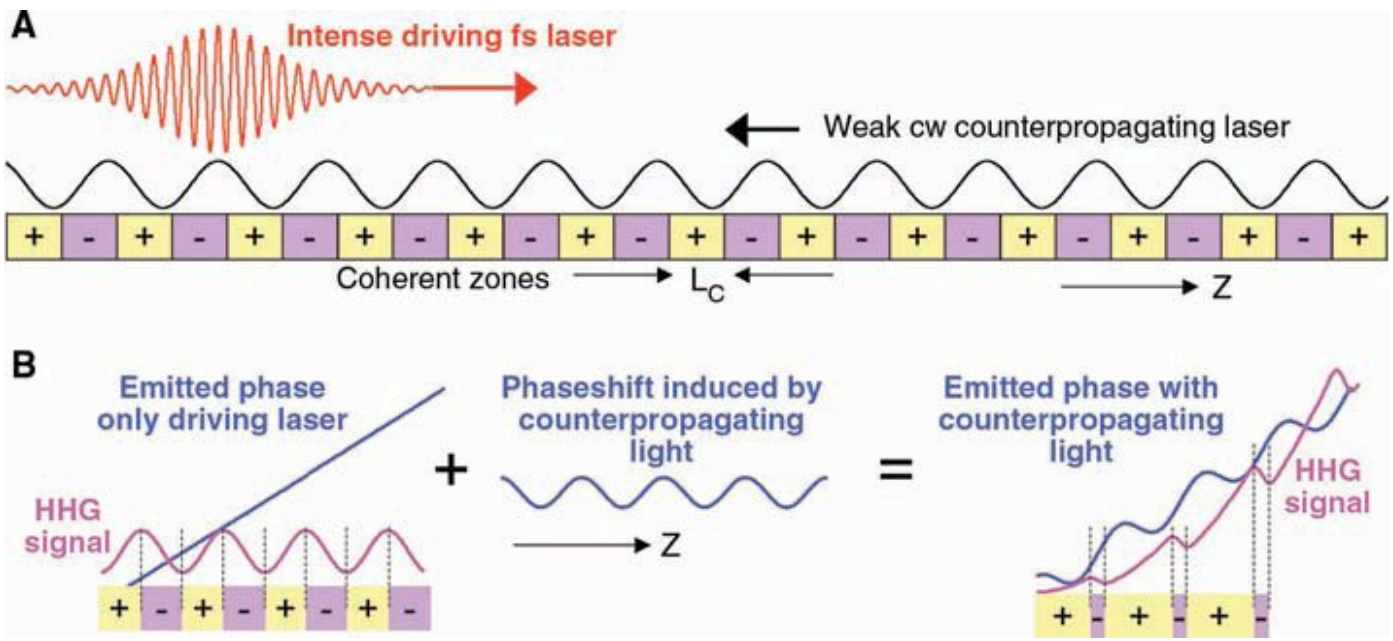




Picosecond pulses, HHG coherence length ~ 1mm.

X. Zhang, A. L. Lytle, H. C. Kapteyn, M. M. Murnane, O. Cohen, Nat. Phys. 3, 270 (2007).

Quasi phasematching using CW counter-propagating laser

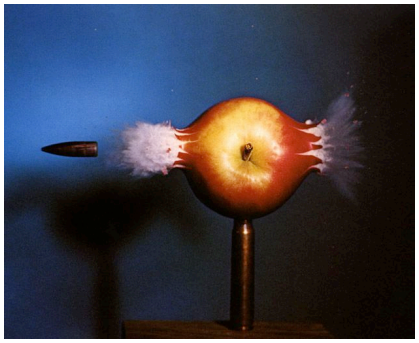


O. Cohen et al., Phys. Rev. Lett. 99, 53902 (2007).

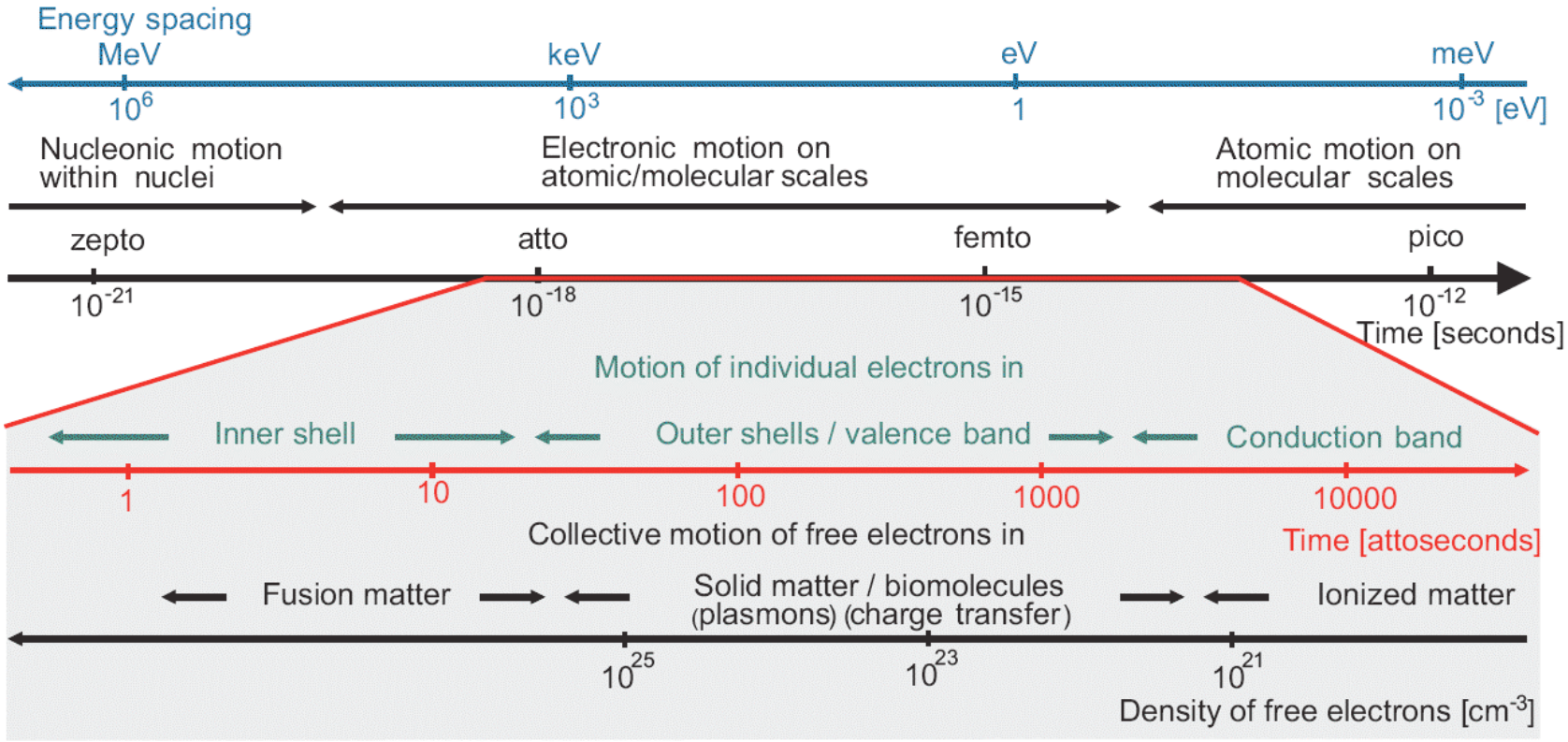
The case for short pulses

Attosecond physics

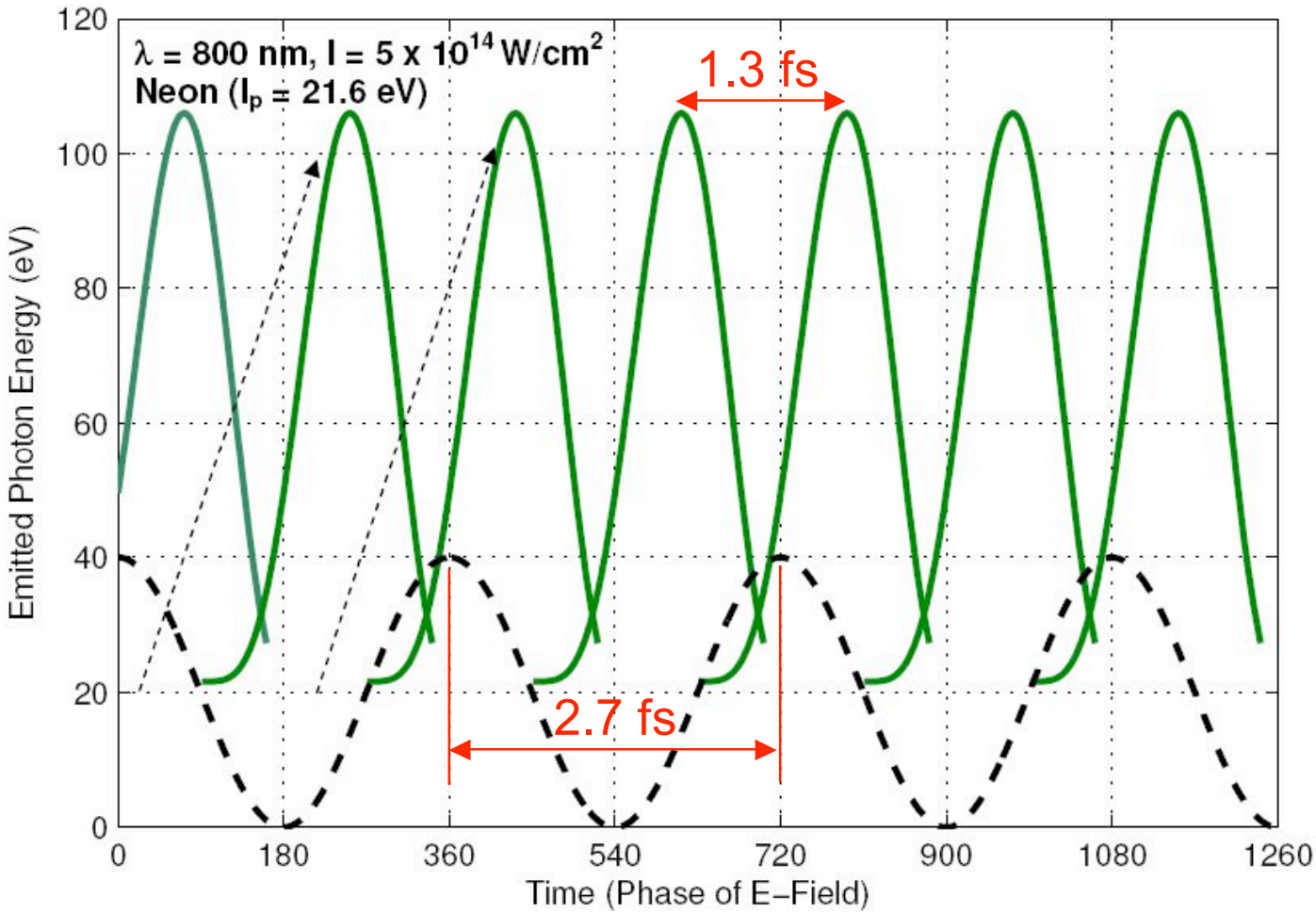
F. Krausz, M. Ivanov
Review of Modern Physics 81, 163 (2009)



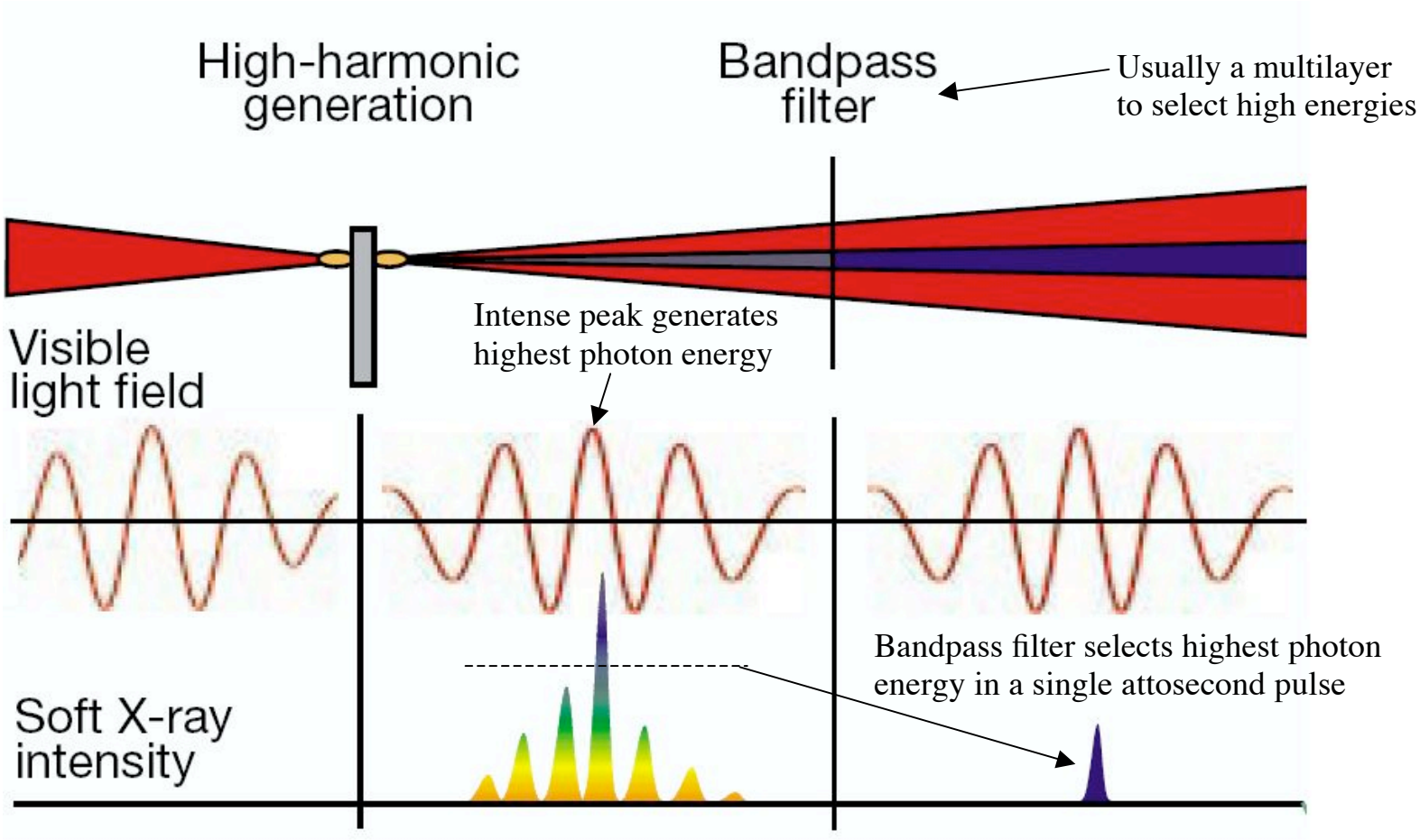
By Harold "Doc" Edgerton, MIT



‘Long’ (many cycles) pump laser generates attosecond pulse train

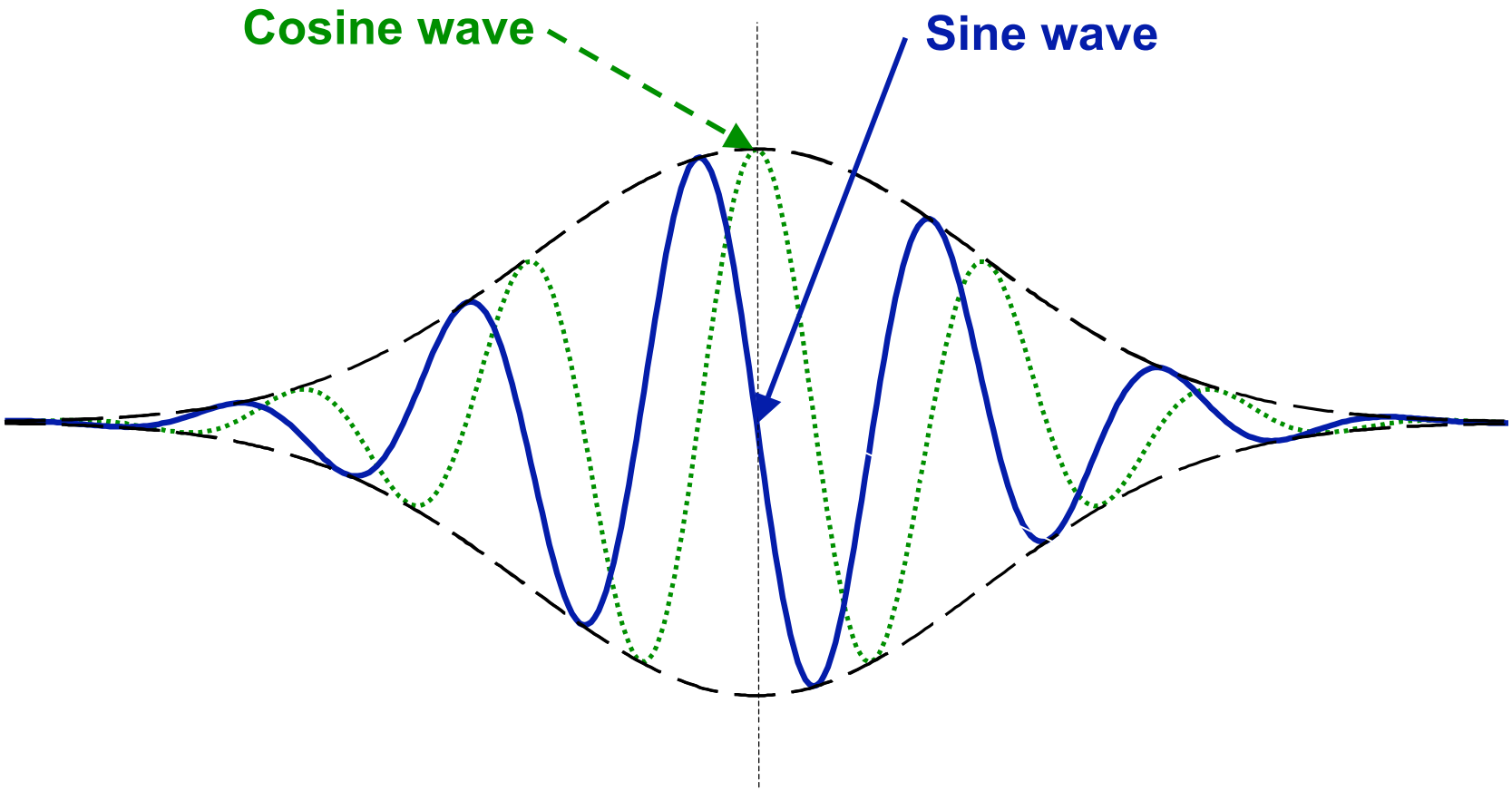


Isolated attosecond pulse generation using few-cycle pump

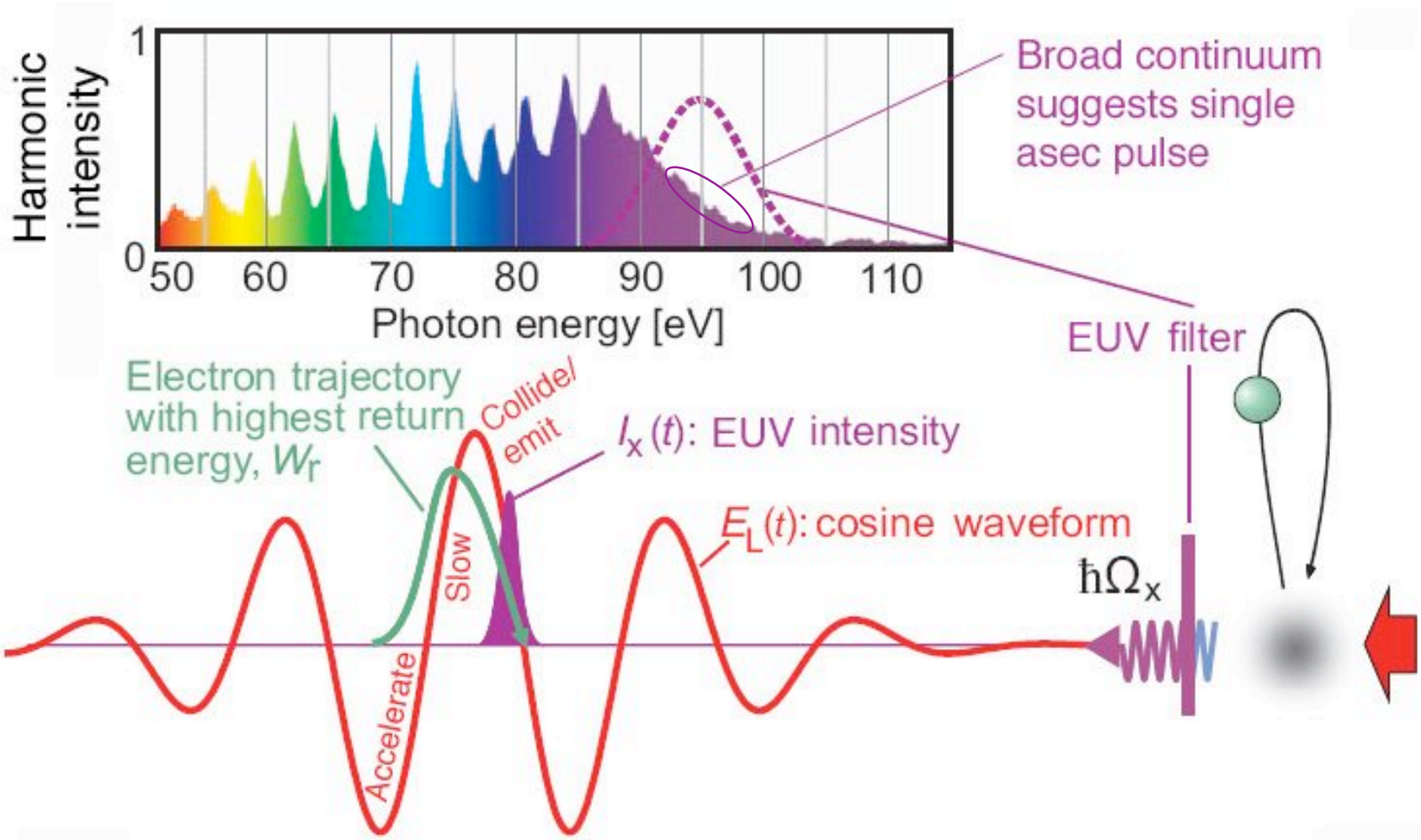


M. Hentschel et al., Nature 414, 509 (2001).

Carrier-Envelope Phase (CEP) of ultrafast pulse

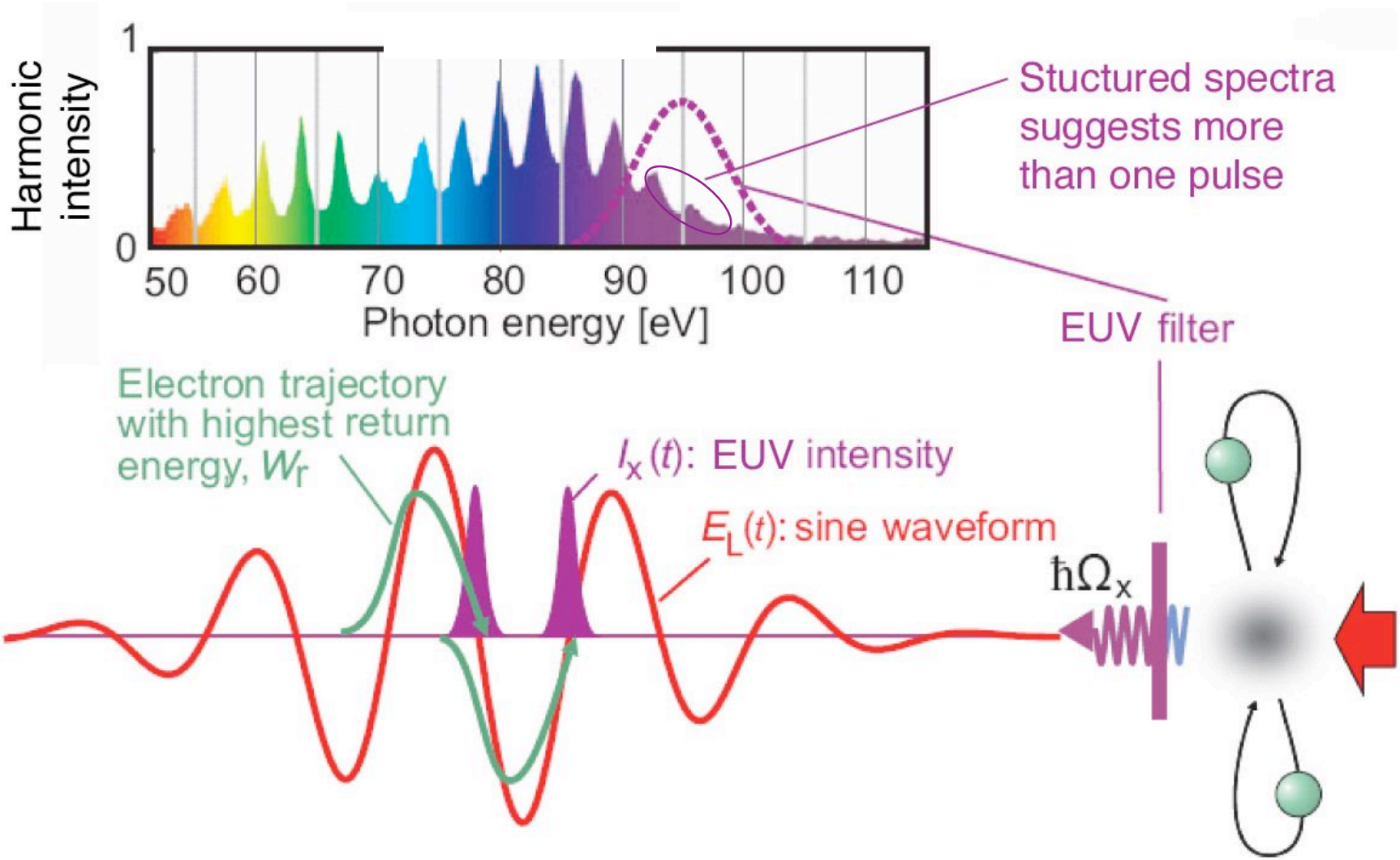


Cosine waveform generates single attosecond pulse



A. Baltuska et al., *Nature* **421**, 611 (2003);
F. Krausz, M. Ivanov, Rev Modern Phy **81**, 163 (2009)

While sine waveform generates two attosecond pulses



A. Baltuska et al., *Nature* **421**, 611 (2003);
F. Krausz, M. Ivanov, Rev Modern Phy **81**, 163 (2009)

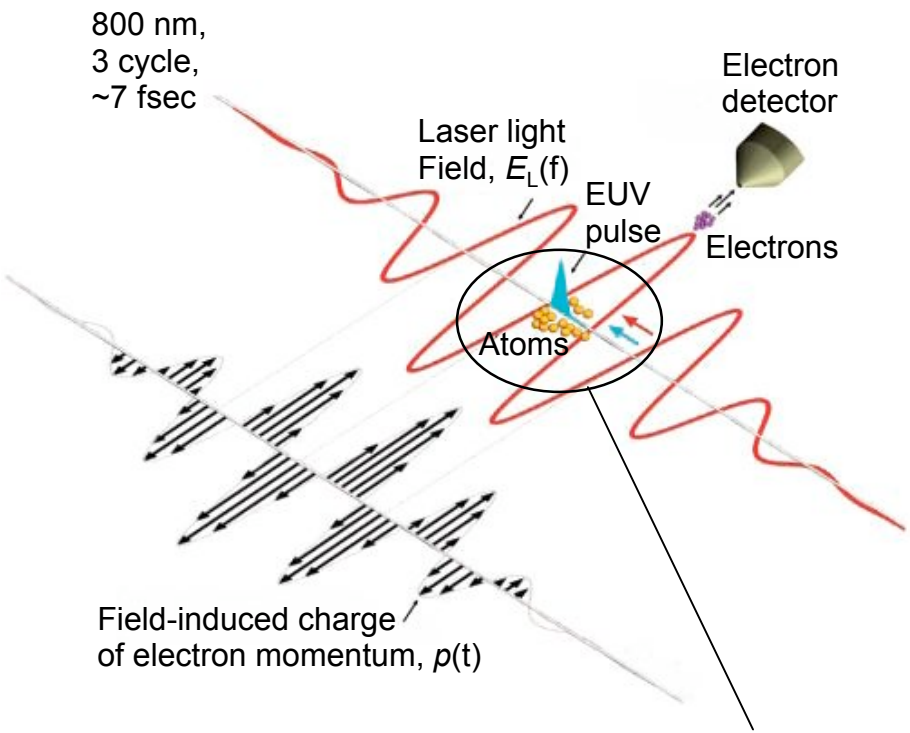
Applications of HHG sources

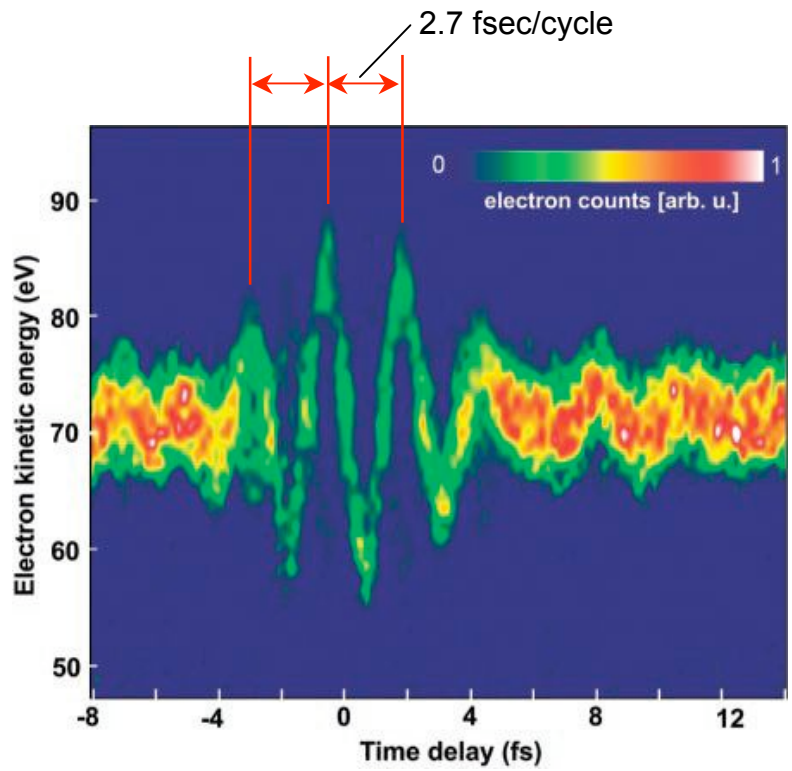
Merits of HHG source:

- Ultrafast EUV/SXR pulses
  - Temporal resolution in fs/as scale, never reached before
  - Molecular dynamics excited by EUV photons
  - Inner-shell probe (high photon energy)
  - Well-controlled pump-probe experiments (automatically synchronized with IR pump)
- Coherent radiation at short wavelengths (nm and fsec)
  - Coherent Diffractive Imaging (CDI, or ‘lenseless’ imaging)
  - Holographic Imaging
  - Zoneplate Imaging

Direct measurement of 750 nm light wave pulse duration

A 250-as EUV pulse is used to map the electric field of 750-nm laser light wave

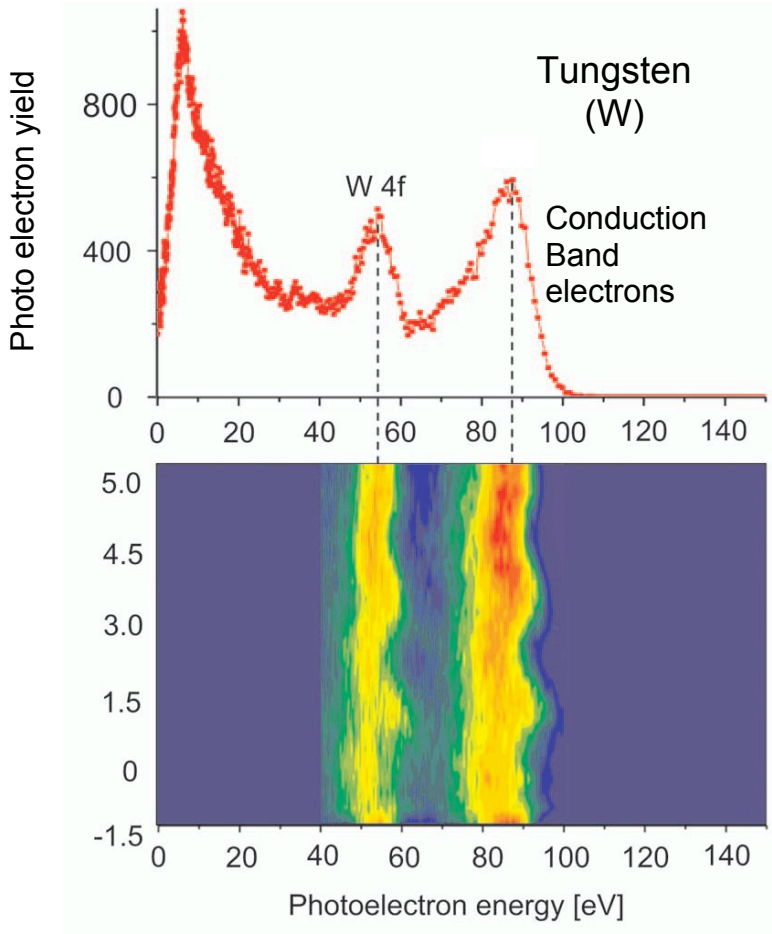




Overlap of 7 fsec IR pulse and 250 asec EUV pulse.
EUV frees electrons, IR electric field accelerates these electrons.
These electrons arrive in waves via time-of-flight tube to detector.

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

Unprecedented time resolution



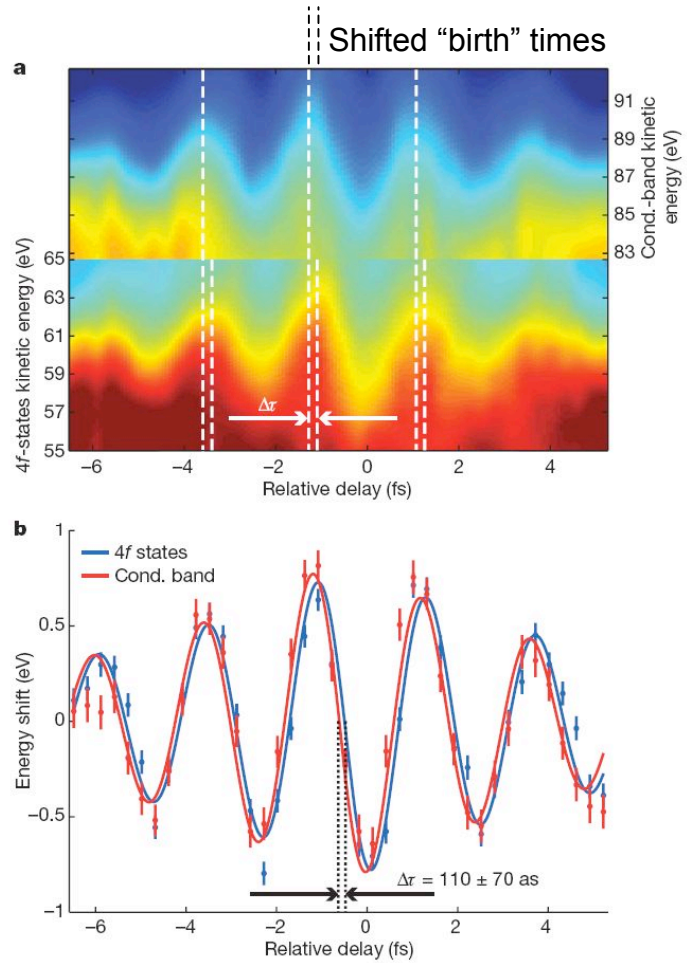
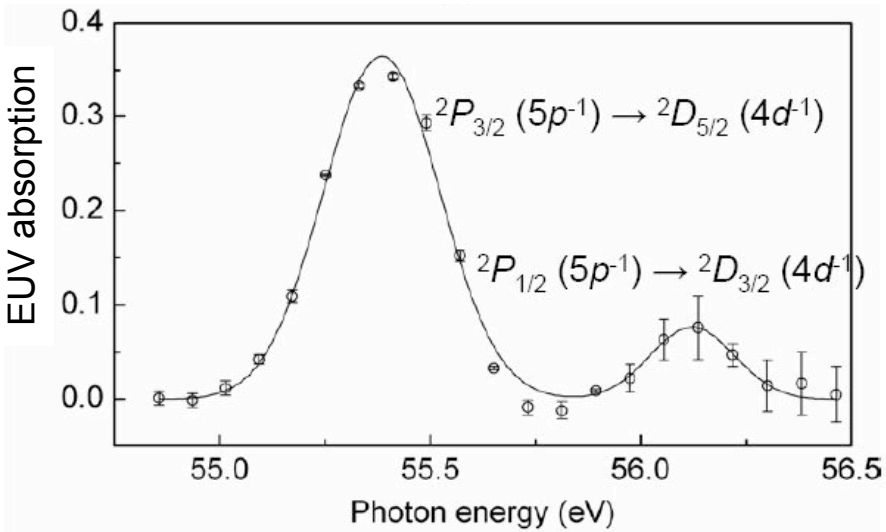


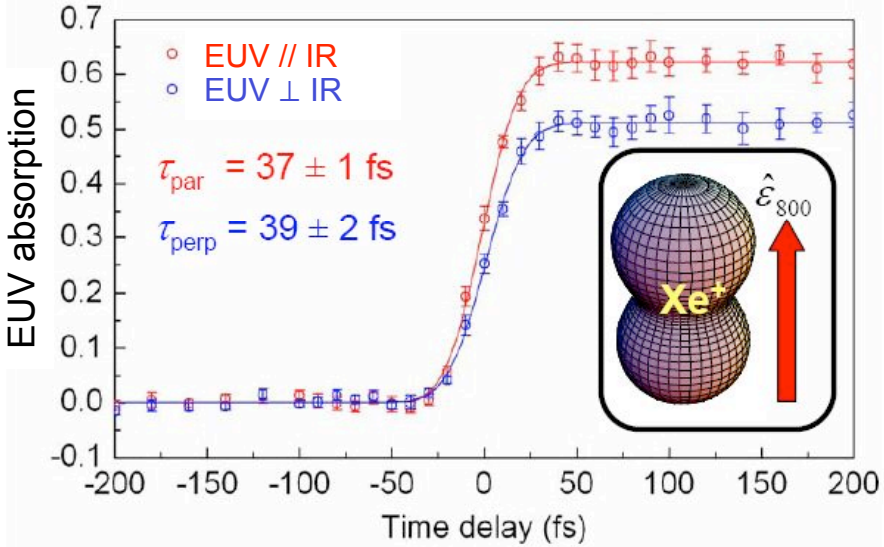
Figure 3 | Evidence of delayed photoemission. **a**, The 4*f* and conduction-

Attosecond spectroscopy in condensed matter
A. L. Cavalieri, et al., Nature 449, 1029 (2007)

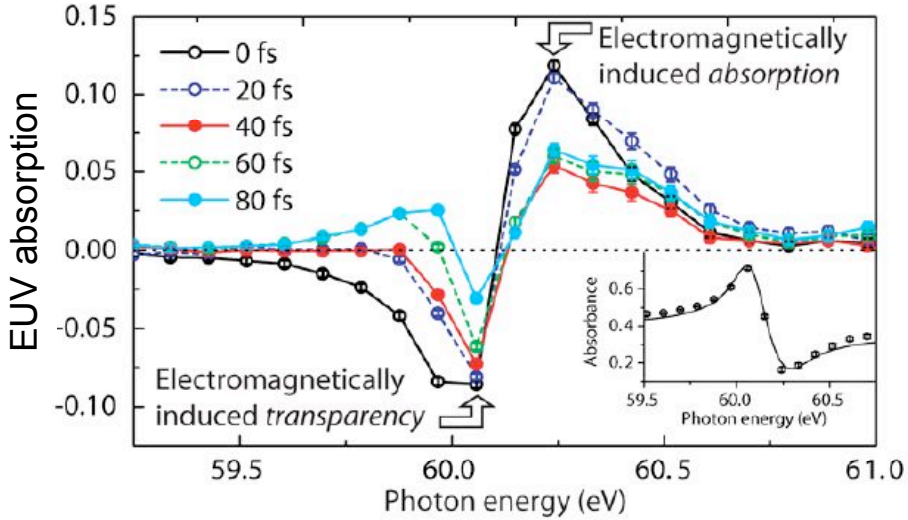
Fs XUV transient absorption spectroscopy

Orbital alignment and nonadiabatic behavior in the strong-field ionization of Xe

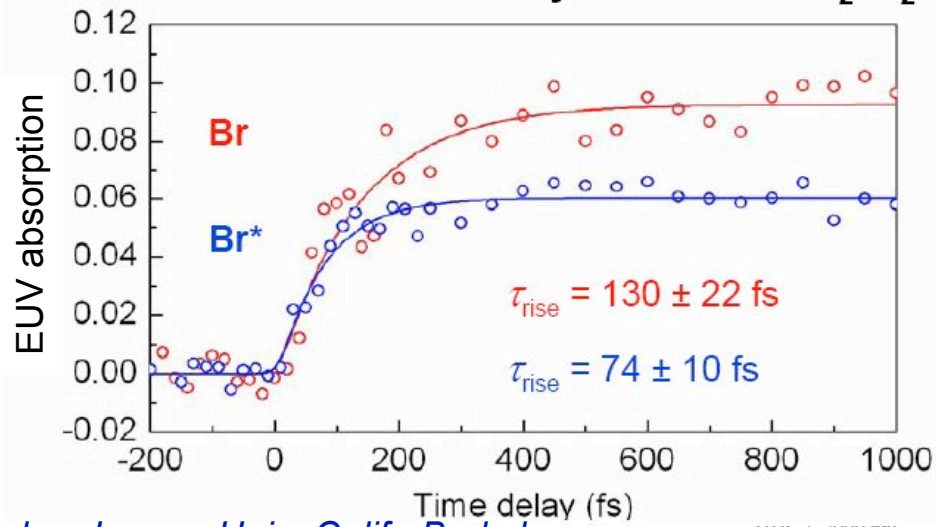




Electromagnetically Induced Transparency (EIT) in the XUV via coherent coupling of He double excitation states



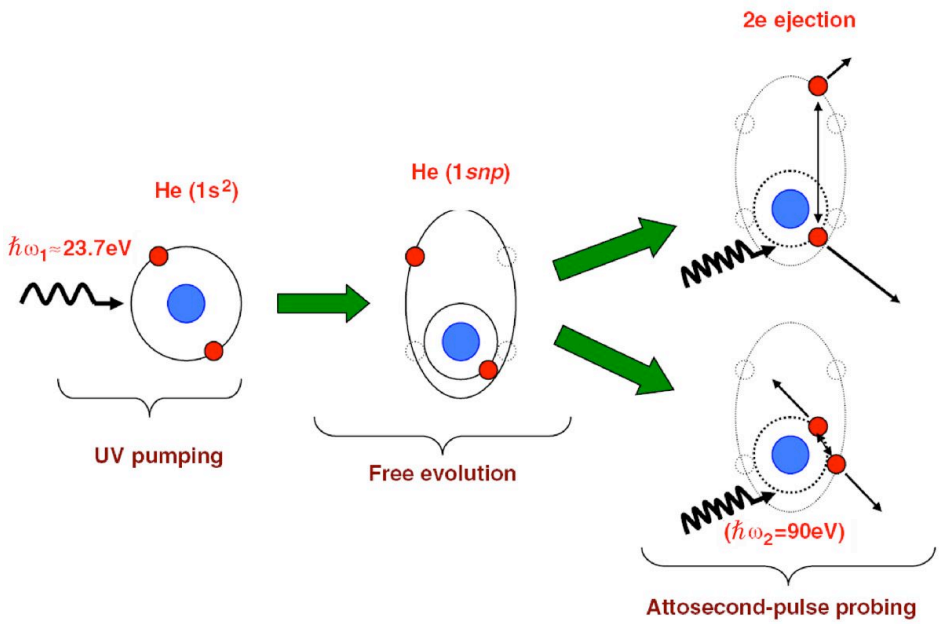
Direct observation of strong-field dissociative ionization dynamics of CH_2Br_2

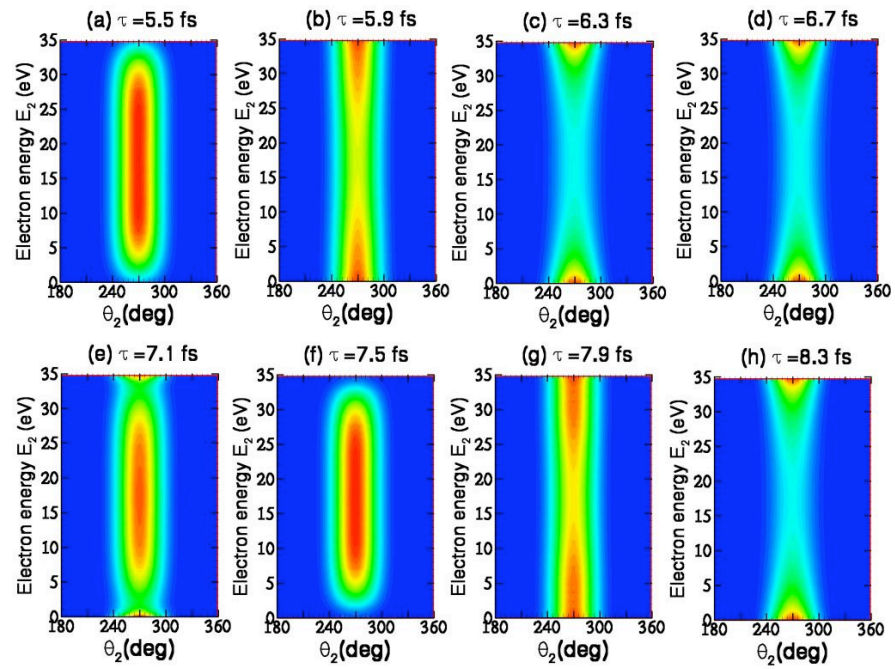


Courtesy of Zhi-Heng Loh and Stephen Leone, Univ. Calif., Berkeley

EUV pump, EUV probe

Electronic motion inside an atom (computational)

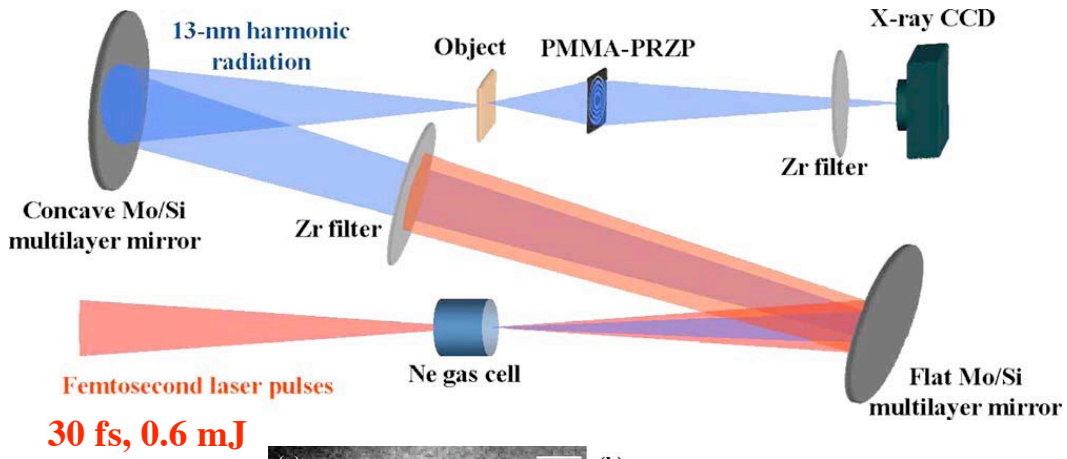


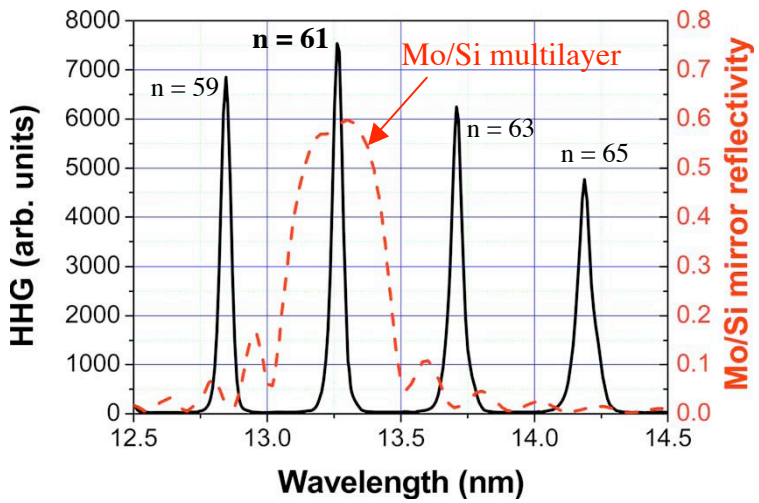


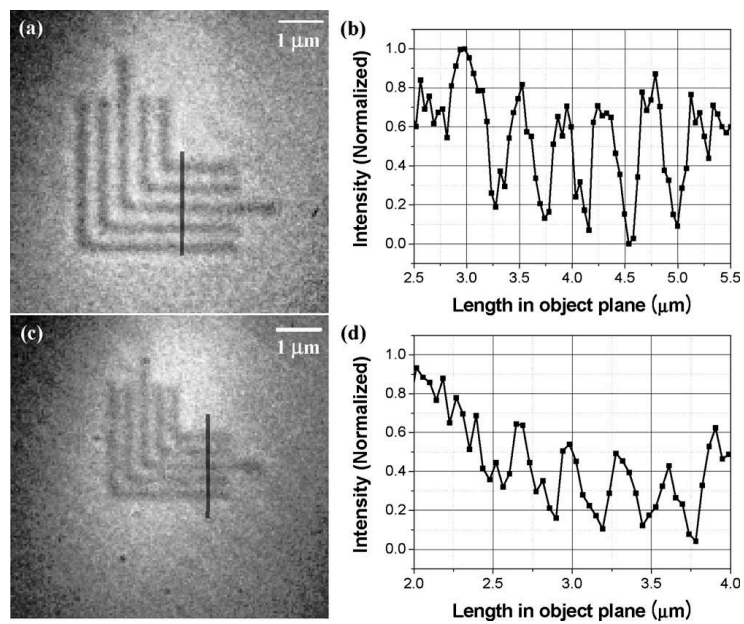
Attosecond Pump Probe: Exploring Ultrafast Electron Motion inside an Atom
S. X. Hu and L. A. Collins, PRL **96,** 073004 (2006)

Zone plate imaging with femtosecond EUV pulses

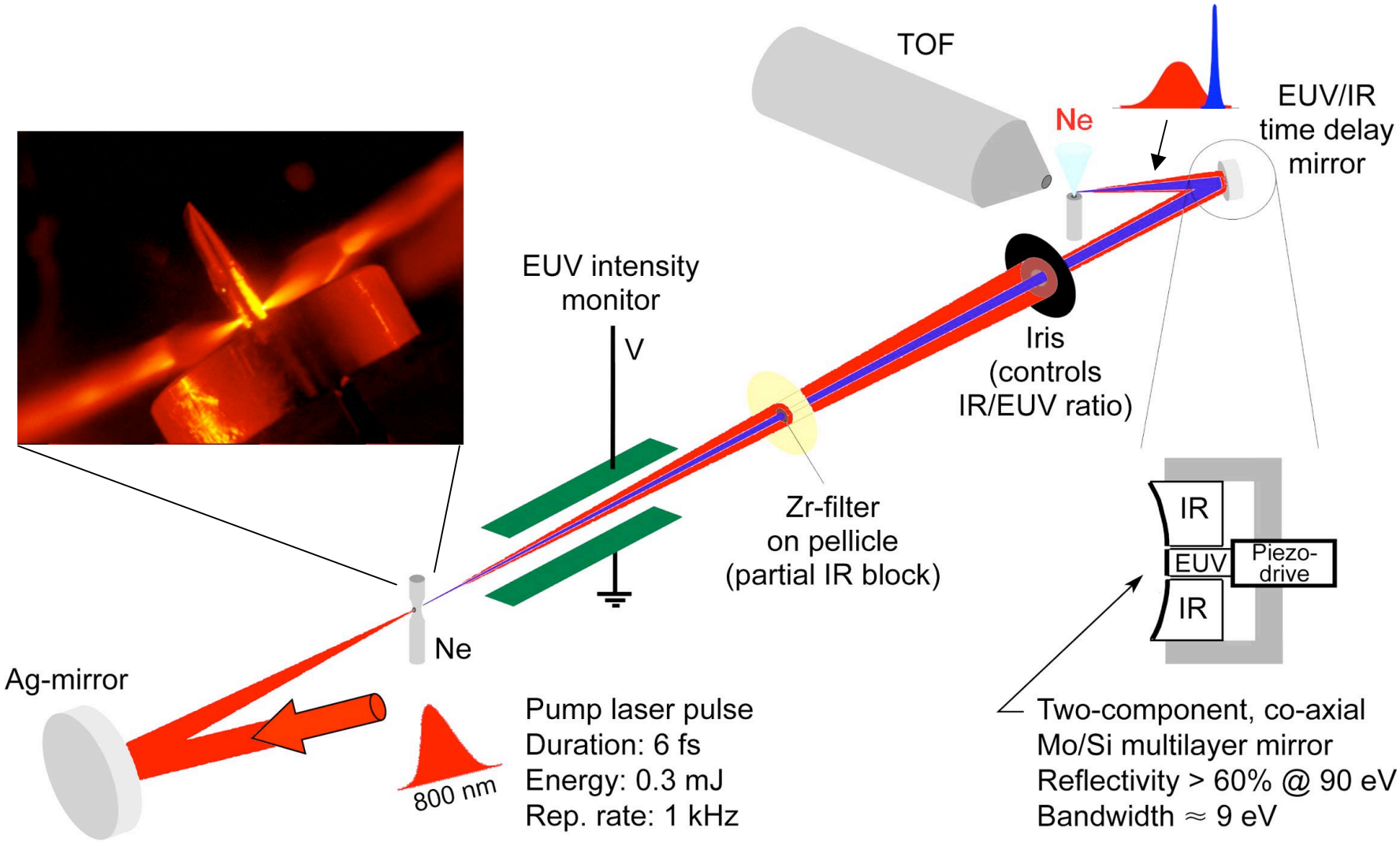
Jong Ju Park et al., "Soft x-ray microscope constructed with a PMMA phase-reversal zone plate," Opt. Lett. **34**, 235-237 (2009)







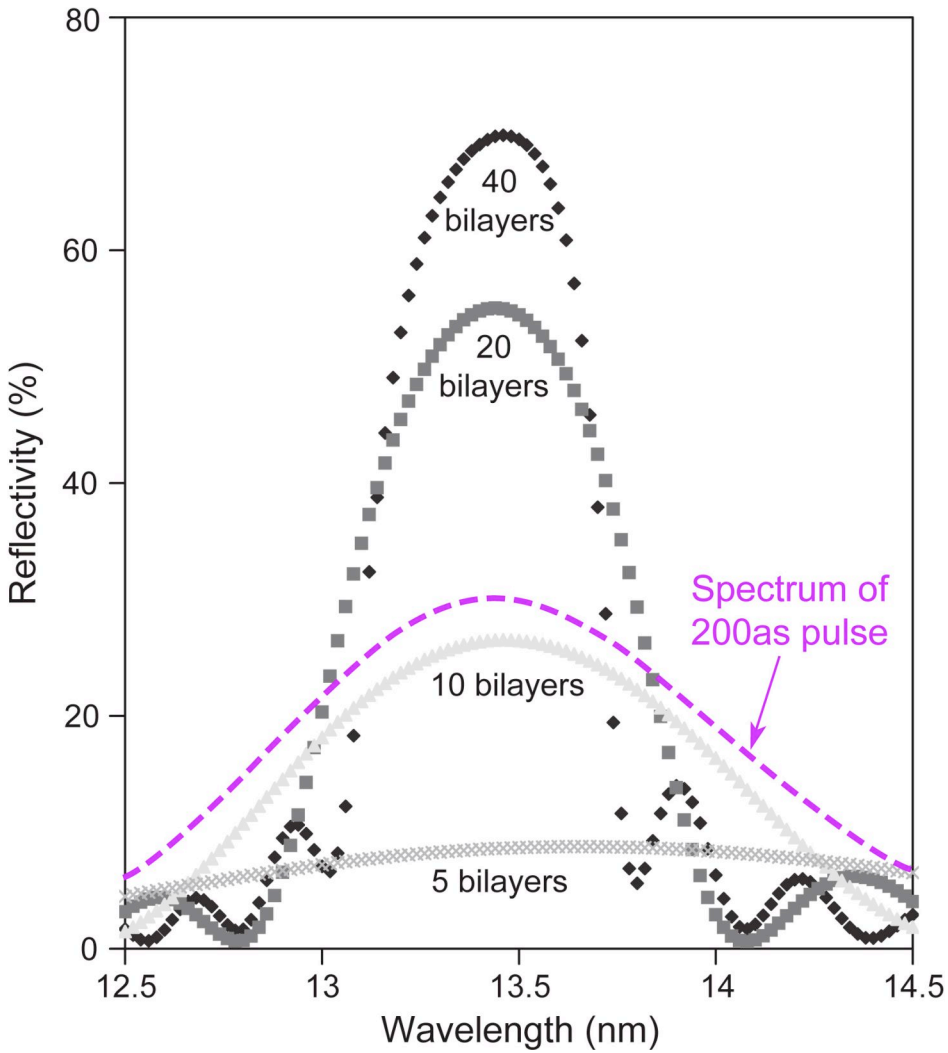
Co-axial multilayer optics used in HHG pump-probe experiments



from Dr. R. Kienberger

M. Drescher *et al.*, Science **291**, 1923 (2001)

Reflectivity ← → Bandwidth



- Multilayer mirrors depend on constructive interference from individual interfaces
- Higher reflectivity needs more layers
- Bandwidth gets narrower with more layers

Attosecond pulse

→ Broad bandwidth

→ Limited number of layers

N<10 layers required for 200 as pulse (@13nm)

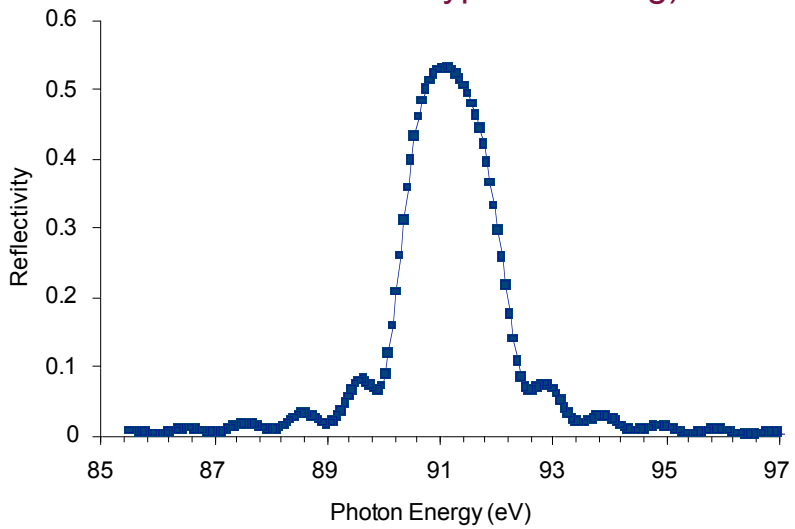
Narrow bandwidth coatings for picking a single harmonic

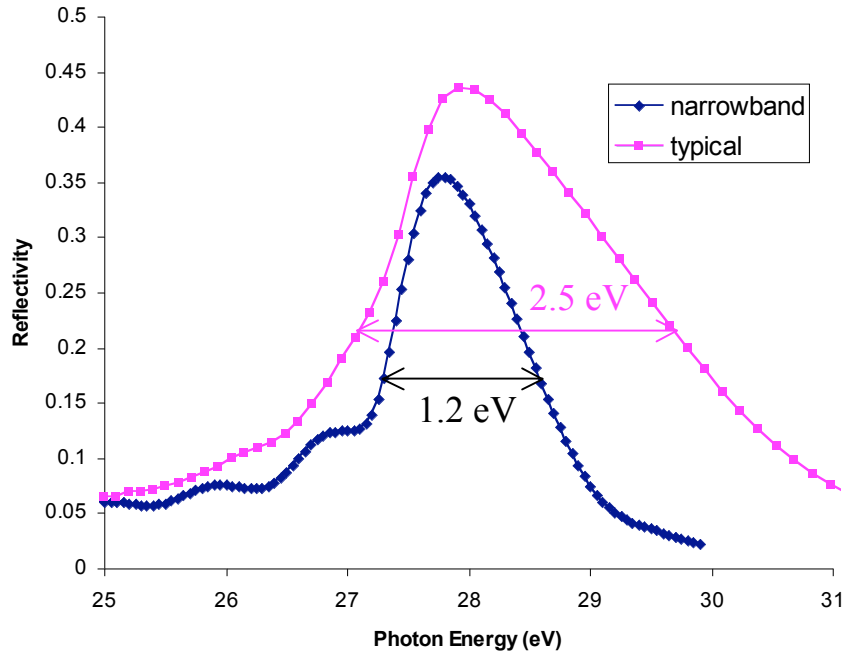
Higher order of multilayer mirror: $2d\sin\theta = m\lambda$ (m= 2,3,…)
Isolate a single harmonic order

FWHM = 1.8 eV

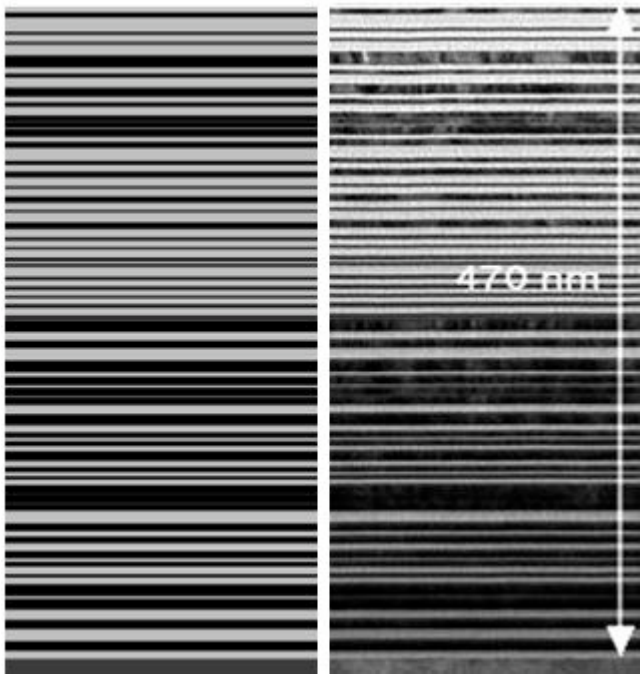
(2% relative bandwidth

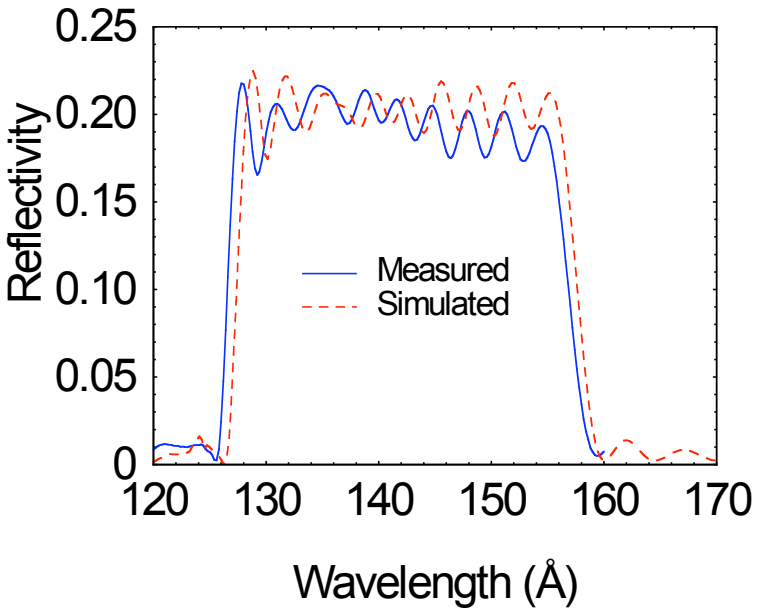
vs 3.5% for typical coating)





Aperiodic ‘supermirror’ with 20 eV bandwidth



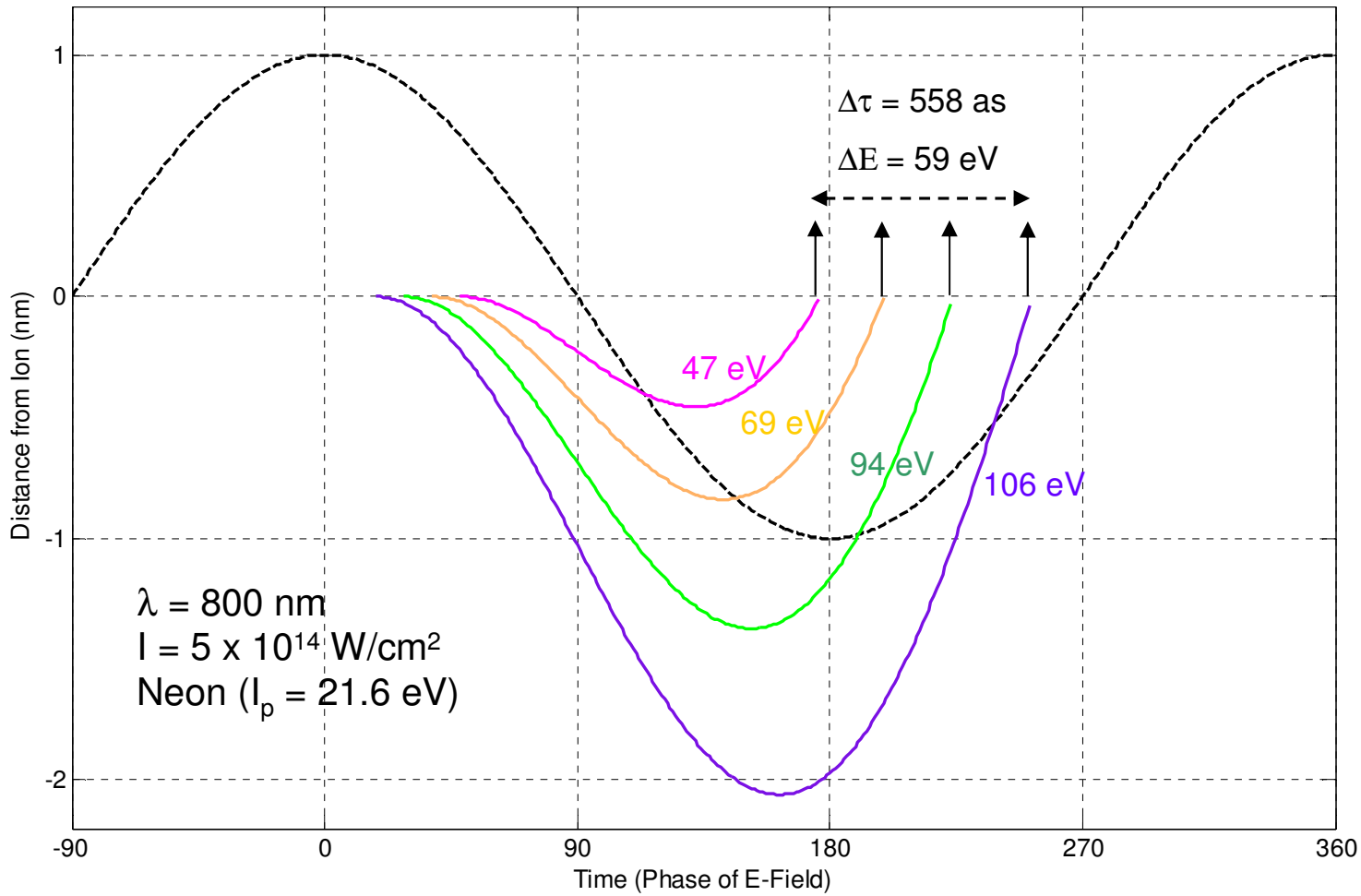


*A. L. Aquila, F. Salmassi, F. Dollar, Y. Liu, and E. Gullikson, "Developments in realistic design for aperiodic Mo/Si multilayer mirrors," Opt. Express **14**, 10073-10078 (2006)*

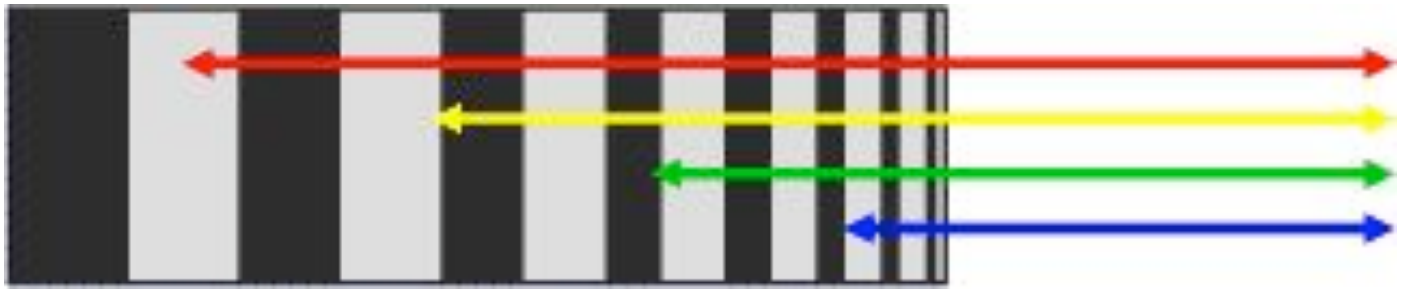
Intrinsic HHG chirp

Chirp in sub-fs scale:

Different energy photons are emitted at slightly different times



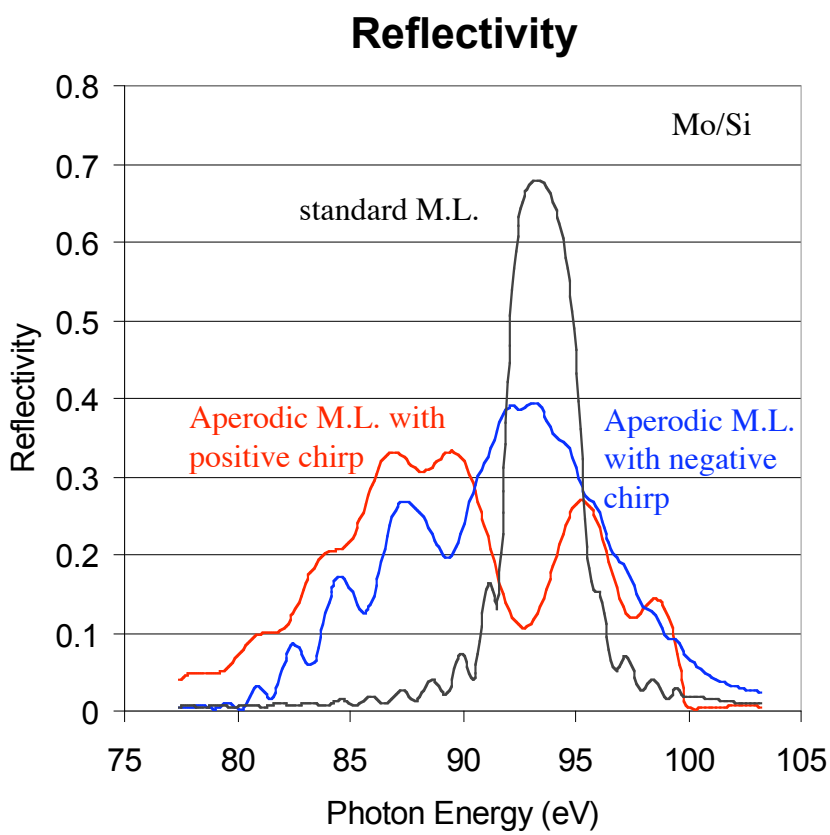
Chirped Mirror for Phase Control

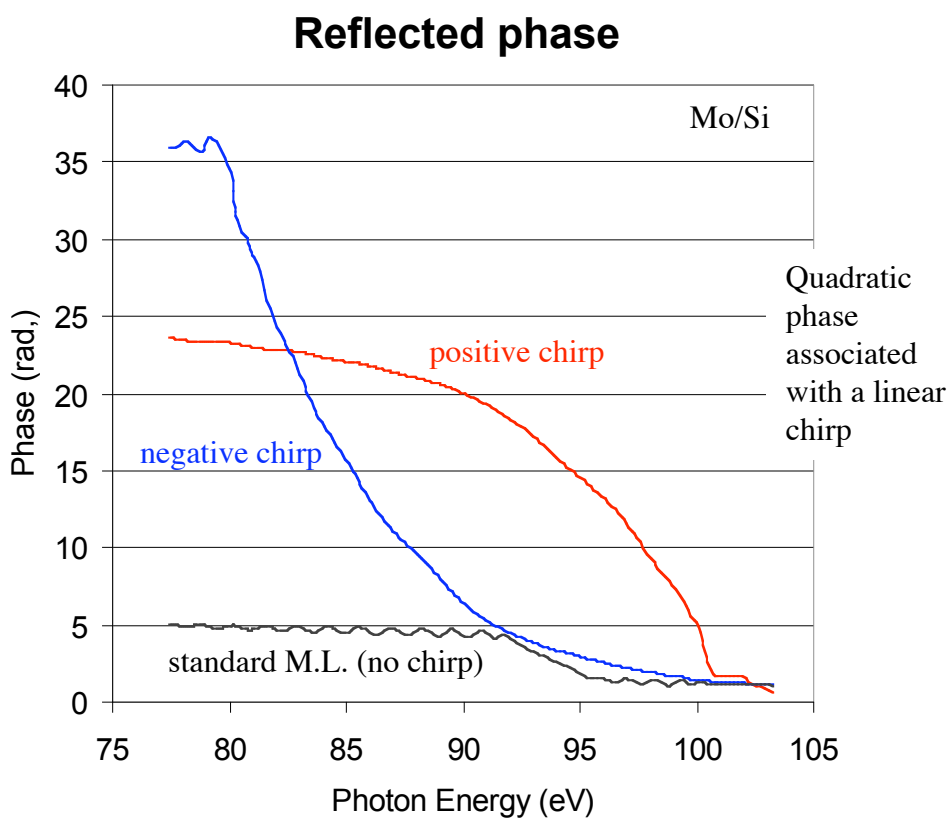


Effective ‘depth’ for different wavelengths are different

Aperodic mirror would provide more control of the spectral phase across wide bandwidth

Chirped multilayer mirrors with controlled phase can be used to compensate chirp for pulse compression





A. Aquila et al, Opt. Lett. 33 (455), 2008

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P. H. Bucksbaum, *Science 317, 766 (2007)*
E. Goulielmakis, *et al.*, *Science 317, 769 (2007)*
H. Kapteyn, et al., *Science 317, 775 (2007)*

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- R. A. Bartels et al., Opt. Lett. 27, 707 (2002).
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