Soft X-Rays and Extreme Ultraviolet Radiation

High-Harmonic Generation II

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

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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)

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

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

Pressure phase matching

Intensity (arbitrary units)
Pressure (torr)

n = 29
2x
n = 31
Fully coherent EUV from HHG in hollow fiber


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

Courtesy of Professors Margaret Murnane and Henry Kapteyn, Univ. Colorado.
Short Modulation Period Capillaries Extend Phase Matching from 85 eV (Unmodulated Fibers) to 160 eV

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
Quasi phase matching using counter-propagating pulses

Picosecond pulses, HHG coherence length ~ 1mm.

Quasi phasematching using CW counter-propagating laser

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

\[ \lambda = 800 \text{ nm}, \ I = 5 \times 10^{14} \text{ W/cm}^2 \]

Neon \((I_p = 21.6 \text{ eV})\)

1.3 fs

2.7 fs
Isolated attosecond pulse generation using few-cycle pump

High-harmonic generation

Bandpass filter

Visible light field

Soft X-ray intensity

Intense peak generates highest photon energy

Bandpass filter selects highest photon energy in a single attosecond pulse

Usually a multilayer to select high energies

Carrier-Envelope Phase (CEP) of ultrafast pulse

Cosine wave

Sine wave
Cosine waveform generates single attosecond pulse

While sine waveform generates two attosecond pulses

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

EUV frees electrons, IR electric field accelerates these electrons. These electrons arrive in waves via time-of-flight tube to detector.

Overlap of 7 fsec IR pulse and 250 as EUV pulse.

Unprecedented time resolution

Attosecond spectroscopy in condensed matter
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 CH₂Br₂

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
Zone plate imaging with femtosecond EUV pulses

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

from Dr. R. Kienberger

Reflectivity $\leftrightarrow$ 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: \(2d \sin \theta = m\lambda\) (\(m = 2, 3, \ldots\))
Isolate a single harmonic order

\[
\text{FWHM} = 1.8 \text{ eV}
\]
(2% relative bandwidth vs 3.5% for typical coating)
Intrinsic HHG chirp

Chirp in sub-fs scale:
Different energy photons are emitted at slightly different times

\[ \Delta \tau = 558 \text{ as} \]
\[ \Delta E = 59 \text{ eV} \]

\[ \lambda = 800 \text{ nm} \]
\[ I = 5 \times 10^{14} \text{ W/cm}^2 \]
Neon (\[ I_p = 21.6 \text{ eV} \])

\[ 47 \text{ eV} \]
\[ 69 \text{ eV} \]
\[ 94 \text{ eV} \]
\[ 106 \text{ eV} \]
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

References


• E. A. Gibson et al., Science 302, 95 (2003).