Science at the Time-scale of the Electron – Coherent Attosecond Soft and Hard X-Ray Harmonics

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I. Bright coherent x-ray beams on a tabletop
   — Ultrafast lasers can manipulate electrons on their fundamental timescale to implement a coherent version of the x-ray tube
   — Bright coherent tabletop beams at > 0.5 keV
   — Duration of 10 attoseconds – soon zeptoseconds!

II. Ultrafast x-rays are an ideal probe of the nanoworld
   — Image reactions at the level of electrons
   — Understand energy/charge transport at the nanoscale
   — Capture correlated electron dynamics (spin, molecules, materials)
   — Elemental and chemical nanoprobe of thick samples
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Coherent light has transformed society

In 1960, research physicist Theodore H. Maiman built the first ruby laser at Hughes Laboratory in Malibu, California.
And so have x-rays....
So, what about coherent x-rays? X-ray lasers

Spontaneous emission

\[ \frac{A_{21}}{B_{21}} = \frac{8\pi h \nu^3}{c^3} \propto \nu^3 \]

Stimulated emission

\[
\text{Power} \propto \left( \frac{1}{\sigma_g} \right) \left( \frac{1}{\tau} \right) (h \nu) \propto \frac{1}{\lambda^5}
\]

- 1 µm -> 1 mW
- 1 nm -> TW
- 1 Å -> 1 PW

LCLS X-ray free electron laser at 1.5 nm
P.A. Franken et al, PRL 7, 118 (1961)

An alternative approach: Nonlinear Optics

An alternative approach: Nonlinear Optics

Ruby laser
Lens
Quartz crystal
Prism
Photographic plate

$E_{\text{Pump}}(t)$
$d(t)$ electric dipole moment

$E_\text{P}(\omega) = E_0 e^{i\omega t}$

$d(\omega) = a_1 e^{i\omega t} + a_2 e^{i2\omega t} + \ldots$

Log intensity
Frequency
Extreme NLO: start with an ultrafast laser pulse

10 fs light pulse:

Δx = 3 micrometers
= 1/50 human hair

First 10 fs Ti:sapphire laser
Optics Letters 18, 977 (1993)
High-order Harmonic Generation–NLO using unbound states

- Coherent x-rays generated by focusing an intense fs laser into a gas
- Broad range of harmonics generated simultaneously from UV – keV
- Discovered in 1987, explained in 1993

High Harmonic Generation

- Coherent x-rays generated by focusing an intense fs laser into a gas
- Broad range of harmonics generated simultaneously from UV - keV

Electron wriggle energy coherently converts to x-rays

\[ h\nu_{\text{max}} \propto I_{\text{laser}} \lambda^2 \]
• Electron takes a few fs to leave vicinity of an atom after being ionized
• During that time, wavefunction is highly modulated due to recollisions
• Modulations give rise to high harmonics in radiated field

Electron wavefunction

X-ray field

40 Å
How to match the laser and x-ray phase velocities?

- Place gas inside a hollow fiber
- Tune the gas pressure to equalize the laser and x-ray phase velocities

\[ \Delta k = q k_{\text{laser}} - k_{\text{HHG}} = 0 \]

\[ \Delta k = q \left( \frac{u_{11}^2 \lambda_0}{4 \pi a^2} - P \left( 1 - \eta \right) \frac{2\pi}{\lambda_0} \Delta \delta - \eta \left[ N_{atm} r e \lambda_0 \right] \right) \]

\[ V_{\text{laser}} = V_{\text{x-ray}} = C \]
Generating bright, coherent, x-ray beams

- Tune the gas pressure to equalize the laser and x-ray phase velocities
- Generate fully coherent, bright, harmonics in EUV region < 150 eV

Fully coherent bright EUV and soft x-rays
10^{-5} or nJ per harmonic, uW average powers
Femtosecond-to-attosecond duration

Science 280, 1412 (1998)
Science 297, 376 (2002)
Limits of phase matching

- To generate high energy x-rays, need high electron energy - ionize gas
- Presence of plasma speeds up laser - no phase matching above 150 eV

\[ \Delta k = qk_{\text{laser}} - k_{\text{HHG}} = 0 \]

\[ \Delta k = q\left(\frac{u^2 \lambda_0}{4\pi a^2}\right) - P\left(1 - \eta\right)\frac{2\pi}{\lambda_0}\Delta \delta - \eta [N_{\text{atm}} r_e \lambda_0] \]

\[ V_{\text{laser}} = V_{\text{X-ray}} \neq C \]
Generating very high energy harmonics

Single atom cutoff photon energy: \( h \nu_{\text{cutoff}} = I_p + 3.2 I_L \lambda_L^2 \)

- \( \lambda_L = 0.8\mu m \)
- \( \lambda_L = 1.6\mu m \)

Ionization high - no phase matching

Single atom yield \( \propto \lambda_L^{-5.5} \)

Nature 433, 596 (2005)

PRL 98, 013901 (2007)
PRL 99 253903 (2007)
PRL 100, 173001 (2008)
Phase matching using mid-infrared lasers

- IR lasers need lower intensity for a given harmonic energy
- Lower laser intensity $\Rightarrow$ lower ionization, better phase matching
- Predict $h\nu_{PM} \propto \lambda_L^{(1.6-1.7)}$
- Single atom response also lower for mid-IR drivers ($\lambda^{-5.5}$)
- BUT phase matching pressure and gas transparency increase and compensate for low yield!

T. Popmintchev et al. CLEO Postdeadline CPDA9 (2008)
Optics Letters 33, 2128 (2008); PNAS, 106, 10516 (2009);
Nature Photonics, tbp (2010)
Full phase matching at $>0.13$ keV using $\lambda_L = 0.8 \mu m$

Full phase matching at >0.3 keV using $\lambda_L = 1.3 \, \mu m$

Popmintchev et al.,
CLEO Postdeadline CPDA9 (2008);
Opt. Lett. 33, 2128 (2008);
PNAS 106, 10516 (2009);
Nature Photonics to be publ. (2010)
Full phase matching at >0.5 keV using $\lambda_L = 2 \mu$m
Broad soft x-ray supercontinuum spans > 300 eV

HHG flux high: $10^6$ photons/s in $\lambda/\Delta\lambda$ of 100

- He / 1000 torr
- Ne / 2400 torr
Bright spatially coherent laser-like output

HHG flux high: $10^6$ photons/s in $\lambda/\Delta\lambda$ of 100

Spatially coherent beams
200 attosecond pulse generation at 50eV

- Phase matching occurs over 1 laser cycle
- Using 15 fs driving laser at 0.8 μm, 200 attosecond pulses can easily be generated
- Full characterization using FROGCRAB

Thomann et al., Optics Express 17, 4611 (2009)
10 attosecond pulse generation at 400eV

- Phase matching occurs over 1 laser cycle
- Using 35 fs driving laser at 2 μm, 10 attosecond pulses can easily be generated
- Predict zeptosecond pulses at > keV
Scaling of phase matched HHG flux to keV and beyond

- Very favorable scaling to multi-keV region!!
- Low gas absorption of HHG
- Large pressure-length products mitigate the low $\chi_{\text{eff}}$
- Low nonlinear distortion of laser pulse due to ionization
- At laser wavelengths $> 3 \, \mu m$, group velocity mismatch and magnetic field effects may reduce the HHG flux

$$dI_q \propto \frac{\omega_q^2 \rho^2 |s_q|^2}{\alpha_q^2 + \Delta k^2} \left(1 + e^{-\frac{L}{L_{\text{abs}}}} - 2e^{-\frac{2L}{L_{\text{abs}}}} \cos \Delta kL\right)$$
First high harmonics driven by 3.55 µm light

- Observed first UV/EUV/SXR high-order harmonics driven by 3 µm lasers
- Magnetic field effects do not reduce HHG yield
- Route to zeptosecond keV harmonics using mid-IR driving lasers at 3.55 µm!
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**Applications of ultrafast coherent x-rays**

### Nanoscience (50 nm)
- Ultrafast, elemental, magnetic switching speeds (thin samples)
- Nanoscale heat flow

### Metrology for EUV Lithography
- EXAFS/PEEM: Image reactions at the level of atoms and electrons. Understand and control function.

### Bio-imaging
- Cellular and materials tomography (10 nm, elemental)

### Nanoscience (<10 nm)
- Dynamic imaging of advanced nano-structures (magnetic, thermal etc.)
- Protein crystallography

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**Soft x-rays are ideal probes of nanoworld:**
- Penetrate thick objects and image small features
- Elemental and chemical specificity if HHG can extend to x-ray absorption edges
- Applications to date limited to $\approx 100$ eV
Applications of coherent ultrafast x-rays span broad range of science

Molecular imaging: image changing electronic orbital and molecular structure

Surface science: probe electronic dynamics on catalysts, photovoltaics

Magnetics: Probe nanodomains, magnetic dynamics

Nanoimaging: High resolution 3D imaging of thick samples using coherent lensless imaging

High frequency acoustic metrology: Characterize thin films, interfaces, adhesion

Nanothermal transport: probe heat flow in nanostructures
Ultrafast coherent magnetism – faster, denser, more efficient?

Beaurepaire et. al PRL 76, 4250 (1996)

Coherent ultrafast magnetism induced by femtosecond laser pulses
Jean-Yves Bigot*, Mircea Vormir and Eric Beaurepaire

Stamm, et al., Nature Mat. 6, 740 (2007)
Ultrafast element-selective demagnetization dynamics

- Demagnetize permalloy using IR laser, probe using M-edge harmonics
- Highest time resolution ≈ 55fs and elemental specific measurement
- First result: Fe and Ni in Permalloy both decay within ≈ 400 fs since they are strongly exchanged coupled even during non-adiabatic heating
- Next steps – attosecond, domain imaging, L-edges etc.
- Collaboration with NIST, Kaiserslautern, Julich

*Phys. Rev. Lett.* **103**, 257402 (Dec 2009)
Ultrafast element-selective demagnetization dynamics

- Demagnetize permalloy using IR laser, probe using M-edge harmonics
- Highest time resolution ≈ 55fs and elemental specific measurement
- Second result: Fe and Ni in Permalloy both decay within ≈ 100 fs at lower sample temperatures
- Next steps – attosecond, domain imaging, L- edges etc.

Phys. Rev. Lett. 103, 257402 (Dec 2009)
Understanding nanoscale heat flow

- Heat is carried by phonons
- In the macroscopic world, Fourier Law applies
  \[ q = -k \nabla T \]
- What happens when a nanostructure is smaller than the phonon mean free path?
- Existing theories of nanoscale heat dissipation disagree
- Fourier law over-estimates the heat flow - need to think of interface \( \approx \) phonon mean free path
- Ronggui Yang, Keith Nelson, Erik Anderson (Nature Materials 9, 26 (2010))
Following entire valence shell electron density rearrange as a bond breaks

W. Li, A. Becker et al., submitted (2010)
Following entire valence shell electron density rearrange as a bond breaks
Following entire valence shell electron density rearranges as a bond breaks

- SFI sensitive to electron density dynamics of entire valence shell
- Many electrons must rearrange as bond breaks!
- Electrons take a surprisingly long time to localize onto the atoms and it happens abruptly!
- See Andreas Becker talk on friday
How fast can we switch the electronic state of a solid?

- TiSe$_2$ undergoes a photo-induced phase transition
- Probe entire band structure using angle-resolved HHG photoemission
- Prof. Michael Bauer, University of Kiel

$T < 200K$: insulating charge density wave state

$T < 200K$: normal metallic phase
Radiation Femtochemistry

- Expt #1: Decay of $\text{N}_2^+$ excited by 43eV
  - Can identify the Rydberg dissociative states
  - Can follow dynamics as system changes from symmetric to 2-center
  - Theory by Xiao-Min Tong

$\text{N} - \text{N} \quad + \quad \text{N} \quad + \quad - \quad \text{N} \quad +$

$\text{N}^+ - \text{N}^+$

$\text{N} - \text{N}^+$

$\text{shake-up state}$

$43\text{eV}$

$\text{Time (fs)}$

$0 \quad 100 \quad 200$

$\text{Science 317, 1374 (2007)}$
• Expt #2: Decay of $\text{O}_2^+$ excited by 43eV, theory by Robin Santra
  – Create long-lived superexcited states in O through Feschbach resonance
  – System still bound up to 30 Å

*Sandhu et al., Science* 322, 1081 (2008)*
Lensless microscopy using coherent x-ray beams

- No aberrations - diffraction-limit in theory
- Image thick samples
- Inherent contrast of x-rays
- Robust geometry, insensitive to vibrations
- Requires a coherent beam of light and an isolated sample

Sayre, Acta Cryst 5, 843 (1952)
High rep rate (1 - 10kHz) femtosecond laser

High average power fs laser system + High harmonic converter + Coherent diffractive microscope
Lensless coherent imaging – 50nm resolution

- Combine lensless imaging with holography for faster, high resolution imaging
- Resolution of 50 nm represents 1.6 λ
- Future: sub-10 nm imaging of thick samples with element specificity
- Applications in bioimaging, magnetics, nano, thermal, lithography…….

Sandberg et al., Opt. Lett. 34, 1618 (2009)
• Take attosecond electron rescattering physics, discovered just over 20 years ago, to generate coherent x-rays and electrons

• Now have coherent soft x-ray laser beams that span to 0.5 keV, with enough flux for expts., and with attosecond pulse duration, and with excellent prospects for hard x-ray laser beams on a tabletop

• Table-top microscopes, nanoprobe, nanomanipulation and x-ray imaging with unprecedented spatial and temporal resolution

• Thanks to NSF, DOE, DOD
Students and postdocs

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