Generation of Femtosecond X-ray Pulses from the Advanced Light Source

Robert W. Schoenlein

Materials Sciences Division
Lawrence Berkeley National Laboratory

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Outline

Scientific Motivation
- Structural dynamics in condensed matter on femtosecond time scale
- X-ray source requirements and experimental considerations

Generation of Femtosecond X-rays from 3rd Generation Synchrotrons
- Manipulation of the stored electron beam with femtosecond laser pulses
- Diagnostics (optical gain measurements, coherent THZ generation)
- Results from proof-of-principle experiments at the ALS
- Future prospects, limitations, practical issues – experimental applications

New Femtosecond X-ray Facility at the Advanced Light Source
- Small-gap, in-vacuum undulator
- Soft x-ray and hard x-ray branchlines
- High average power (high repetition rate) femtosecond laser system

Future Femtosecond X-ray Sources
**Fundamental Scientific Challenge in Condensed Matter:**

**Understanding the interplay between atomic and electronic structure**
- beyond single-electron band structure model – correlated systems (charge, spin, orbit, lattice)
- beyond simple adiabatic potential energy surfaces

**Fundamental Time Scales in Condensed Matter**

**Atomic Structural Dynamics**
- fundamental time scale for atomic motion
  - vibrational period: $T_{\text{vib}} \sim 100$ fs
  - (femto $\sim 10^{-15}$)
- ultrafast chemical reactions
- ultrafast phase transitions
- surface dynamics
- ultrafast biological processes

**Electronic Structural Dynamics**
- fundamental time scales for electron dynamics
  - electron-phonon interaction times $\sim 1$ ps
  - e-e scattering times $\sim 10$ fs
  - correlation time $\sim 100$ attoseconds ($a/V_{\text{Fermi}}$)
- (atto $\sim 10^{-18}$)
- charge transfer
- electronic phase transitions
- correlated electron systems
  - charge/orbital ordering
  - CMR
  - high $T_c$ superconductivity

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**Ultrafast X-ray Science**
Rapidly emerging field of research - Physics, Chemistry and Biology
Femtosecond Spectroscopy

- transmission
- reflection
- photoemission

- non-linear (probe) $\sim |I_{\text{probe}}(\Delta t)|^n$
  harmonic $\omega_1 \pm \omega_2$

- transient four-wave mixing
- photon echo
Rhodopsin - photoreceptor for vision

- **cis-trans isomerization complete in 200 fs**
- **vibrationally coherent**

![Graph and diagram showing wavelength vs. delay and structural changes with Schoenlein et al. Science (1991) and Wang et al. Science (1994).]
Femtosecond X-ray Science

time-resolved x-ray spectroscopy

EXAFS – **local atomic structure** and coordination
(extended x-ray absorption fine structure)

NEXAFS – **local electronic structure**, bonding geometry,
magnetization/dichroism

(element specific)

molecular systems and reactions
complex/disordered materials

surface EXAFS, μEXAFS ....

\[ f(r) \sim \frac{\sin(2kr + \phi(k))}{kr^2} \]

**time-resolved x-ray diffraction**
atomic structure in systems with long-range order/periodicity
phase transitions, coherent phonons

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Fe$^{II}$ Spin-Crossover Molecules

Motivation:
- relationship between structure, electronic, and magnetic properties

Do the structural distortions facilitate the spin-crossover reaction?
- electron transfer mechanistic role in biochemical processes (cytochrome P450)
- magnetic and optical storage material
Fe(II) Spin-Crossover X-ray Spectroscopy

Time-resolved XAS - ALS Beamline 5.3.1

- Laser: 75 fs, 400 nm
- Flowing jet: 0.5 mm, ~30 mM
- Ge crystal mono chopper (2 kHz)
- APD detector
- Synchrotron (70 ps, 7.1 keV)


<table>
<thead>
<tr>
<th></th>
<th>Reactant</th>
<th>Photoexcited</th>
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<tbody>
<tr>
<td>N</td>
<td>6 ±0.5</td>
<td>6.5 ±1</td>
</tr>
<tr>
<td>R (Å)</td>
<td>1.94 ± 0.01</td>
<td>2.15 ±0.03</td>
</tr>
<tr>
<td>σ (Å²)</td>
<td>0.001</td>
<td>0.009</td>
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Characteristics for Ideal Source

(1) temporal resolution $\sim 100$ fs
   • pulse duration
   • synchronization to laser trigger

(2) high average flux $10^8$-$10^{13}$ photons/sec/0.1% BW
   • high average brightness $<1$ mrad source divergence

(3) tunable 0.3 keV - 10 keV
   • broadband - spectroscopy
   • soft x-rays (electronic structure)
   • hard x-rays (atomic structure)

(4) rep. rate: 100 Hz - 10 kHz
   • sample recovery or replacement
   • signal averaging (high stability)
3rd Generation Synchrotrons - Advanced Light Source

Microprobe

Crystallography

Interferometry

Microscopy

Microfabrication

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**3rd Generation Synchrotrons**

**Advanced Light Source**

- **Electron Storage Ring**: 1.9 GeV, 500 MHz, 400 mA, 70 ps bunch length
- **12 Straight Sections for Insertion Devices**
- **36 Bend Magnets**
- **High-Brightness Source of Soft X-rays 1-100 Å**

\[ \Delta \theta \sim \frac{1}{\gamma} \sqrt{\frac{1 + K^2/2}{Nn}} \]

\[ \frac{\Delta \lambda_n}{\lambda_n} \sim \frac{1}{nN} \]

\[ K = \frac{eB_n \lambda_w}{2\pi nc} \]

\[ \Delta \theta_{vert} \sim \frac{1}{\gamma} \]

\[ \Delta \theta_{horiz} \sim \frac{K}{\gamma} \]
Generation of Femtosecond X-rays from the ALS

Energy Modulation in the Wiggler

Wiggler - permanent magnets

Total field energy:
\[ A \sim \left \{ E_L(\omega, \tilde{r}) + E_R(\omega, \tilde{r}) \right \}^2 dSd\omega = A_L + A_R + 2 \sqrt{A_L A_R} \frac{\Delta \omega_L}{\Delta \omega_R} \cos \phi \]

Wiggler radiated energy:
\[ A_R \equiv \pi c \hbar \omega_K \frac{K^2 / 2}{(1 + K^2 / 2)} \]

Laser requirements:
\[ \hbar \omega_L = 1.55 \text{ eV} \]
\[ \Delta \omega_L = 27 \text{ period wiggler } \Rightarrow 36 \text{ fs laser pulse (72 fs)} \]
\[ A_L = 610 \mu J \text{ (780 } \mu J) \]

Since \( \sigma_E \sim E_o^2 \) and we want \( \Delta E \sim \sigma_E \) then the required laser pulse energy scales as: \( A_L \sim E_o^4 \)
Dynamics of Modulated Electron Beam

- ~70 ps electron bunch
- ~100 fs laser pulse
- $z,t$
- $x,E$

ALS Storage Ring Dispersion

- $\sigma_{\text{dispersion}} \sim 85$ fs per full arc
- (200 fs FWHM)

- $T_{\text{orbit}} = 656$ ns
- $300$ fs (BL6.3.2)
- $100$ fs (BL5.3.1)

- $t_{\text{damp}} = 6$ ms
Laser Gain through Wiggler (FEL Gain)

total optical field \[ \mathcal{E}_{\text{total}}(\omega) = \mathcal{E}_{\text{laser}}(\omega) + e_s(\omega) \]

\[ \mathcal{E}_{\text{total}}(\omega)^2 = \mathcal{E}_{\text{laser}}(\omega)^2 + e_s(\omega)^2 + 2\mathcal{E}_{\text{laser}}(\omega)e_s(\omega) \]

\[ e_s(\omega) = e_s(\omega_o) + \frac{\partial e_s(\omega_o)}{\partial \omega} \Delta \omega + \cdots \]

\[ \lambda_{\text{radiated}} = \frac{\lambda_{\text{wiggler}}}{2\gamma^2} \quad -\frac{d\lambda_r}{\lambda_r} = \frac{d\omega_r}{\omega_r} = \frac{2\Delta E}{E} \]

\[ \Delta E \propto \mathcal{E}_{\text{laser}} \]

\[ \mathcal{E}_{\text{total}}^2 = \mathcal{E}_{\text{laser}}^2 \left[ 1 + 2 \left( \frac{\partial e_s(\omega)}{\partial \omega} \frac{2\omega}{E} \xi \right) \right] + e_s(\omega)^2 + 2e_s(\omega_o)\mathcal{E}_{\text{laser}} \]

gain

\[ \text{gain} = \frac{(2\pi M)^2}{\gamma} \pi \frac{L_{\text{peak}}}{2 \int_A} \frac{d}{dx} \left( \frac{\sin^2(x)}{x^2} \right) \eta \quad x = \frac{\pi M (\omega - \omega_o)}{\omega_o} \]

\[ \eta \]

\[ I(t) \]

\[ I(\omega) \]

\[ \omega - \omega_o \]
Measurement of Laser Gain through Wiggler

\[ \lambda_{\text{radiated}} = \frac{\lambda_W}{2\gamma^2} \left(1 + K^2/2\right) = \lambda_{\text{laser}} \]

- **laser**
- **e-beam**
- **wiggler**
- **IF \(\lambda_1\)**
- **IF \(\lambda_2\)**

**Example Graph**: 
- **gain function**
- **laser spectrum**

**Graph Parameters**: 
- **delay (ps)**
- **gain (x10^{-3})**
- **central wiggler emission \(\lambda_0\) (nm)**

**Additional Information**: 
- \(\sigma = 16.6 \text{ ps}\)
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Femtosecond Pulses of Synchrotron Radiation

Femtosecond Pulses of Synchrotron Radiation

Calculated Electron Density Distribution

Laser System

\[ \hbar \omega_1 = 1.55 \text{ eV} \]
\[ \hbar \omega_2 = \approx 2 \text{ eV} \]
Coherent Infrared Radiation from Modulated Electron Bunch

- longitudinal modulation of electron density $\sim \lambda_{\text{IR}}$
- vacuum chamber aperture must accommodate source divergence (formation length)

\[
\begin{align*}
AB_{\text{photon}} &= 2 \rho \sin \left( \frac{\theta}{2} \right) \\
AB_e &= \rho \theta \\
AB_e - AB_{\text{photon}} &= \lambda_R \approx \rho \frac{\theta^3}{24} \\
L_{\text{formation}} &= \rho \theta \approx Z_R = \frac{\pi \omega_o^2}{\lambda_R}
\end{align*}
\]

\[\lambda_R \leq \rho^{-1/2} \omega_0^{3/2}\]

$\lambda_R \leq 1.4 \text{ mm} \ (\rho = 4 \text{ m}, \omega_o = 2 \text{ cm})$
Coherent Infrared – Measurement Schematic

Coherent Synchrotron Radiation

Coherent IR Detection

Waveguide Effects

Femtosecond x-ray Beamline 5.3.1

• 1:1 image of bend magnet source
  250 μm (H) x 50 μm (V)

• white beam, 0.1-12 keV
  (possibility for Laue diffraction)

• flux ~10^{13} ph/sec/0.1% BW (30 ps pulse duration)

flux ~10^5 ph/sec/0.1% BW
brightness ~10^8 ph/s/mm^2/mrad^2/0.1% BW
100 fs pulse duration
(5 kHz repetition rate)
Separation of Femtosecond X-rays

- Laser modulation of e-beam energy ($\Delta E$)
  - Laser power, wiggler matching
- Storage ring dispersion ($\Delta E \rightarrow \Delta x$)
  - Emittance and lifetime degradation
- Beamline image quality
  - Mirror scattering (non-specular)
  - Depth of source effects

**Requirement** – gated detectors for isolating individual bunches

- Avalanche photodiodes
- Gated microchannel plates

**Signature (fsec)**

\[
\frac{\text{Sig (fsec)}}{\text{background}} > 1
\]
Δρ(E) = ρ(E)_{laser on} - ρ(E)_{laser off}

Modulation: 3σ_E to 10σ_E  τ_{laser}/τ_e = 10^{-3}
Beam Profile Measurements – BL5.3.1 Camshaft Bunch

\[ \lambda = 1.2 \text{ Å} \]
\[ \sigma_x = 100 \mu\text{m} \]

- fraction of e-beam that is modulated = \( 75 \text{ fs} \times 0.1 /75 \text{ psec} = 10^{-4} \)

\[ \sigma_E/\Delta E=1/10 \]
Differential Beam Profile (Laser On-Laser Off)

- $\Delta E_{\text{max}} = 6\sigma_E$ (11.4 MeV)
- $\Delta E_{\text{max}} = 7\sigma_E$ (13.3 MeV)
- $\Delta E_{\text{max}} = 8\sigma_E$ (15.2 MeV)

Calculated Electron Density Distribution

Signal/bkg ratio

Femtosecond Flux / Background

Peak Laser Power ~ 10 GW
(0.8 mJ, 75 fs)
Science at time-resolved x-ray science beamline (ALS BL5.3.1)

- mÅ charge displacement – THz phonon-polariton mode in LiTaO$_3$

- Ultrafast insulator-metal transition in VO$_2$ - XANES

- Time-resolved EXAFS - spin-crossover transition in Fe[tren(py)$_3$]$^{2+}$
  M. Khalil et al., *J. Phys. Chem.* (in press)

- Bonding Properties of Liquid Silicon and Liquid Carbon

- X-ray/laser ionization dynamics in atomic systems
  M. Hertlein, A. Belkacem et al.
Femtosecond X-ray Facility – Scaling the X-ray Flux

- phase factor $\eta_1 = 0.1$ (fraction of electrons in optimum phase)
- pulse duration $\eta_2 = \frac{\tau_{\text{laser}}}{\tau_{\text{synchrotron}}} = 10^{-3}$ ($\tau_{\text{x-ray}} \approx 170$ fs)
- repetition rate $\eta_3 = \frac{f_{\text{laser}}}{f_{\text{synchrotron}}} = 2 \times 10^{-6}$

$$f_{\text{laser}} / f_{\text{synchrotron}} \approx 3 \times \frac{\text{number of bunches}}{\tau_{\text{damping}}} = 150 \text{ kHz}$$

**Average Femtosecond X-ray Flux ~ Average Femtosecond Laser Power**

**Bend Magnet**
- flux $\sim 10^{13}$ ph/sec/0.1% BW
- brightness $\sim 10^{16}$ ph/sec/0.1% BW

**Undulator**
- flux $\sim 10^{15}$ ph/sec/0.1% BW
- brightness $\sim 10^{19}$ ph/sec/0.1% BW
Femtosecond Undulator Beamline – Overview

I. Insertion Device
• highest possible flux and brightness 0.2-10 keV
• small-gap undulator/wiggler (1.5 T, 50 x 3cm period)
  \( \times 10^2 \) increase in flux, \( \times 10^3 \) increase in brightness

II. Beamlines for Femtosecond X-ray Science
• isolation of femtosecond x-ray, 0.2-2 keV, 2-10 keV
  sector 6 - proximity to existing wiggler 200 fs x-rays

III. Laser: average power/repetition rate
• 30 W (1.5 mJ per pulse, 20 kHz)
  \( \times 10 \) increase in flux

IV. Storage Ring Modifications
• local vertical dispersion bump – sector 6 and/or 5
In-Vacuum Undulator/Wiggler

S. Marks et al.

Specifications

- Magnetic gap: 5.5 mm
- Period: 30 mm
- No. periods: 50
- Vacuum gap: >5 mm
- $B_0$: 1.45 T
ALS Femtosecond Undulator Beamline 6.0 Layout

P. Heimann, D. Plate, H. Padmore, R. Duarte, D. Cambie et al.

- **Hard x-ray branchline**
  - (2-10 keV)
  - Separate M1 mirrors for hard and soft x-rays

- **Soft x-ray branchline**
  - (250 eV – 2 keV)

- **20 kHz femtosecond laser system**
  - 2x – 30 W

- **Hard x-ray hutch**

- **Sample chamber**
  - (dispersive soft x-ray)

- **Crystal monochromator**

- **Slits**
  - (intermediate focus)

- **20 kHz choppers**

- **Soft x-ray spectrograph**
ALS Femtosecond Undulator Branchlines

P. Heimann, D. Plate, H. Padmore, R. Duarte, D. Cambie et al.

**soft x-ray branch (0.2-2 keV)**

- **Source**: U3
- **Aperture for separating fs x-ray pulse**: M11, M21
- **Mirror**: M11, M21
- **Chopper**: Focus: 710 μm (H), 72 μm (V)
- **VLS Grating**: Focus: 60 μm (H), 37 μm (V)
- **M13 mirror**
- **Endstation 3 dispersive detector**

**Hard x-ray branch (2-10 keV)**

- **Source**: U3
- **Aperture for separating fs x-ray pulse**: M11, M21
- **Mirror**: M11, M21
- **Chopper**: Focus: 83 μm (H), 97 μm (V)
- **Ge(111), Si(111) crystal mono**
- **M23 mirror**
- **M24 mirror**
- **Endstation 2**

**Commission**

- **2005**
- **Fall 2006**
X-ray Chopper

- Absorbed x-ray power: ~500 W
- Slot dimensions: 3 mm horizontal x 300 mm vertical
- Rotating disk, water-cooled, in vacuum – ferro-fluidic feedthroughs
- Frequency: 20 kHz matched to laser repetition rate (100 slots at >12,000 RPM)
- Synchronization to the laser frequency: allowable phase error 250 ns
- LBNL/Rigaku design
First Light 10/4/05

M203

Sample Chamber and slits

M201 (behind wall)

Grating mono

Femtosecond Soft X-ray Beamline 6.0.1.2
**Femtosecond Laser System**

*R. Wilcox, R. Schoenlein*

*Electron beam interaction requirements:*
- ~1.5 mJ pulse energy, 60 fs FWHM, at ~800 nm
- 20 kHz repetition rate, 30 W average power
- Diffraction limited focusing, beam parameter: $M^2 \leq 1.1$

*Excitation pump pulses for time-resolved experiments:*
- ~1 mJ pulse energy at 800 nm (OPA)
- 60 fs pulse duration, 20 kHz repetition rate
- ~500 ns relative delay

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**Diagram:**

- **Oscillator**
  - Pulse duration: <75 fs
  - Energy: 2.0 nJ

- **Stretcher**
  - Pulse duration: 0.5 ns
  - Energy: 1.0 nJ

- **Regen**
  - Pulse duration: 0.5 ns
  - Energy: 300 μJ

- **2-Pass Amplifier**
  - Pulse duration: 0.5 ns
  - Energy: 300 μJ

- **Compressor**
  - Pulse duration: 60 fs
  - Energy: 1.5 mJ

- **2-Pass Amplifier**
  - Pulse duration: 0.5 ns
  - Energy: 5 μJ

- **Compressor**
  - Pulse duration: 60 fs
  - Energy: 1 mJ

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**cryogenic power amplifier**

- **PUMP BEAM**
  - Nd:YLF (2λ)
  - ~90 W, 10 kHz

- **HIGH REFLECTOR**
- **FARADAY ROTATOR**
- **1/2 wave plate**
- **77 K**
- **T:SAPPHIRE ROD**

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~1 mJ pulse energy at 800 nm (OPA)
60 fs pulse duration, 20 kHz repetition rate
~500 ns relative delay
Femtosecond Pulse Compression

Diffraction Grating Compressor

Transmission Grating Compressor

Power Densities: 10-100 W/cm²
Femtosecond X-ray Flux

- HHG flux from F. Krausz, laser: 10 fs, 3 mJ/pulse, 30 W
- Plasma source flux in mrad² laser: 40 fs, 1 mJ/pulse, 30 W (continuum includes projected $10^5$ improvement)
- Cu $K_{\alpha}$: $10^{10}$ ph/s/4π (proj. $10^{13}$ with Hg target)
- Cont. $6\times10^7$ ph/s/4π (integ. from 7-8 keV)

**ALS typical average x-ray flux**
- Undulator ~$10^{15}$ ph/s/0.1% BW
- Bend-magnet ~$10^{13}$ ph/s/0.1% BW
Ultrafast X-ray Facilities

- Femtosecond soft x-ray ‘slicing’ beamline – BESSY (operational)
- Femtosecond hard x-ray ‘slicing’ beamline – Swiss Light Source (commissioning)
- Femtosecond x-ray beamline – SOLEIL (planning)
- VUV free electron laser – DESSY (operational)
- X-ray free-electron laser – LCSL SLAC (under construction)
- X-ray free-electron laser – EURO X-FEL Hamburg (planning)
- FERMI@ELETTRA soft x-ray FEL – Trieste (design/construction)
- Soft x-ray FEL – BESSY (planning)
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