Femtosecond X-ray Science at the ALS: Recent Results and Future Plans

R.W. Schoenlein
H.W. Chong
C.V. Shank

T.E. Glover
P.A. Heimann

A. Zholents
M. Zolotorev

Materials Sciences Division
Advanced Light Source - Experimental Systems Group
Accelerator and Fusion Research Division - Center for Beam Physics

Outline

Scientific Motivation

- structural dynamics in condensed matter on femtosecond time scale

Generation of femtosecond x-rays at the ALS

- femtosecond slicing of electron beam
- results from proof-of-principle experiments
- practical issues/challenges

Future plans for femtosecond x-ray science at the ALS

- femtosecond x-ray beamline 5.3.1
- in-vacuum insertion device beamline for femtosecond x-rays

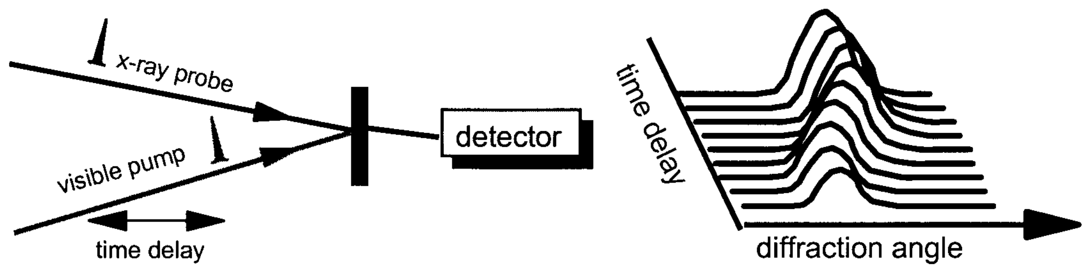
Structural Dynamics in Condensed Matter

fundamental time scale for atomic motion
vibrational period: h/kT ~ 100 fs

- ultrafast chemical reactions
- ultrafast phase transitions
- surface dynamics
- ultrafast biological processes

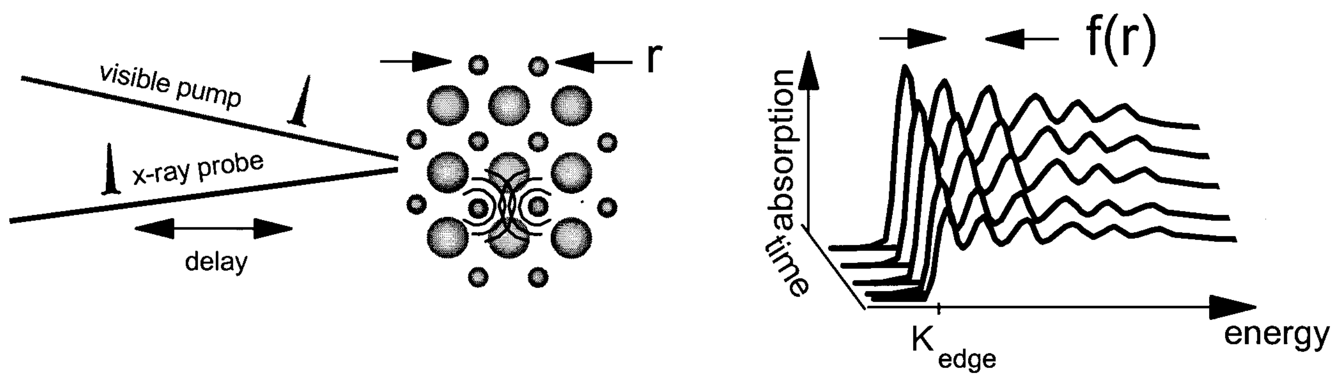
Rapidly emerging filed of research
Important scientific applications in Physics, Chemistry and Biology

time-resolved x-ray diffraction



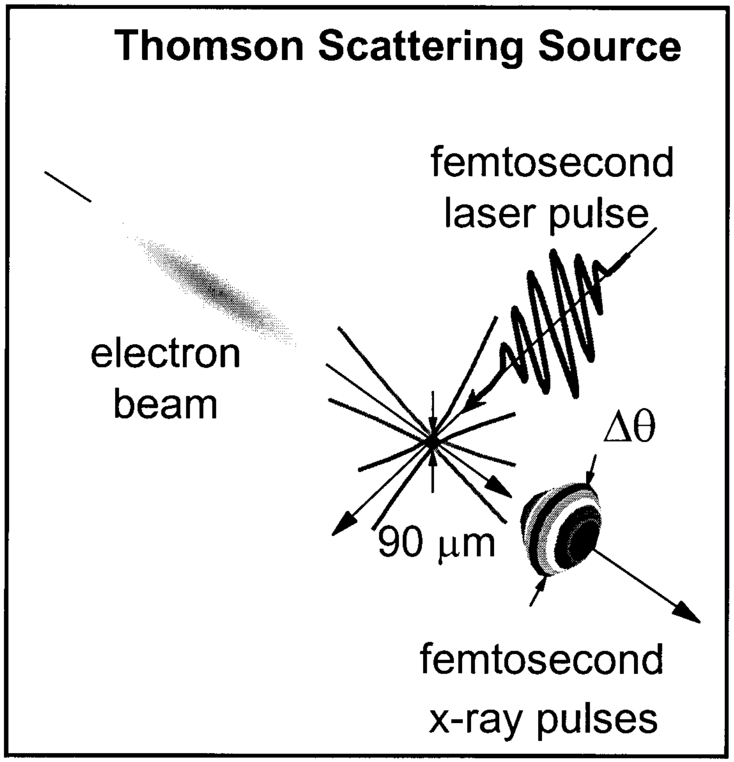
ordered crystals - phase transitions, coherent phonons

time-resolved EXAFS, NEXAFS, surface EXAFS

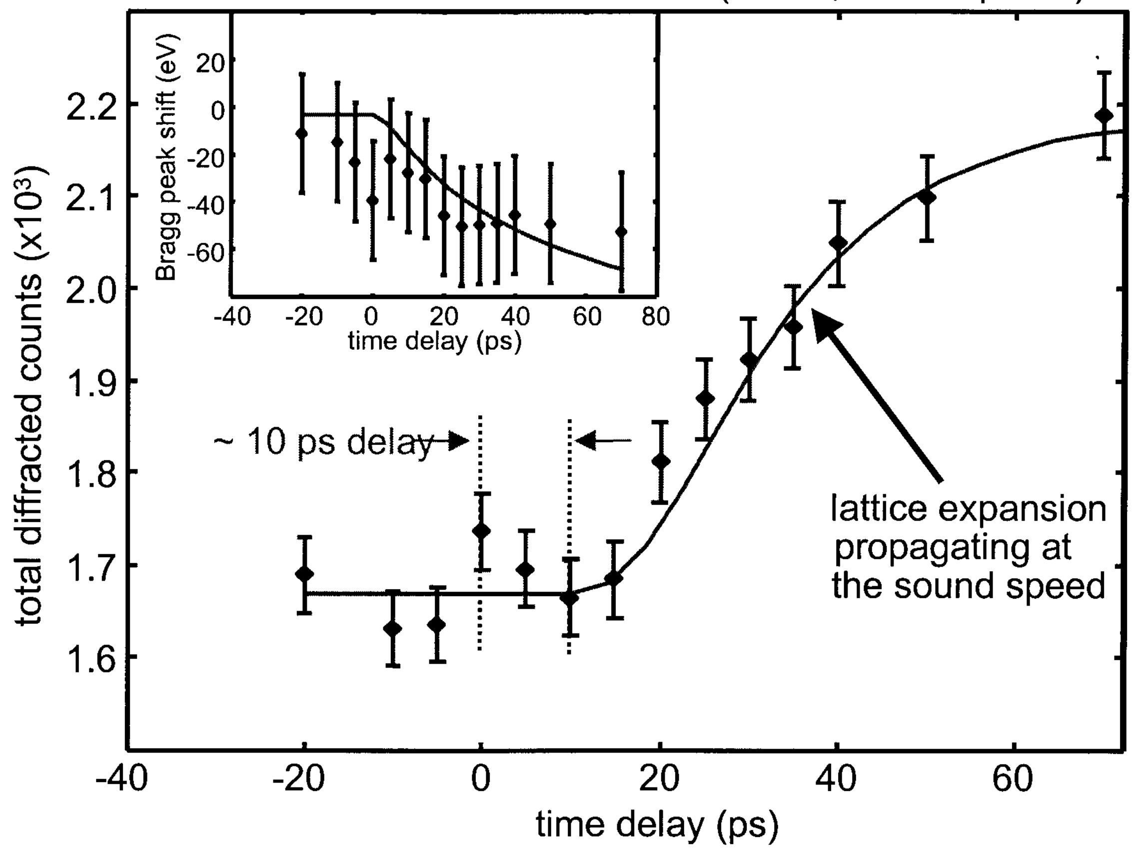


complex/disordered materials - chemical reactions, surface dynamics
bonding geometry

Laser Heated InSb Data



(300 fs, 30 keV probe)

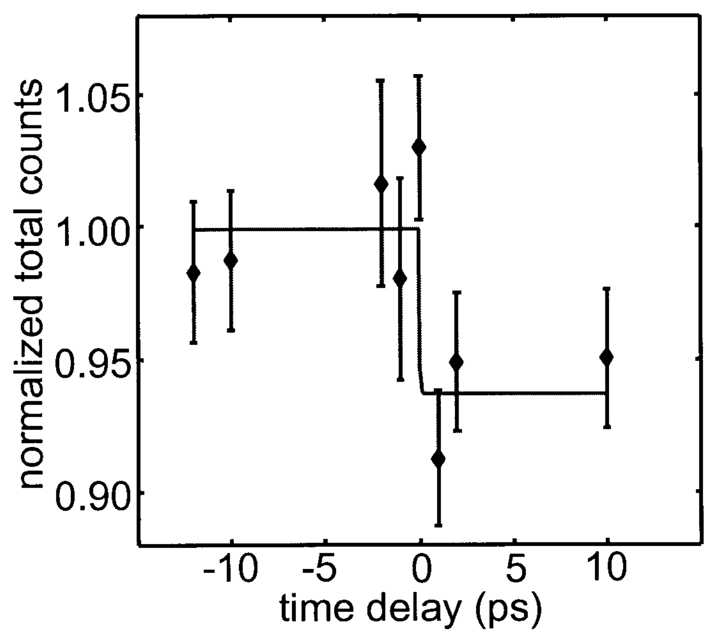


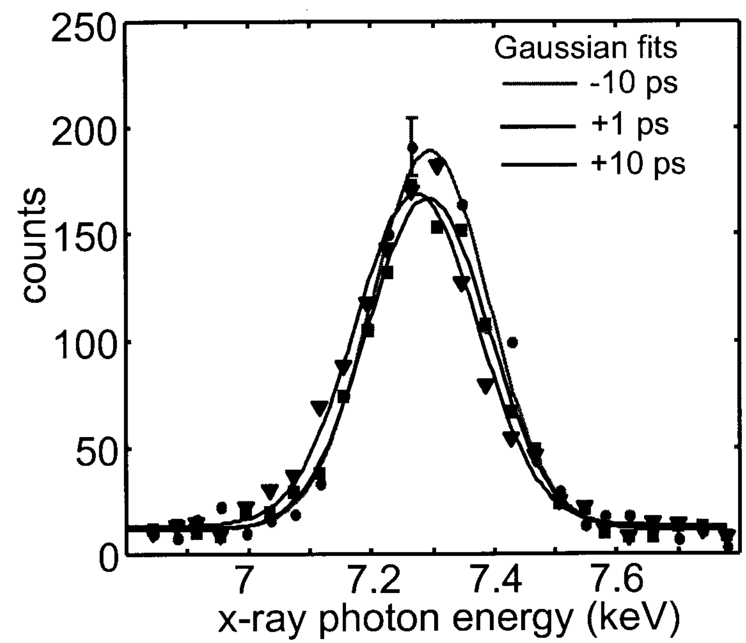
Indication of Ultrafast Disordering

- ~6 % reduction in integrated diffracted photons for τ_{delay}<1 ps
- sub-picosecond formation of ~ 30 Å melt depth
- higher sensitivity to the surface

7.5 keV photons from Thomson source (25 MeV e-beam)

InSb 12.8 degrees off (111)





Ultrafast Structural Dynamics in Crystalline Solids

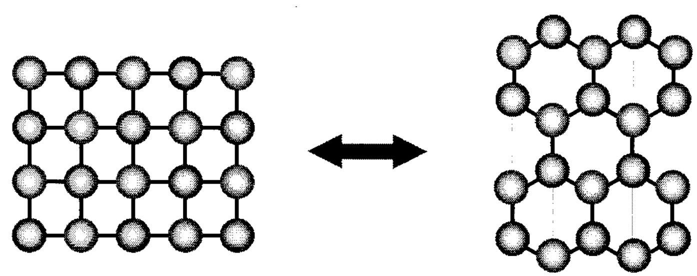
beyond Bragg studies of order disorder transitions

Femtosecond NEXAFS and EXAFS experiments

- amorphous/polycrystalline materials
- surface sensitivity
- changes in coordination and bonding

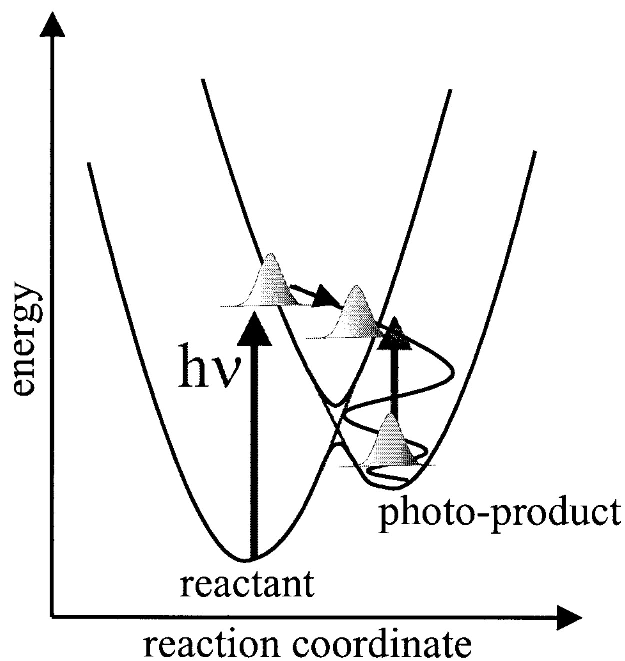
Future experiments on nanocrystal materials

- reversible solid/solid phase transitions

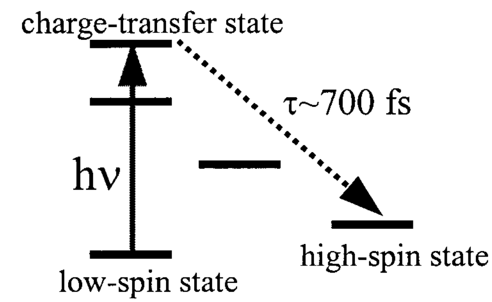


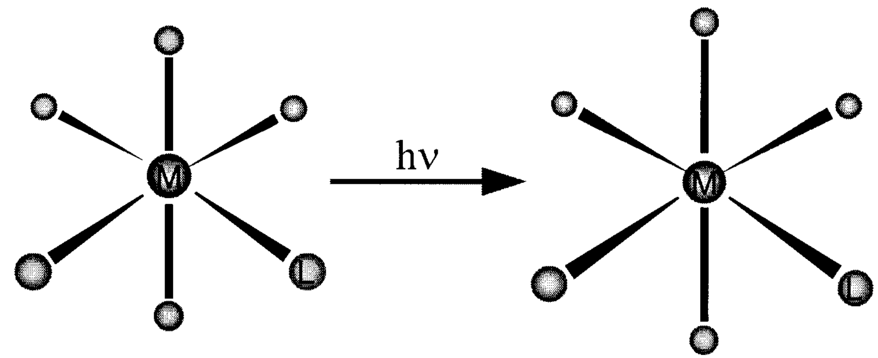
Ultrafast Chemical Reactions

Structural Dynamics of the Transition State



Organo-Metallic Spin-Crossover Molecules





- ~10-15% increase in metal-ligand bond distances
- structural dynamics concomitant with changes in optical and magnetic properties

Structural Dynamics of Ultrafast Biological Processes

Role of protein environment in chemical reaction?

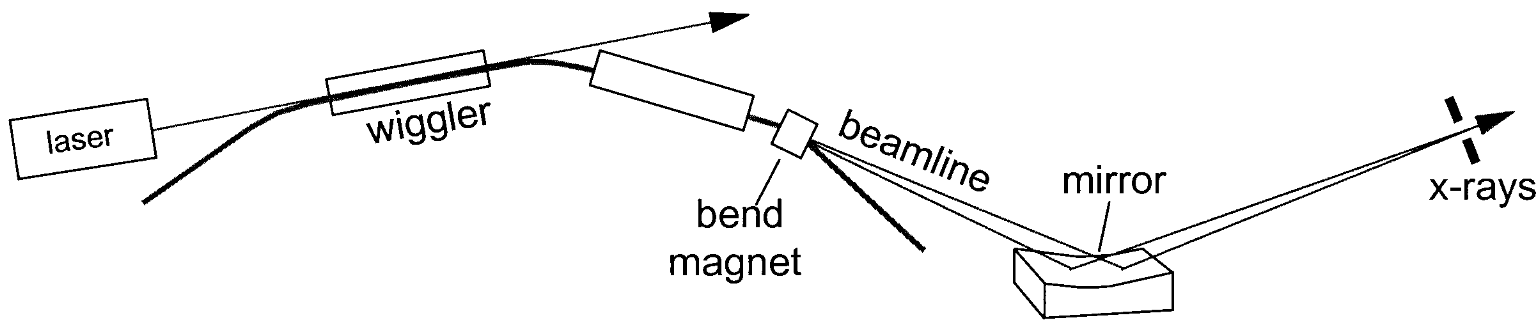
Rhodopsin - photoreceptor for vision
- cis-trans isomerization complete in 200 fs
- vibrationally coherent

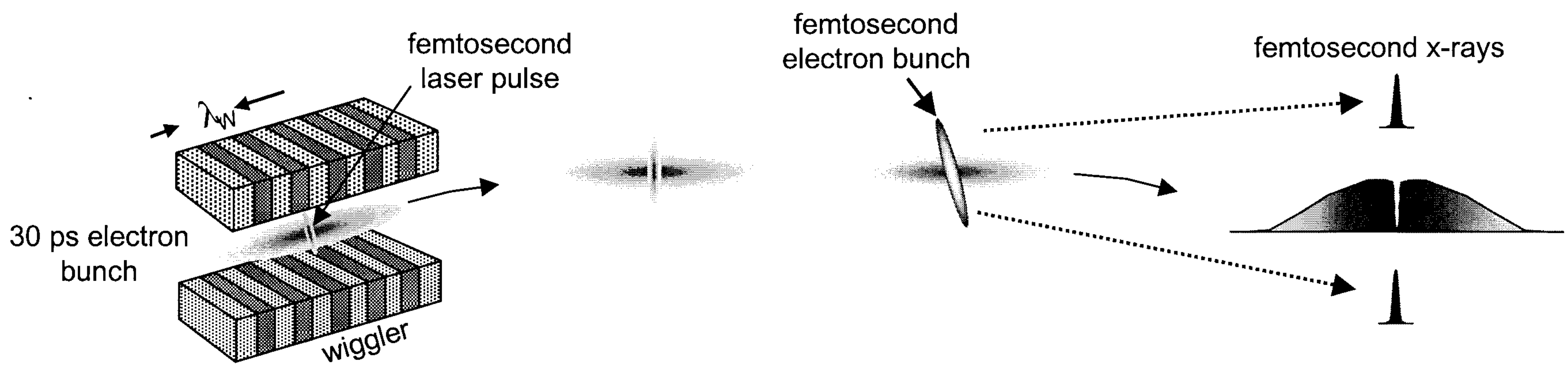
Heme Protein Dynamics
- structural changes associated with ligand binding and dissociation?
- vibrationally coherent photodissociation
  (ultrafast optical spectroscopy)

Previous Time Resolved X-ray Diffraction and NEXAFS Studies

• Bacteriorhodopsin photocycle	msec	DESY
• CO-myoglobin recombination	nsec	ESRF, CHESS
• Xanthopsin - photoactive yellow protein	nsec	ESRF

Generation of Femtosecond X-rays from the ALS





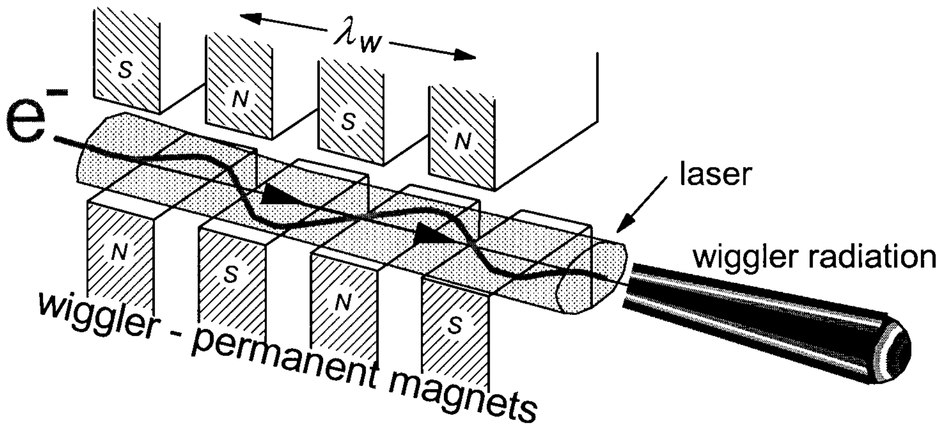
electron-photon interaction in wiggler

spatial separation dispersive bend

bend magnet beamline

Zholents and Zolotorev, *Physical Review Letters*, **76**, 916,1996.

Energy Modulation in the Wiggler



$$\lambda_{radiated} = \frac{\lambda_w}{2\gamma^2}(1+K^2/2) = \lambda_{laser}$$

resonance condition

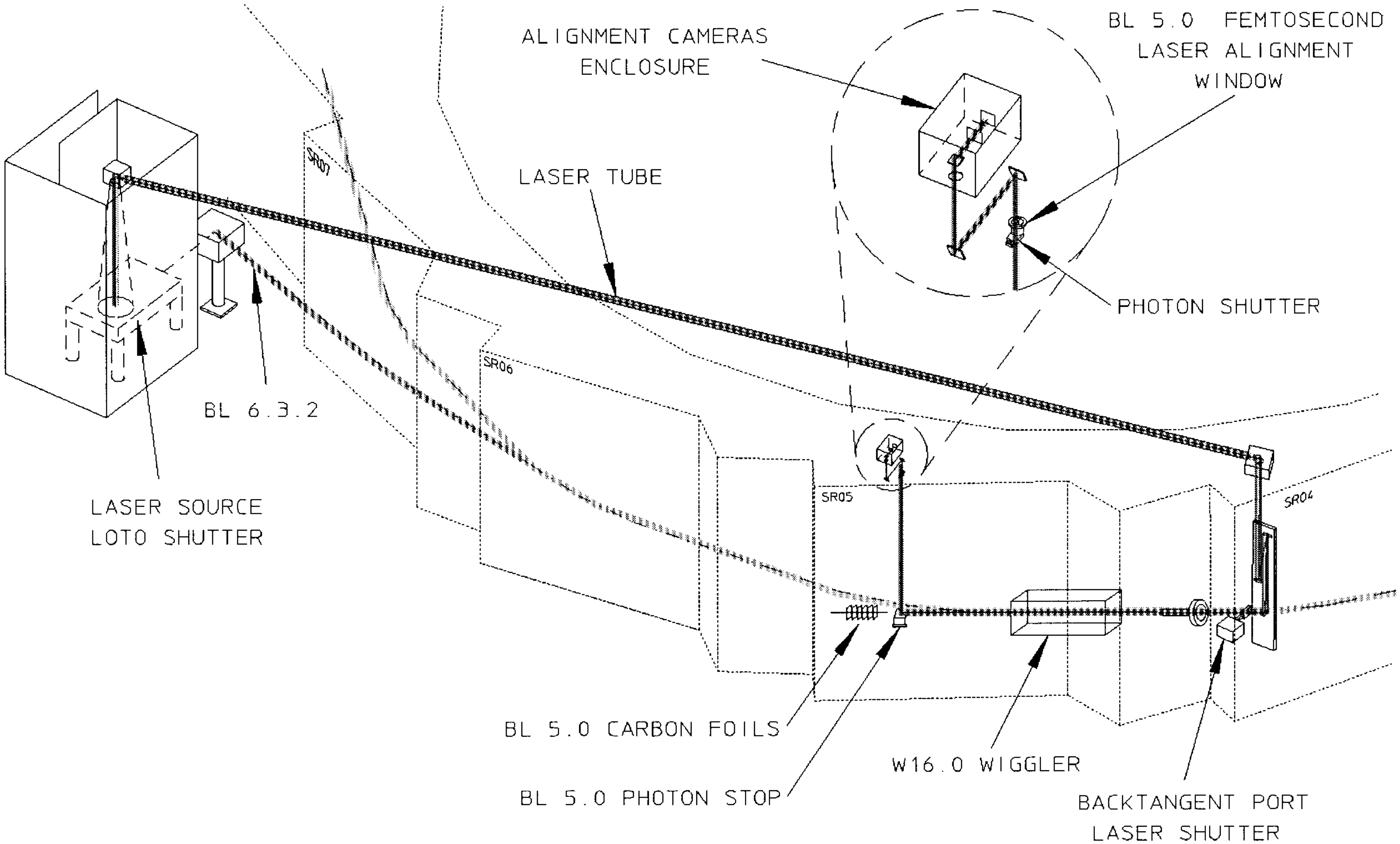
$$K = \frac{eB_o\lambda_W}{2\pi mc}$$

total field energy
$$A \sim \iint \left|E_L(\omega,\vec{r}) + E_R(\omega,\vec{r})\right|^2 dS\,d\omega$$
$$= A_L + A_R + 2\sqrt{A_L A_R \Delta\omega_L / \Delta\omega_R}\cos\phi$$

wiggler radiated energy
$$A_R \cong \pi\alpha\hbar\omega_R \frac{K^2/2}{(1+K^2/2)}$$

amplitude of energy modulation
$$(\Delta E)^2 \cong 4A_L A_R \frac{\Delta\omega_L}{\Delta\omega_R}$$

Synchrotron Beam Slicing - Layout



Laser Gain through Wiggler (FEL Gain)

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

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

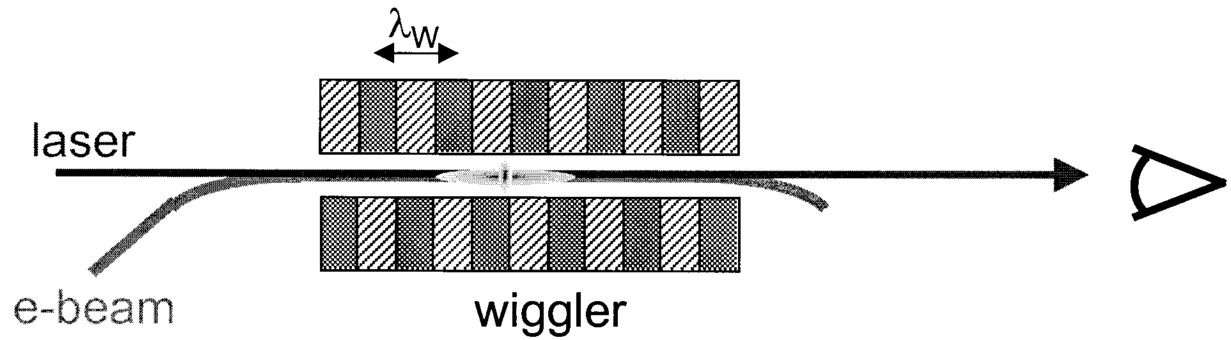
$$e_s(\omega) = e_s(\omega_o) + \frac{\partial e_s(\omega_o)}{\partial \omega}\Delta\omega + \cdots$$

$$\lambda_{radiated} = \frac{\lambda_{wiggler}}{2\gamma^2} \qquad \frac{d\lambda_r}{\lambda_r} = \frac{-2\Delta\gamma}{\gamma} = \frac{-2\Delta E}{E} = \frac{-d\omega_r}{\omega_r} \qquad \frac{d\omega_r}{\omega_r} = \frac{2\Delta \mathrm{E}}{\mathrm{E}} \qquad \Delta \mathrm{E} \propto \mathcal{E}_{laser}$$

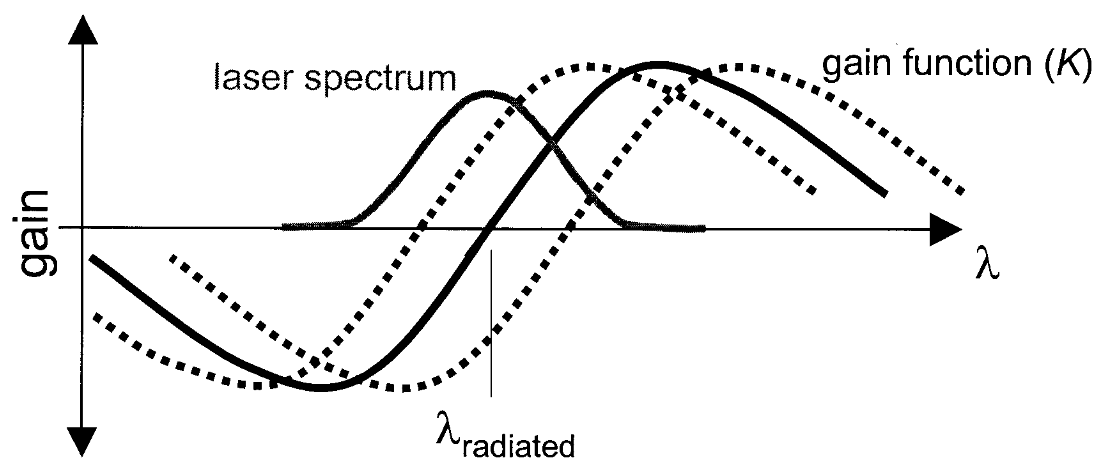
$$\boxed{\mathcal{E}_{total} = \mathcal{E}_{laser}{}^2\left[1 + \underbrace{2\left(\frac{\partial e_s(\omega)}{\partial \omega}\frac{2\omega}{\mathrm{E}}\xi\right)}_{gain}\right] + e_s(\omega)^2 + 2e_s(\omega_o)\mathcal{E}_{laser}}$$

$$gain = \frac{(2\pi M)^2}{\gamma}\frac{\pi}{2}\frac{I_{peak}}{I_A}\frac{d}{dx}\left(\frac{\sin^2(x)}{x^2}\right)\eta \qquad x = \frac{\pi M(\omega-\omega_o)}{\omega_o}$$

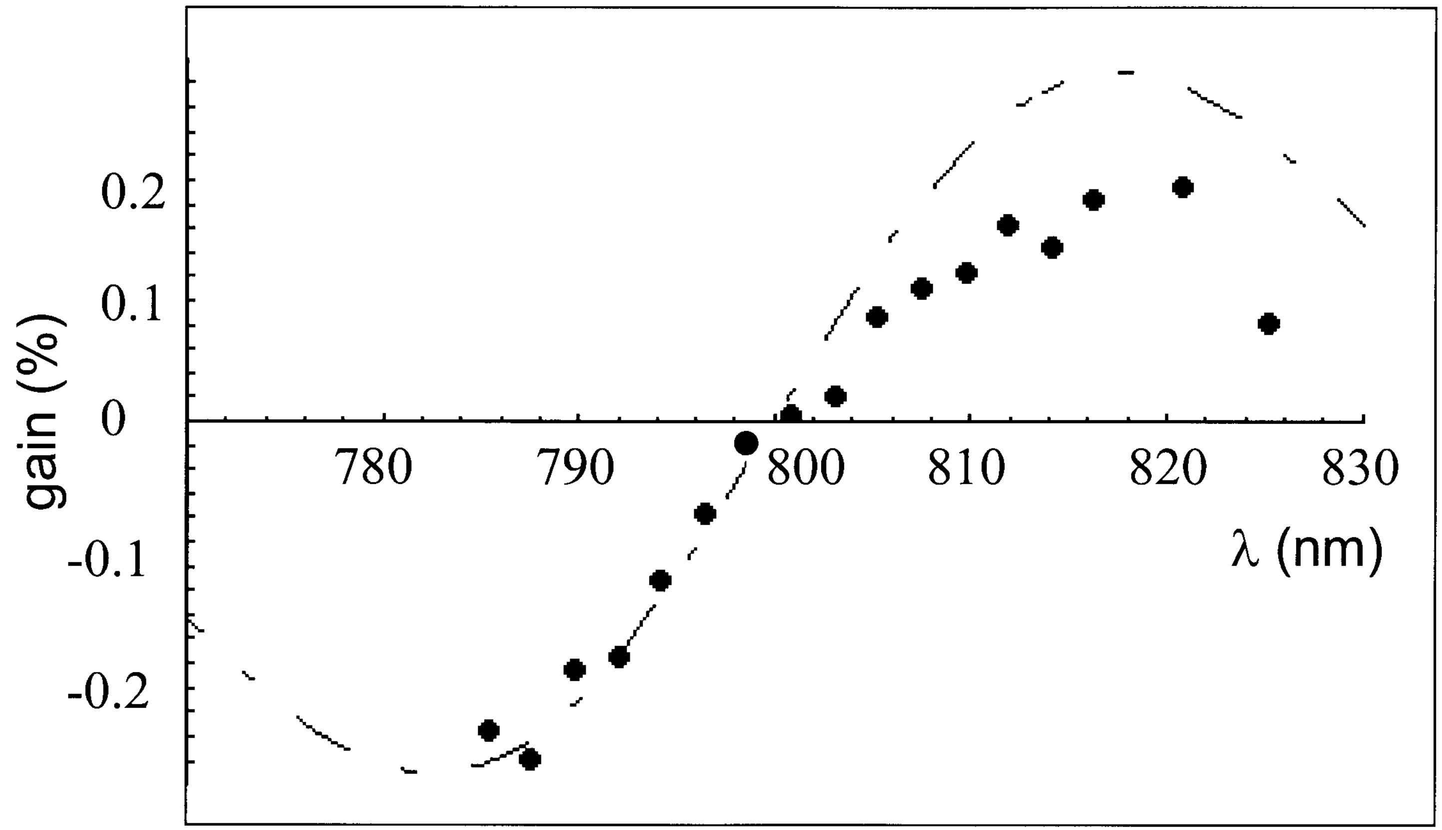
Measurement of Laser Gain through Wiggler



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

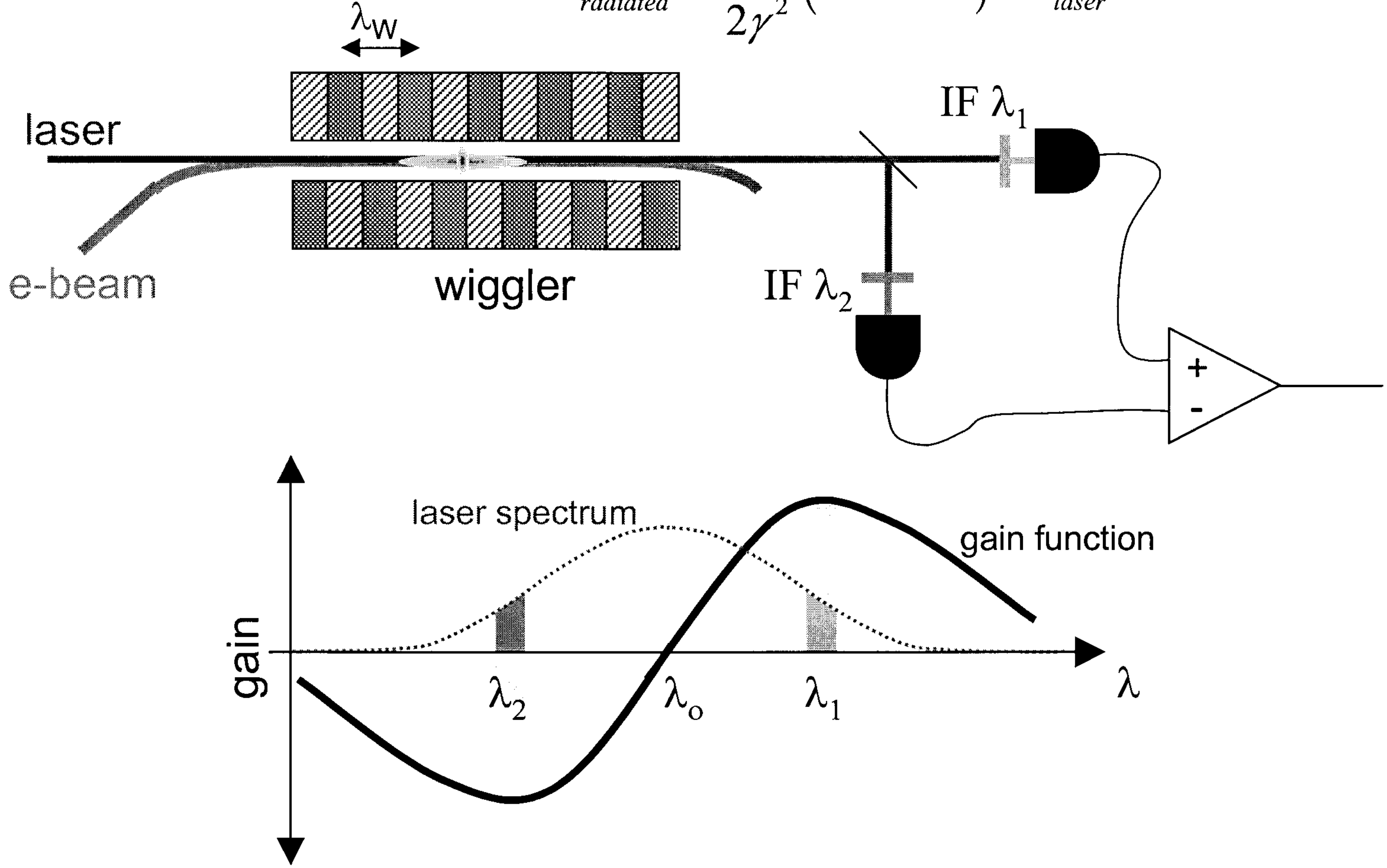


Laser Gain Through Wiggler (FEL Gain)

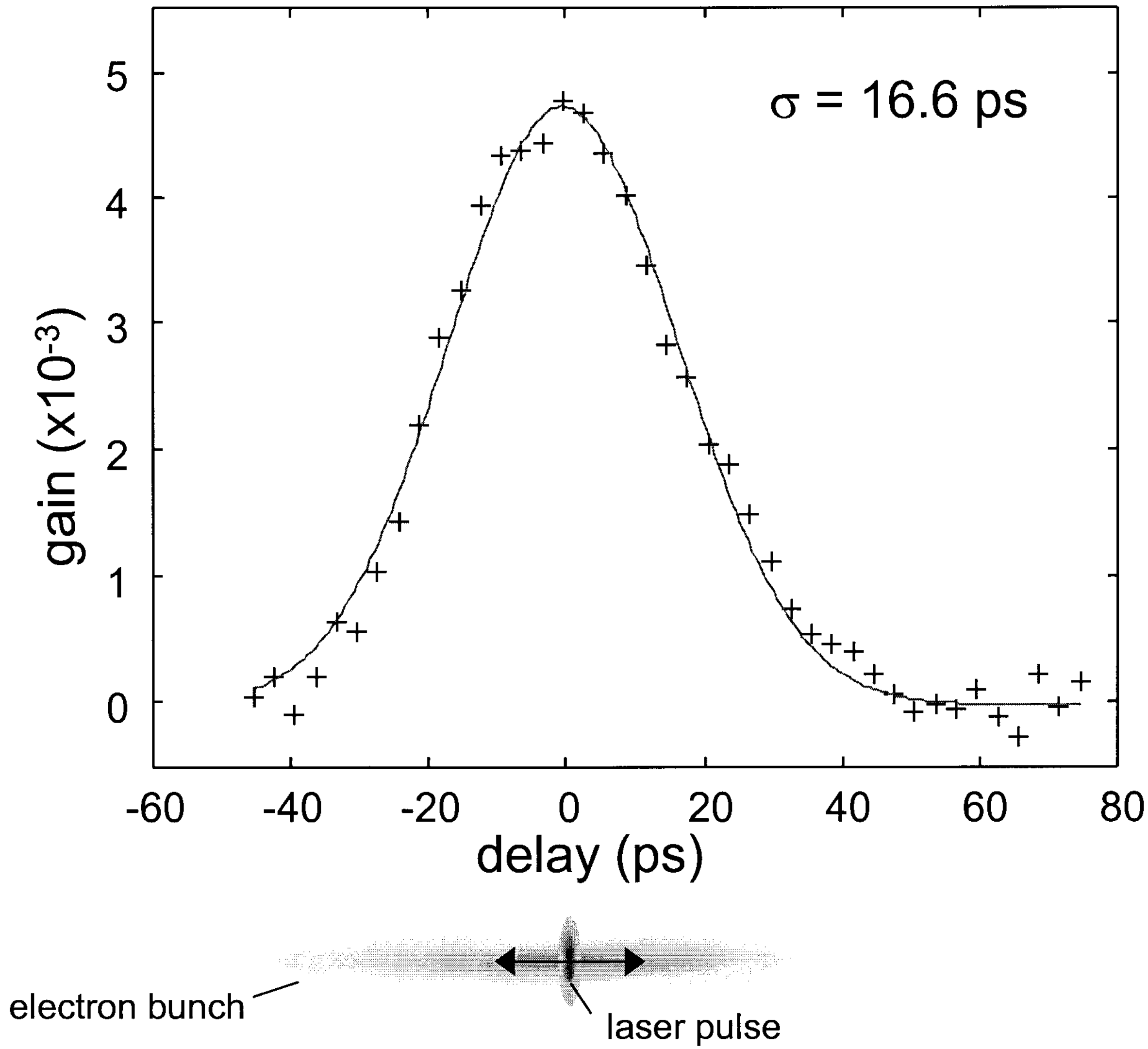


Measurement of Laser Gain through Wiggler

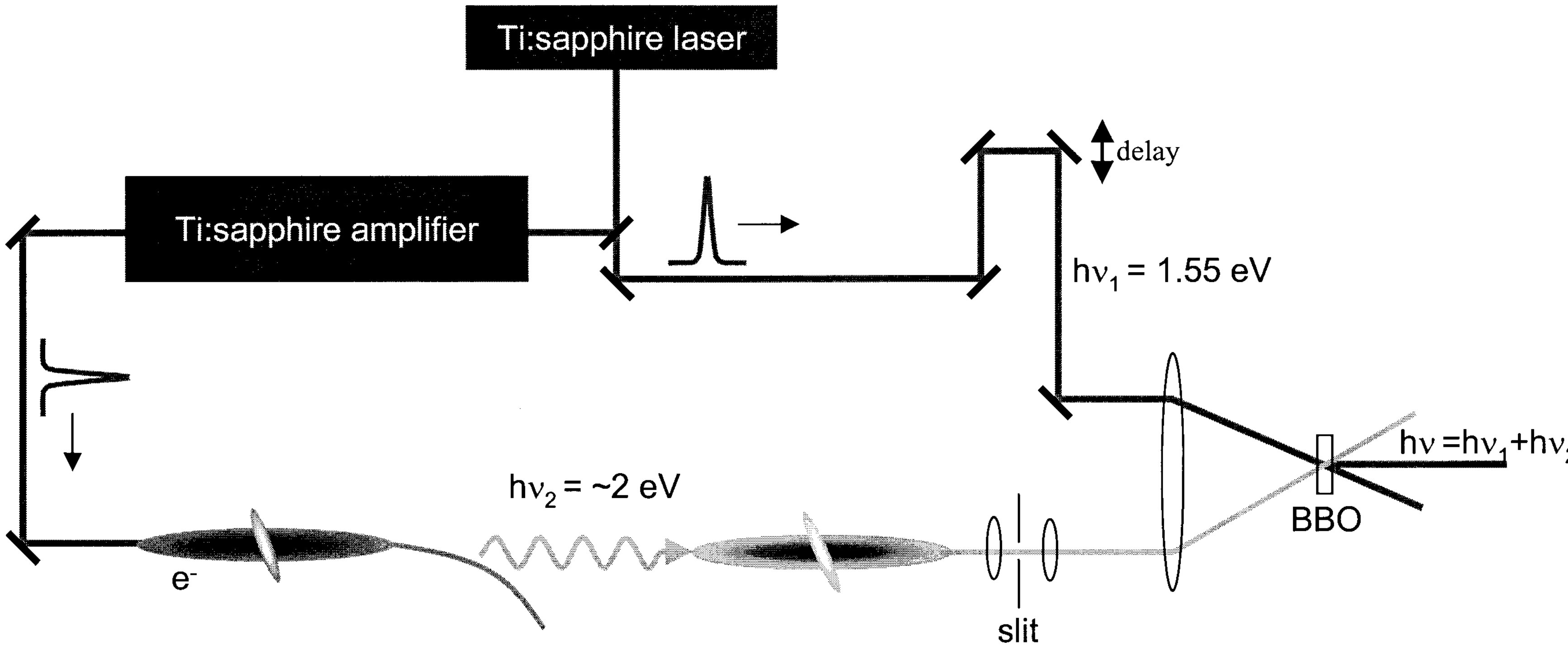
$$\lambda_{radiated} = \frac{\lambda_W}{2\gamma^2}\left(1 + K^2/2\right) = \lambda_{laser}$$



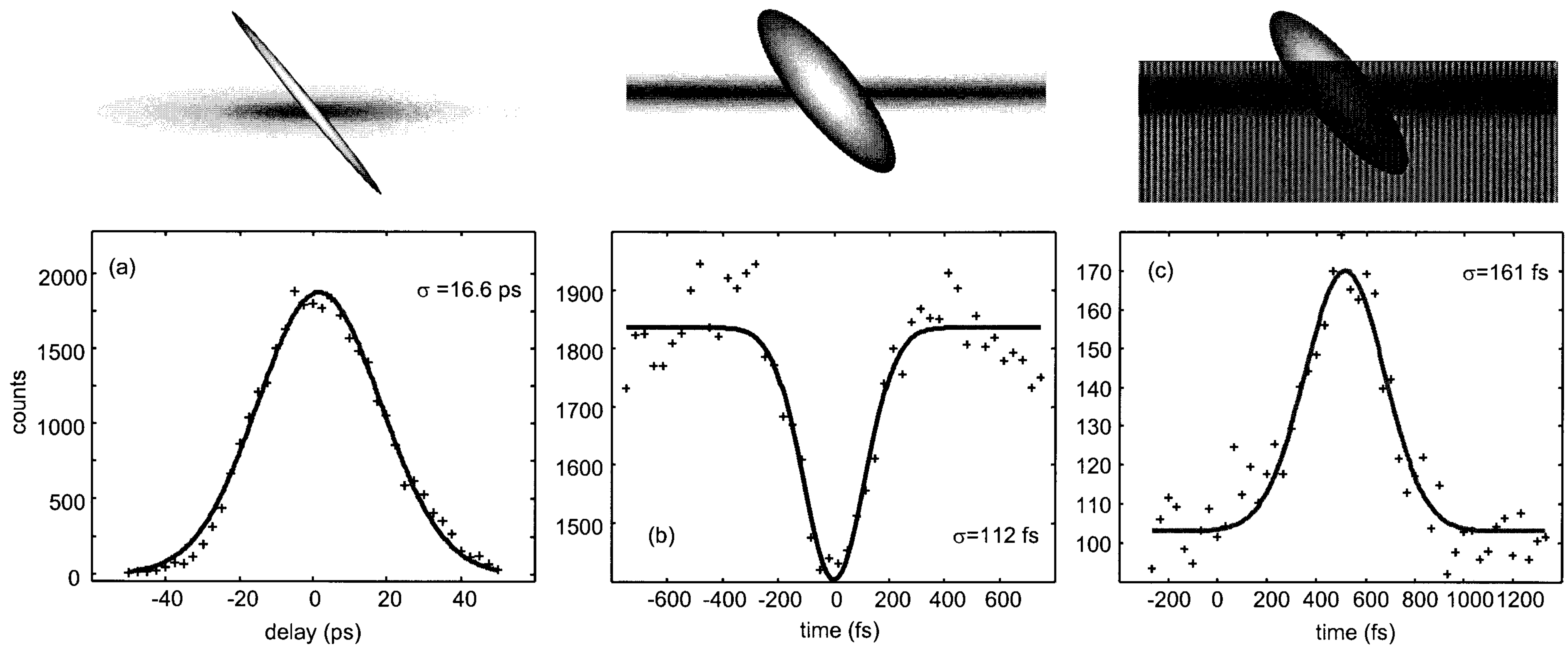
Laser Gain through Wiggler (FEL gain)



Measurement of Femtosecond Synchrotron Pulses via Frequency Upconversion



Correlation Measurements of Visible Femtosecond Synchrotron Pulses



Beyond Proof-of-Principle Experiments

BERKELEY LAB

Femtosecond X-ray Beamline

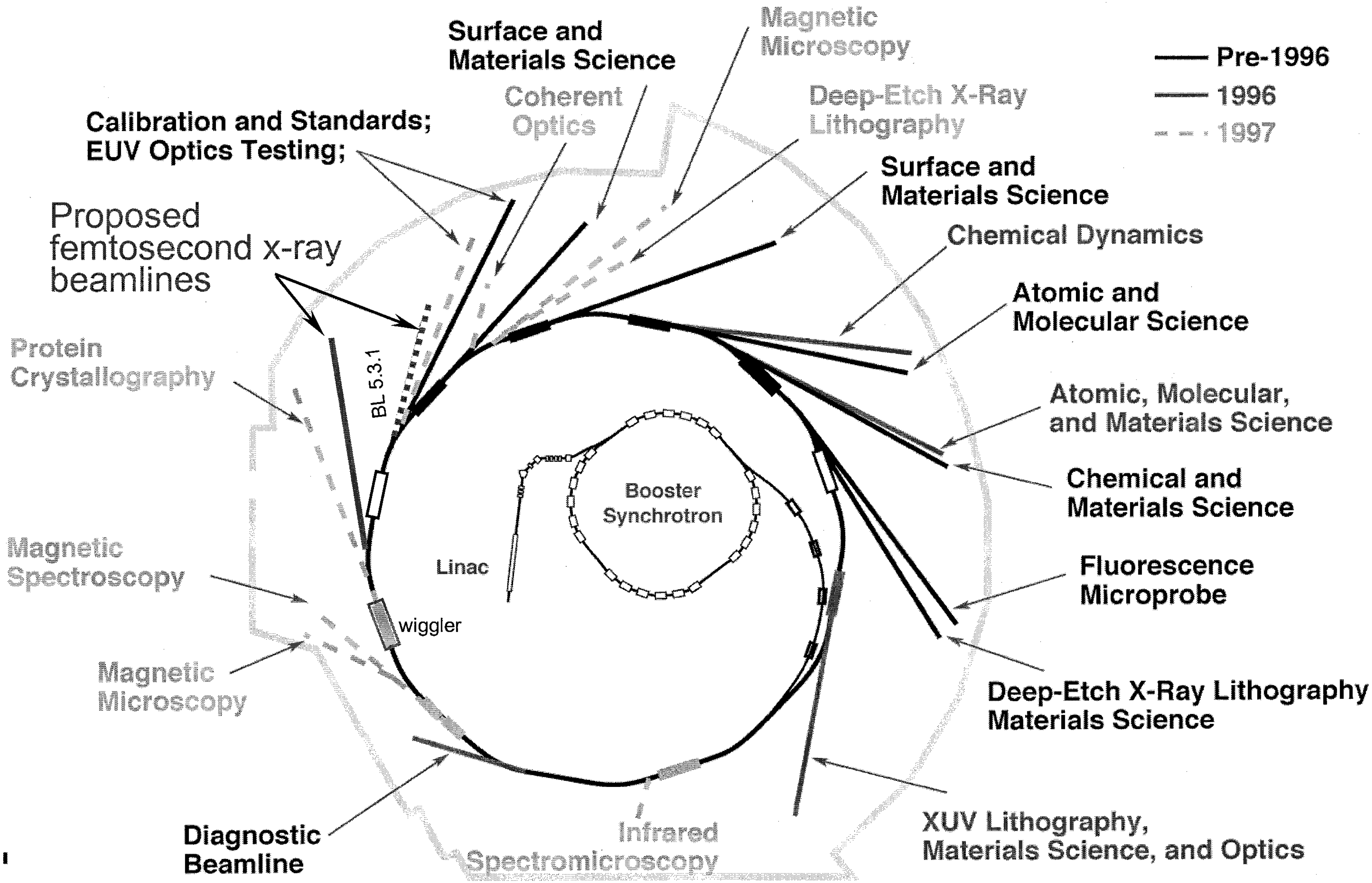
(1) Beamline proximity to wiggler W16 $\rightarrow$ 100 fs x-ray pulses

- storage ring dispersion t_D~200 fs (FWHM) per arc sector

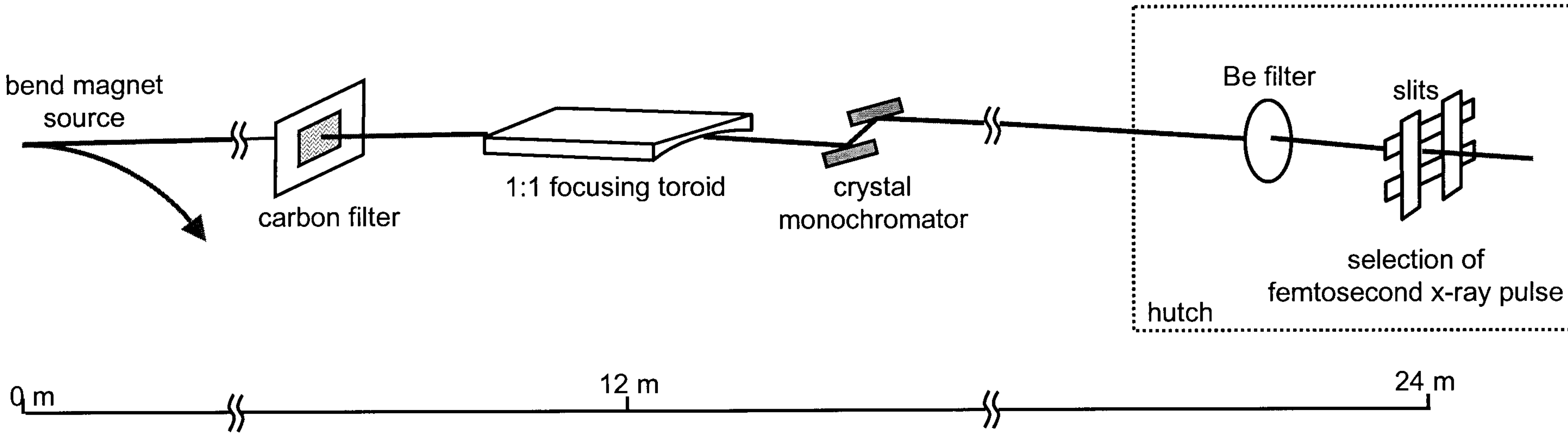
(2) Beamline optics

- minimize non-specular scattering
- maximum wavelength coverage

Advanced Light Source



Femtosecond x-ray Beamline 5.3.1



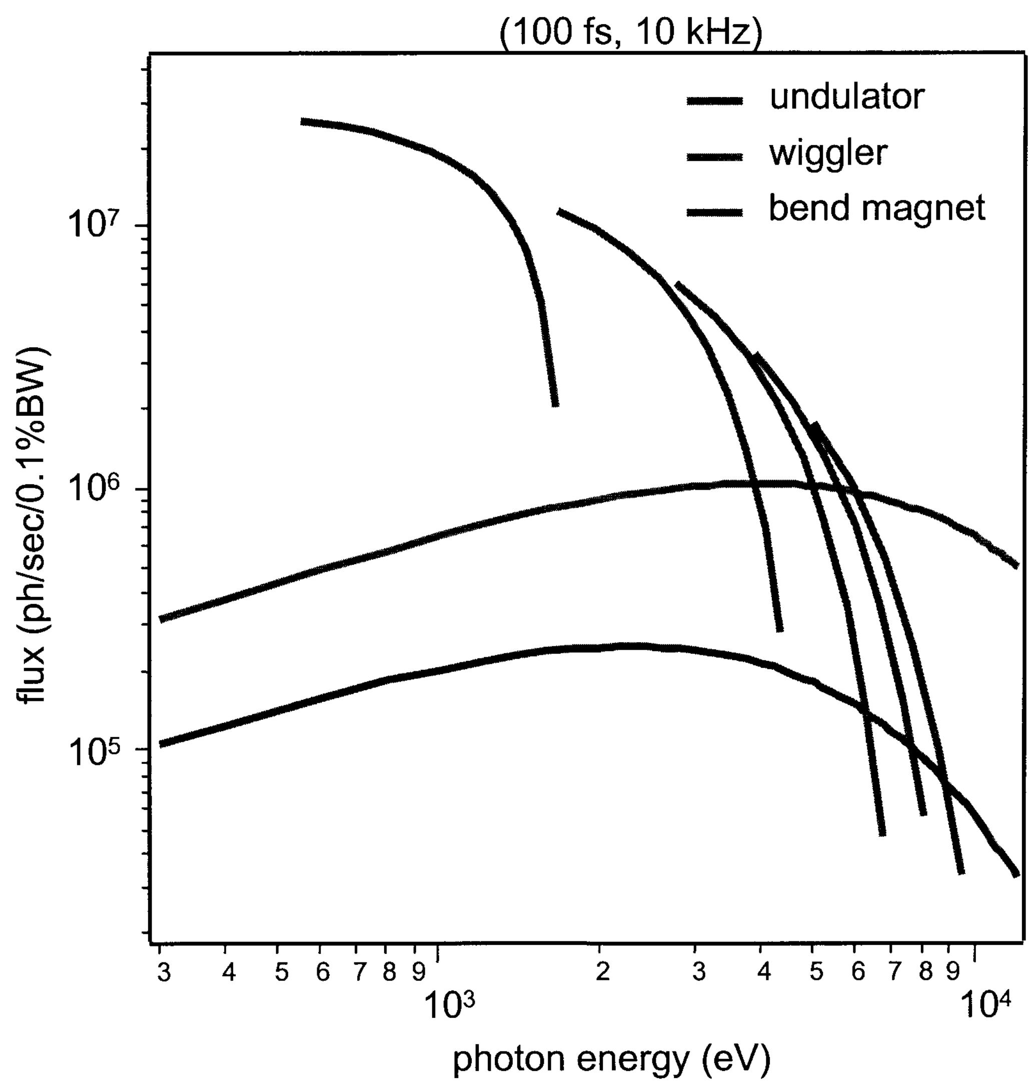
- 1:1 image of bend magnet source
  250 μm (H) x 50 μm (V)
- white beam, 2-12 keV
  (possibility for Laue diffraction)
- flux ~10^{13} ph/sec/0.1% BW (30 ps pulse duration)

flux ~10^{5} ph/sec/0.1% BW
100 femtosecond pulse duration

Femtosecond X-ray Flux from Synchrotron Beamline

- pulse duration $\eta_1 = \tau_{laser} / \tau_{syn} = 3\text{x}10^{-3}$
  (100 fs) (30 ps)
- repetition rate $\eta_2 = f_{laser} / f_{syn} = 2\text{x}10^{-5}$
  (10 kHz) (500 MHz)
- collection efficiency $\eta_3 \cong 0.2$

overall efficiency $\sim 10^{-8}$



Long Term Plans

I. Insertion Device Beamline for Femtosecond X-ray Science

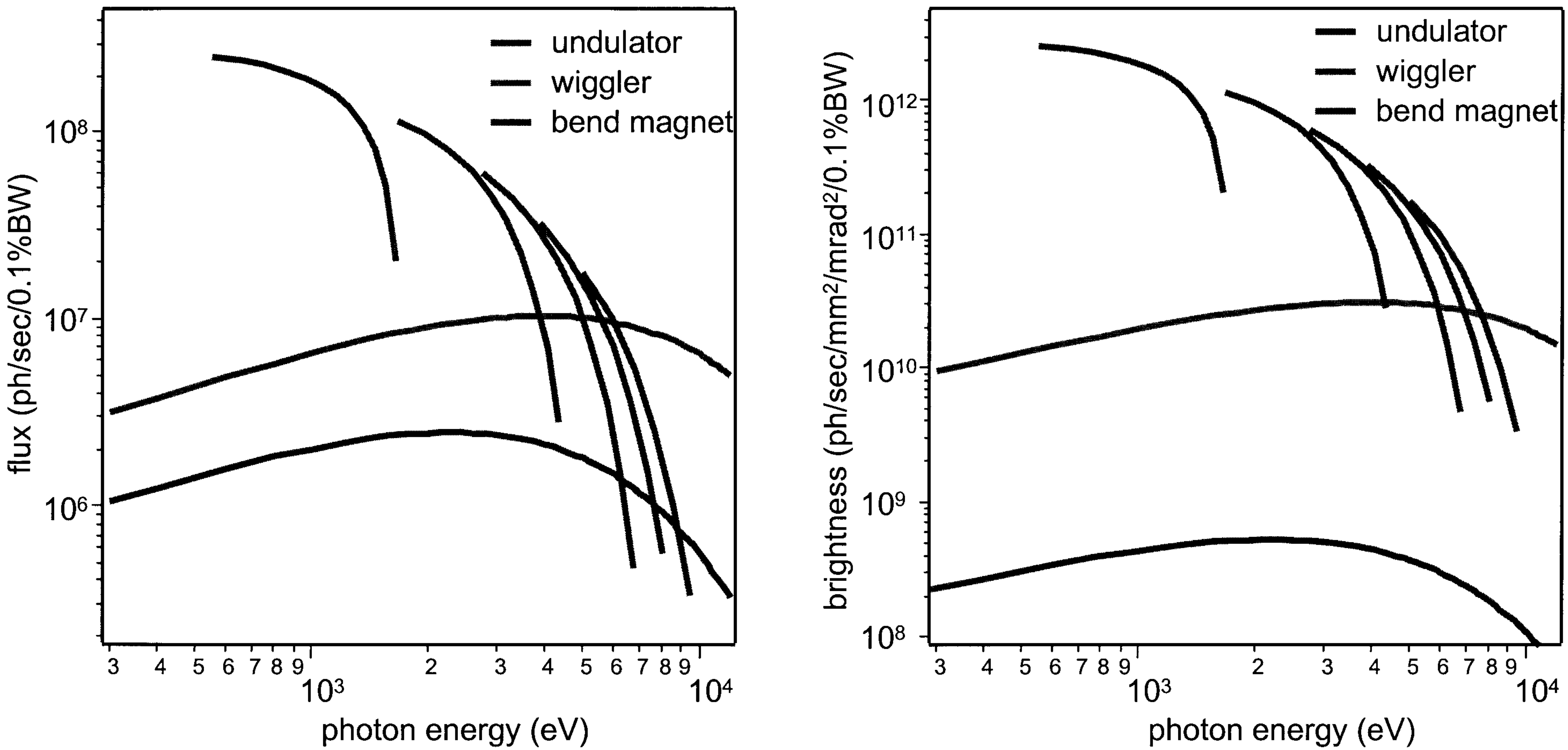
- **10 x increase in flux**
- in-vacuum undulator highest possible flux and brightness 1-8 keV
  - compatible with femtosecond slicing source
  - vertical dispersion bump in storage ring lattice
- location - sector 6, proximity to existing wiggler, presently empty

II. Laser: average power/repetition rate

- **10 x increase in flux**
- 100 W (1 mJ per pulse, 100 kHz)

Femtosecond X-ray Flux

(100 fs, 100 kHz)



Summary

Generation of femtosecond pulses from the Advanced Light Source

- slicing of relativistic e-beam on 100 fs time scale
  co-propagation of e-beam and optical pulse through wiggler
- ~300 femtosecond pulses - visible radiation from bend magnet (BL 6.3.2)
  spectral range from near infrared to x-ray

New beamline 5.3.1 for ultrafast x-ray spectroscopy

- 10^5 photons/sec/0.1% BW (10 kHz laser repetition rate)
- ~100 femtosecond pulse duration, 2-12 keV

In-vacuum insertion device beamline in sector 6

- 10^6-10^7 photons/sec/0.1% BW (10 kHz laser repetition rate)

Time-resolved Measurements of Structural Dynamics in Condensed Matter