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

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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

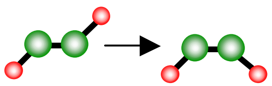
Fundamental Time Scales in Condensed Matter

Atomic Structural Dynamics

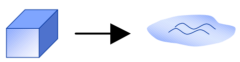
fundamental time scale for atomic motion
vibrational period: T_{vib} ~ 100 fs

(femto – 10^{-15})

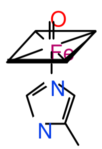
- ultrafast chemical reactions



- ultrafast phase transitions



- surface dynamics
- ultrafast biological processes



Electronic Structural Dynamics

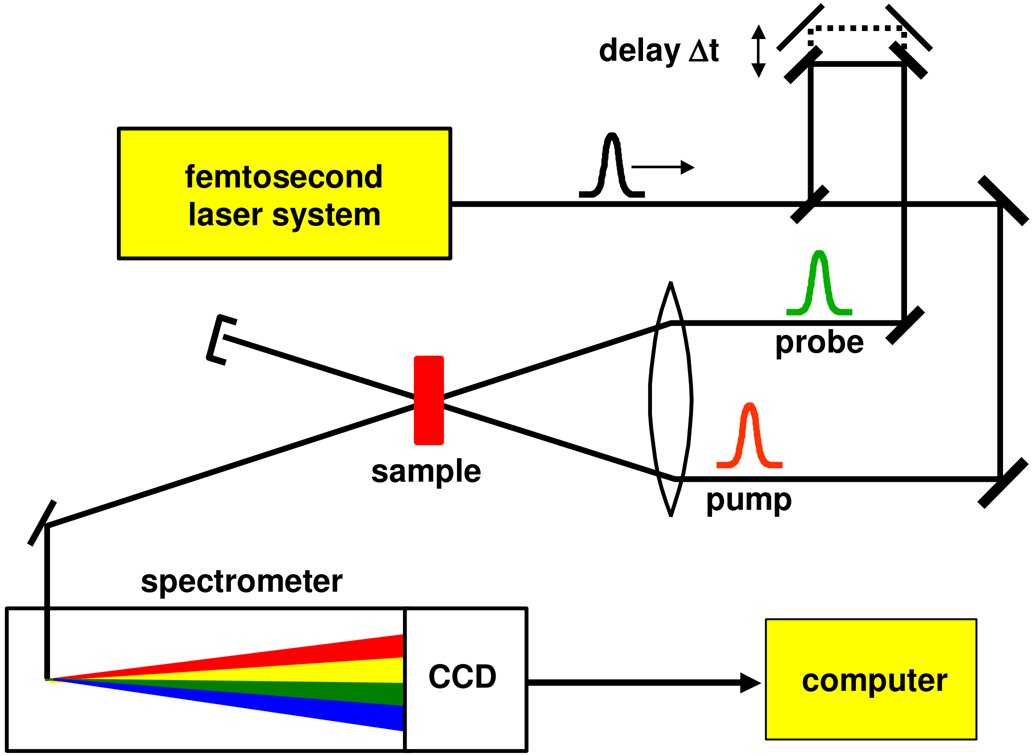
fundamental time scales for electron dynamics
electron-phonon interaction times ~ 1 ps
e-e scattering times ~10 fs
correlation time ~100 attoseconds (a/V_{Fermi})

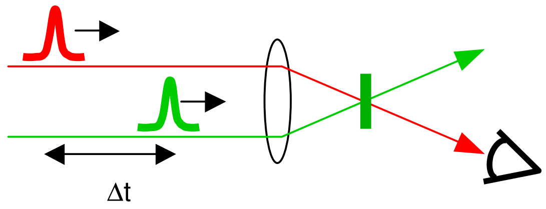
(atto – 10^{-18})

- charge transfer
- electronic phase transitions
- correlated electron systems
  - charge/orbital ordering
  - CMR
  - high T_c superconductivity

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

Femtosecond Spectroscopy





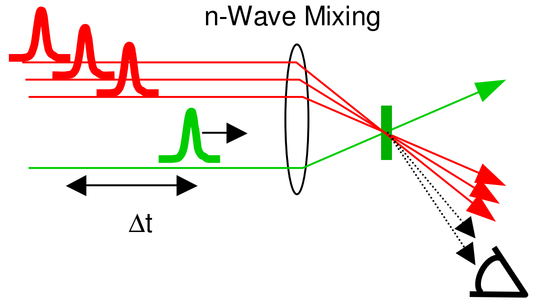
- transmission
- reflection
- photoemission

} $\sim I_{probe}(\Delta t)$

- non-linear (probe) $\sim[I_{probe}(\Delta t)]^n$

  harmonic $\omega_1 \pm \omega_2$

n-Wave Mixing

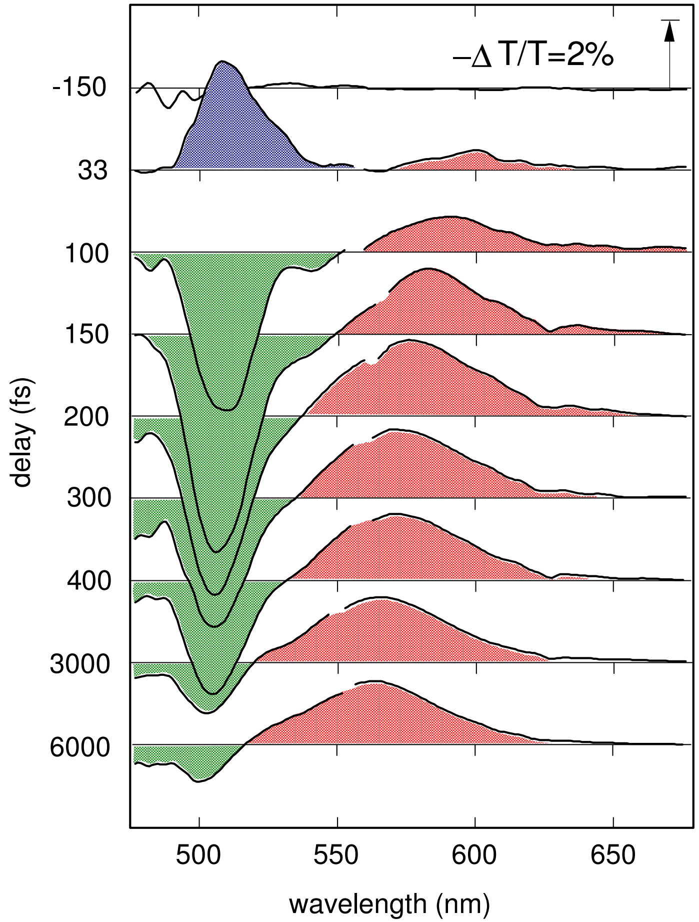


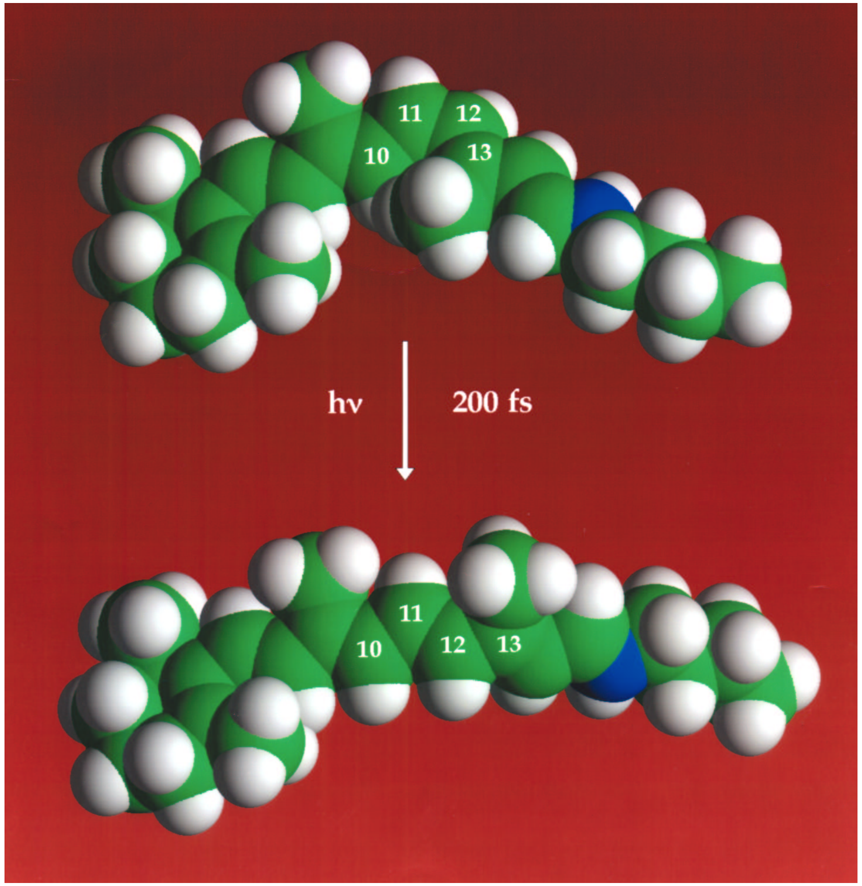
- transient four-wave mixing
- photon echo

Structural Dynamics of Ultrafast Biological Processes

Rhodopsin - photoreceptor for vision

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





Schoenlein et al. *Science* (1991)
Wang et al. *Science* (1994)

Femtosecond X-ray Science

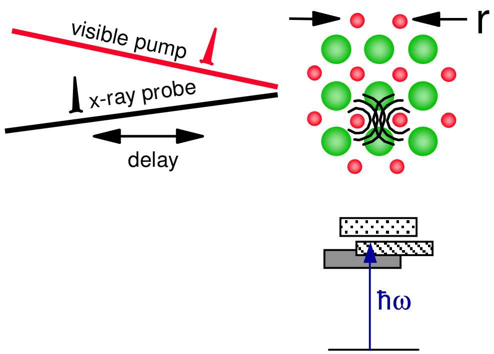
time-resolved x-ray spectroscopy

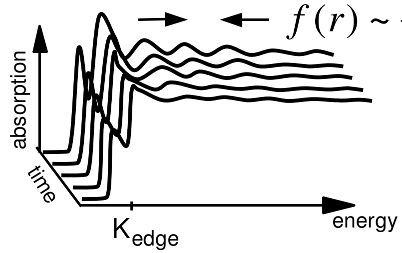
EXAFS – local atomic structure and coordination
(extended x-ray absorption fine structure)

NEXAFS – local electronic structure, bonding geometry, magnetization/dichroism
(near-edge x-ray absorption fine structure)

- element specific
- molecular systems and reactions
- complex/disordered materials

surface EXAFS, μEXAFS ….

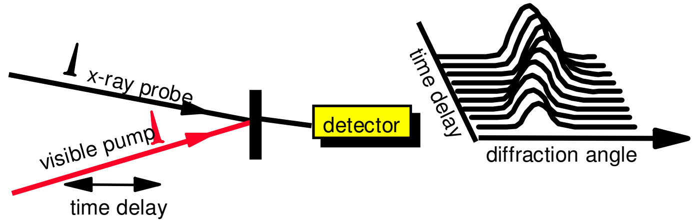


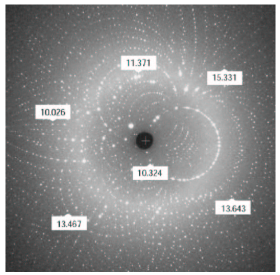


$$f(r) \sim \frac{\sin\left(2kr + \phi(k)\right)}{kr^2}$$

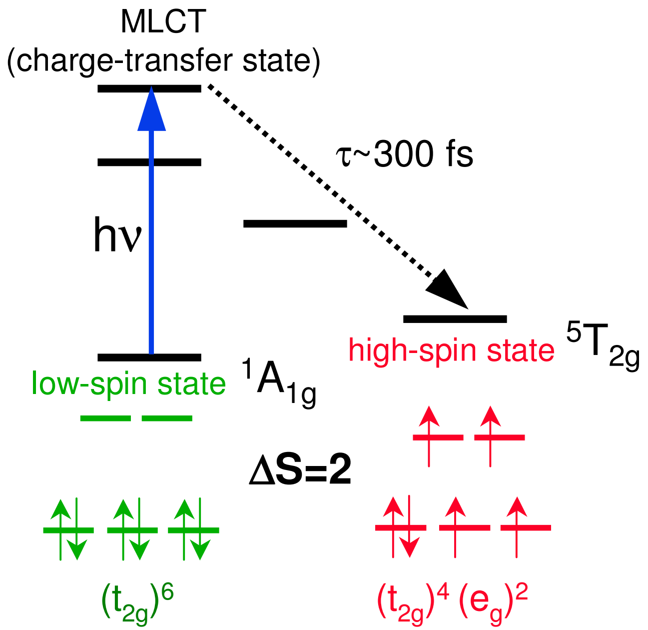
time-resolved x-ray diffraction

atomic structure in systems with long-range order/periodicity
phase transitions, coherent phonons

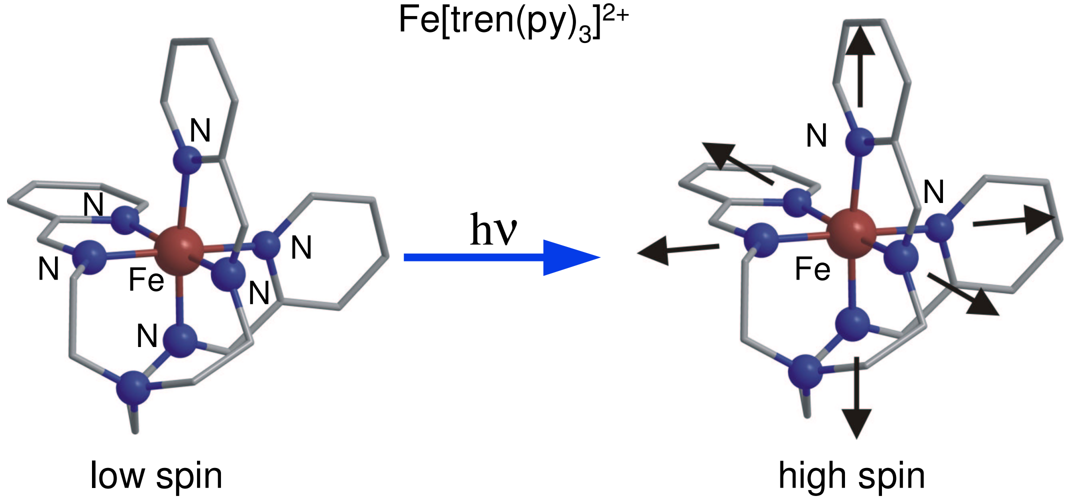




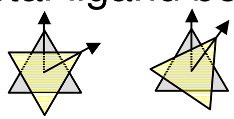
Fe^{II} Spin-Crossover Molecules



$Fe[tren(py)_3]^{2+}$



- ~10-15% increase in metal-ligand bond distances
- trigonal cage distortion?



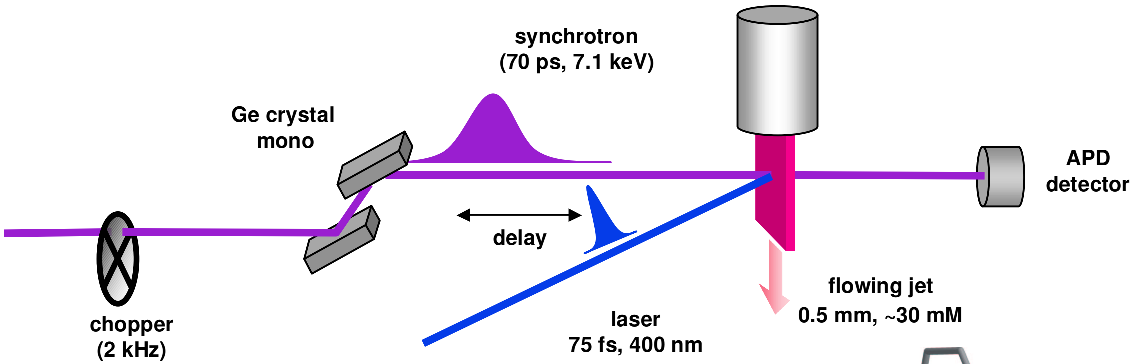
Motivation:

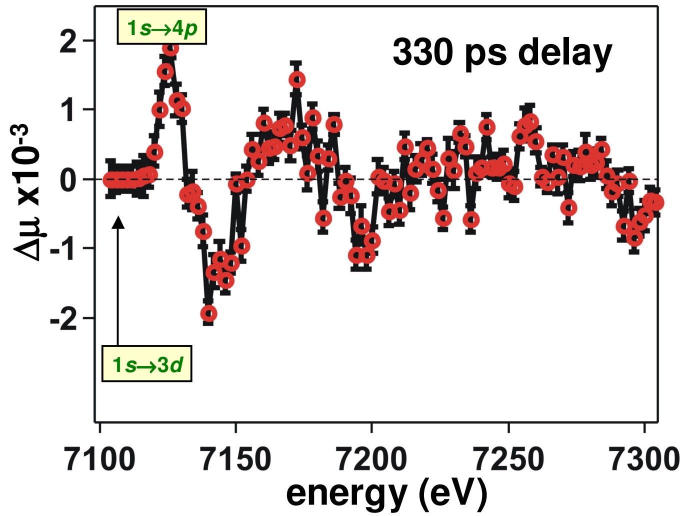
- relationship between structure, electronic, and magnetic properties

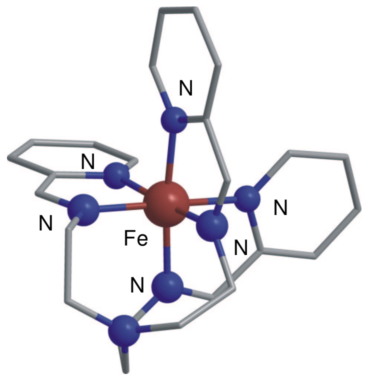
  Do the structural distortions facilitate the spin-crossover reaction?
- electron transfer mechanistic role in biochemical processes (cytochrome P450)
- magnetic and optical storage material

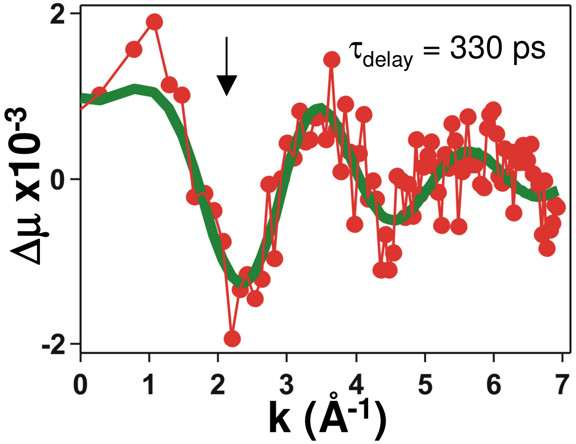
Fe(II) Spin-Crossover X-ray Spectroscopy

Time-resolved XAS - ALS Beamline 5.3.1)









	Reactant $^1A_1$ ($\tau = 0$)	Photoexcited $^5T_2$ ($\tau = 330$ ps)
N	6 ±0.5	6.5 ±1
R (Å)	1.94 ± 0.01	2.15 ±0.03
σ (Å^2)	0.001	0.009

M. Khalil et al., *J. Phys. Chem.* (2006)

X-rays for Ultrafast Structural Dynamics

flux - photons/sec/0.1% BW
brightness - photons/sec/mm^2/$mrad^2$/0.1% BW

Characteristics for Ideal Source

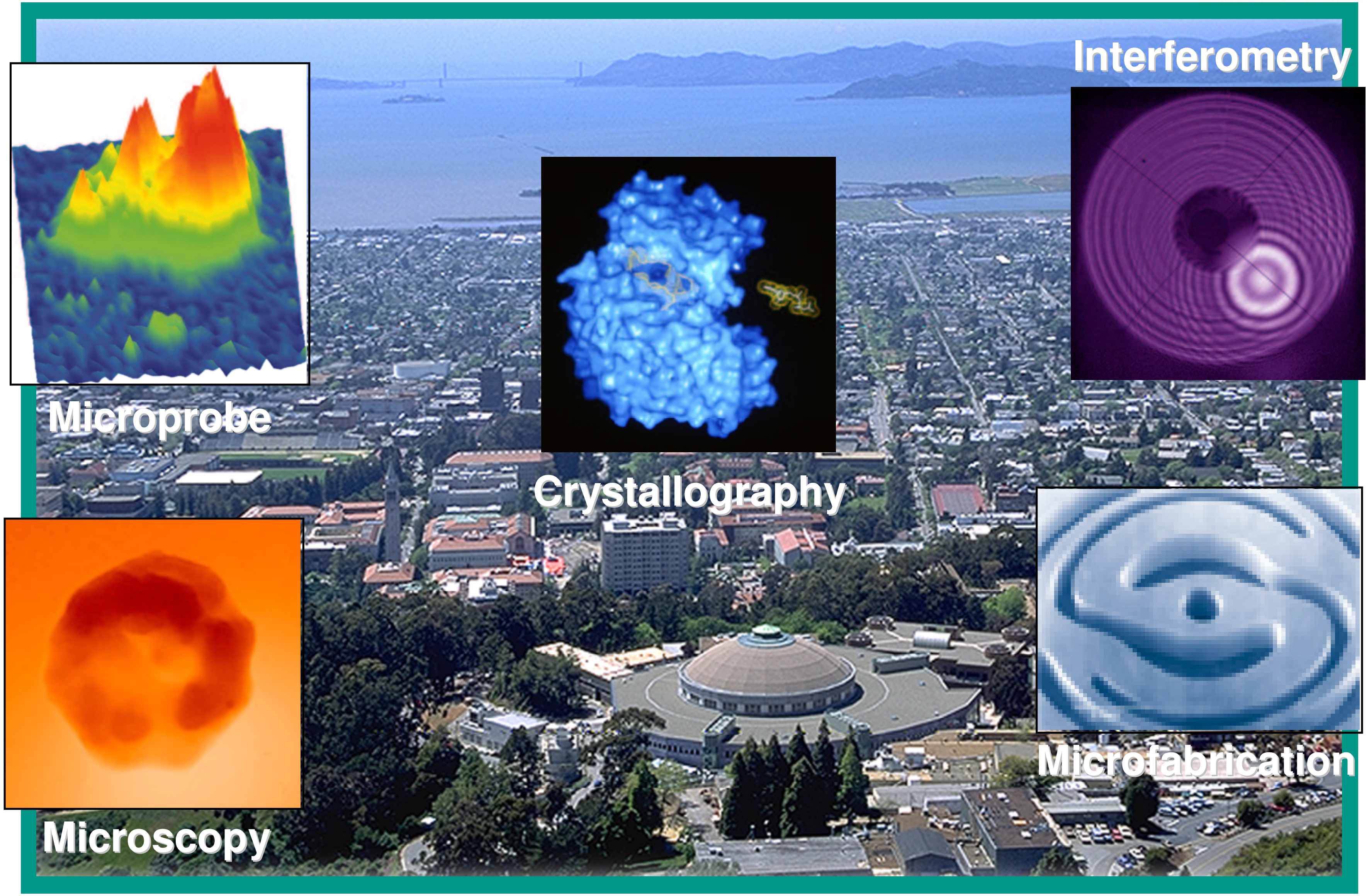
(1) temporal resolution ~ 100 fs
- pulse duration
- synchronization to laser trigger

(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)

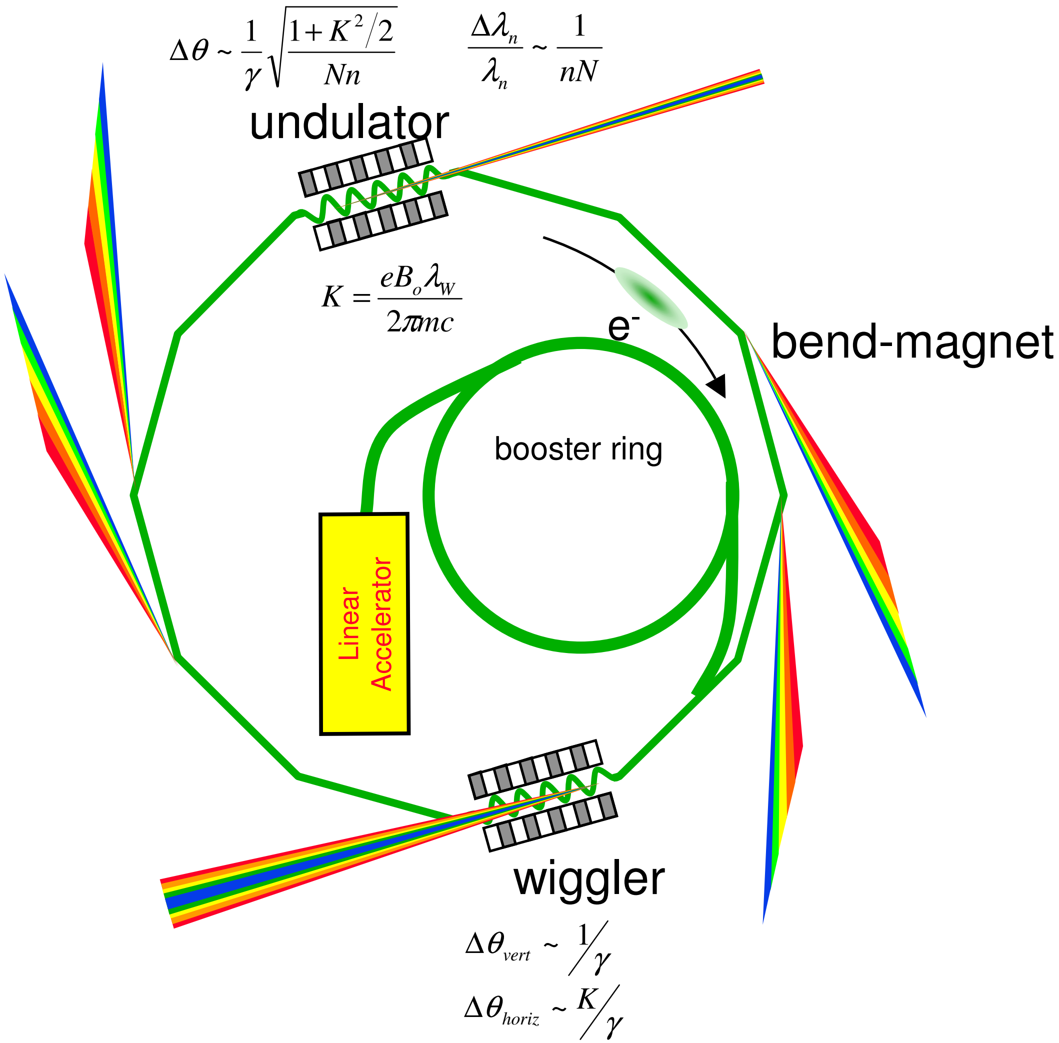
(4) rep. rate: 100 Hz - 10 kHz
- sample recovery or replacement
- signal averaging (high stability)

3rd Generation Synchrotrons - Advanced Light Source



3rd Generation Synchrotrons

Advanced Light Source



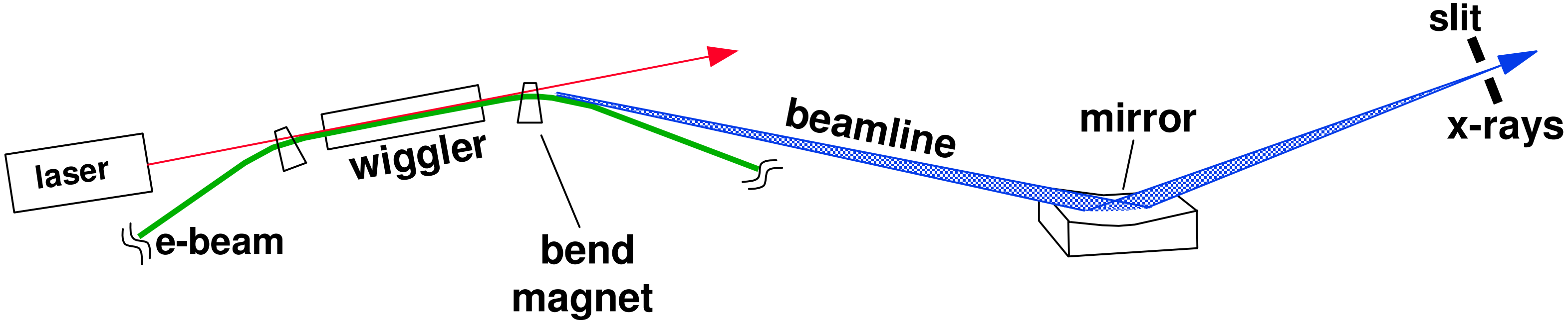
electron storage ring:
- 1.9 GeV
- 500 MHz, 400 mA
- 70 ps bunch length

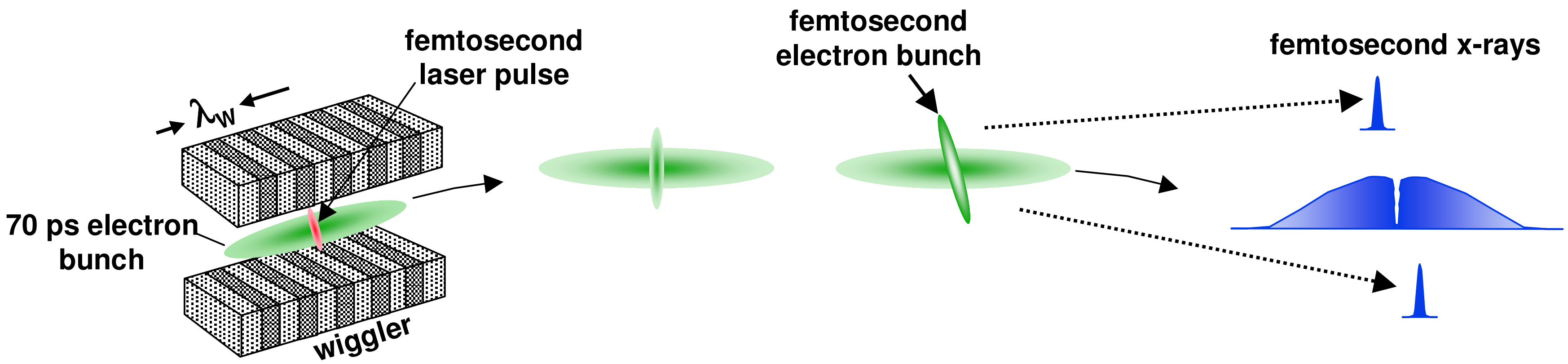
12 straight sections for insertion devices

36 bend magnets

high-brightness source of soft x-rays 1-100 Å

Generation of Femtosecond X-rays from the ALS





electron-photon interaction in wiggler

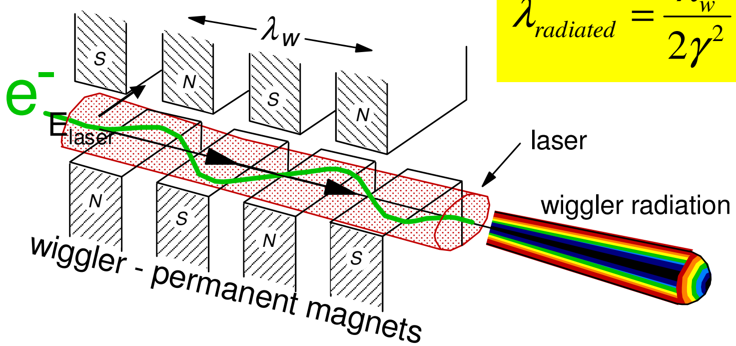
spatial separation dispersive bend

bend magnet beamline

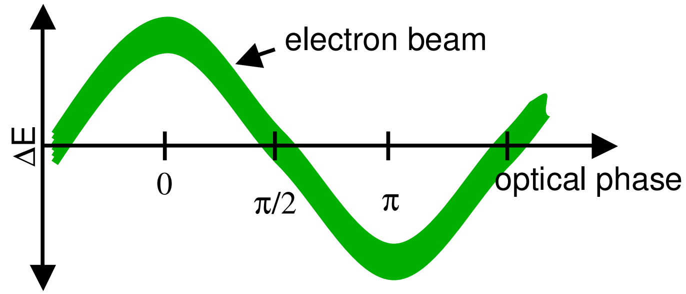
A. Zholents and M. Zolotorev, *Phys. Rev. Lett.*, 76, 916,1996.

Energy Modulation in the Wiggler

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



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



total field energy:

$$A \sim \iint \left| E_L(\omega,\vec{r}) + E_R(\omega,\vec{r}) \right|^2 dS d\omega = A_L + A_R + \underbrace{2\sqrt{A_L A_R \frac{\Delta\omega_L}{\Delta\omega_R}}\cos\phi}_{\Delta E \text{ (energy mod)}}$$

wiggler radiated energy:

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

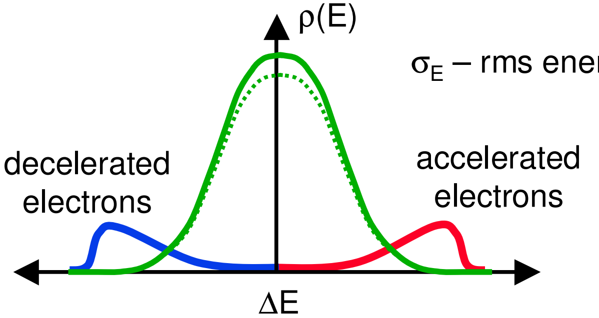
Laser requirements:

$\hbar\omega_L = 1.55$ eV

$\Delta\omega_L = 27$ period wiggler $\Rightarrow$ 36 fs laser pulse
(72 fs)

$A_L = 610\ \mu J$
(780 μJ)

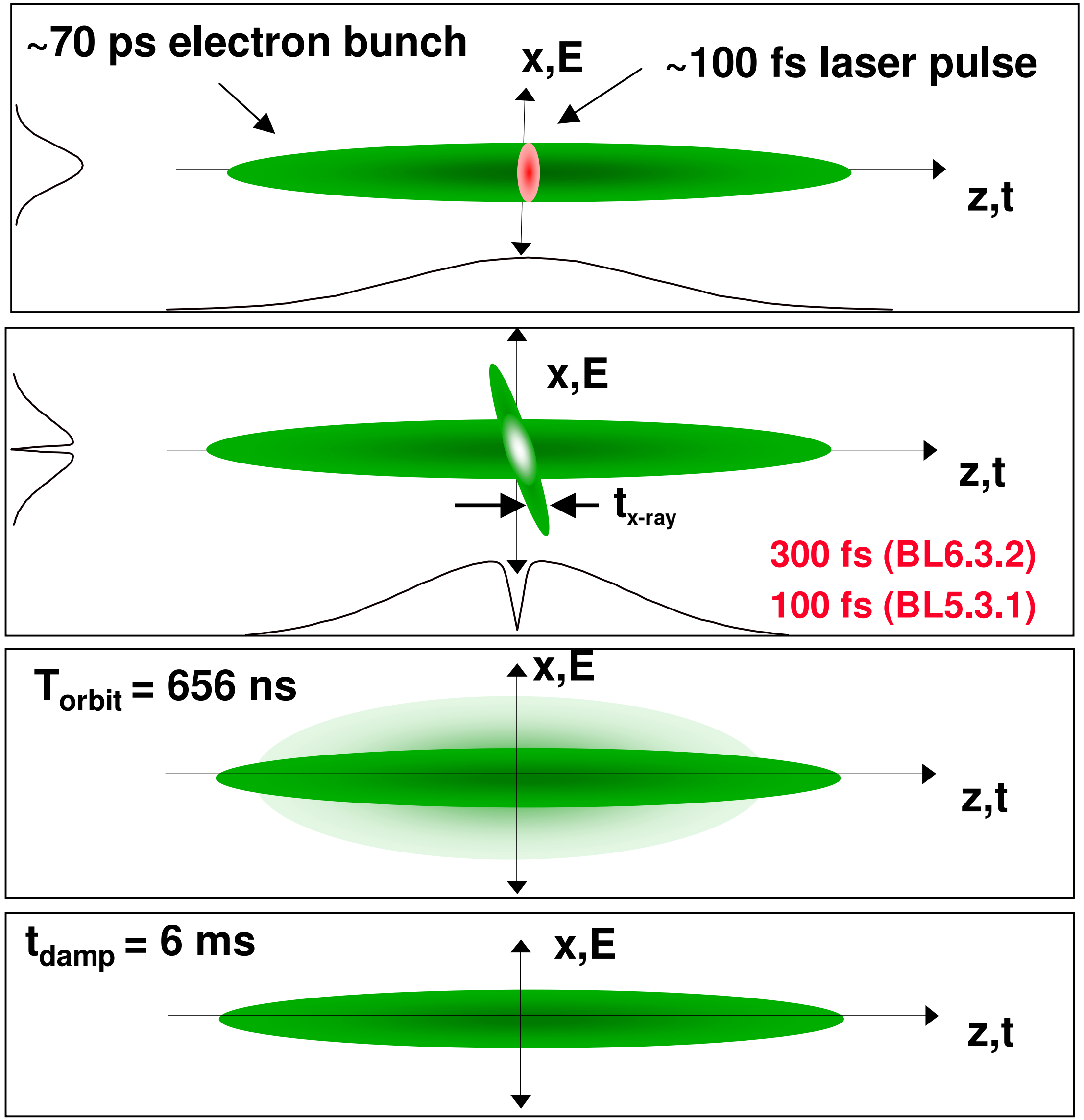
$\Delta E \cong 17$ MeV
$9\sigma_E$



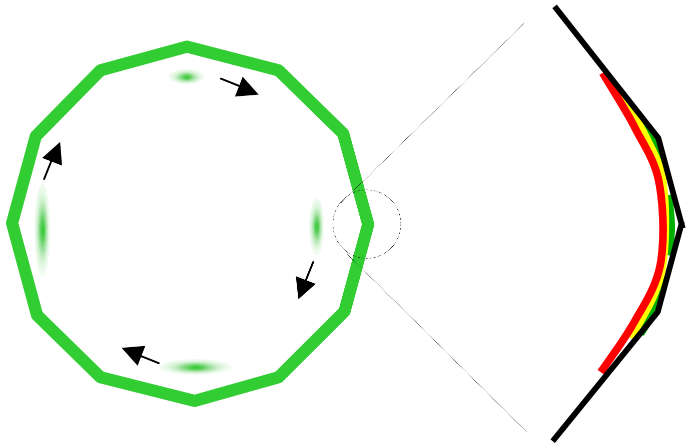
ALS beam energy spread σ_E ~1.9 MeV E_o =1.9 GeV

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

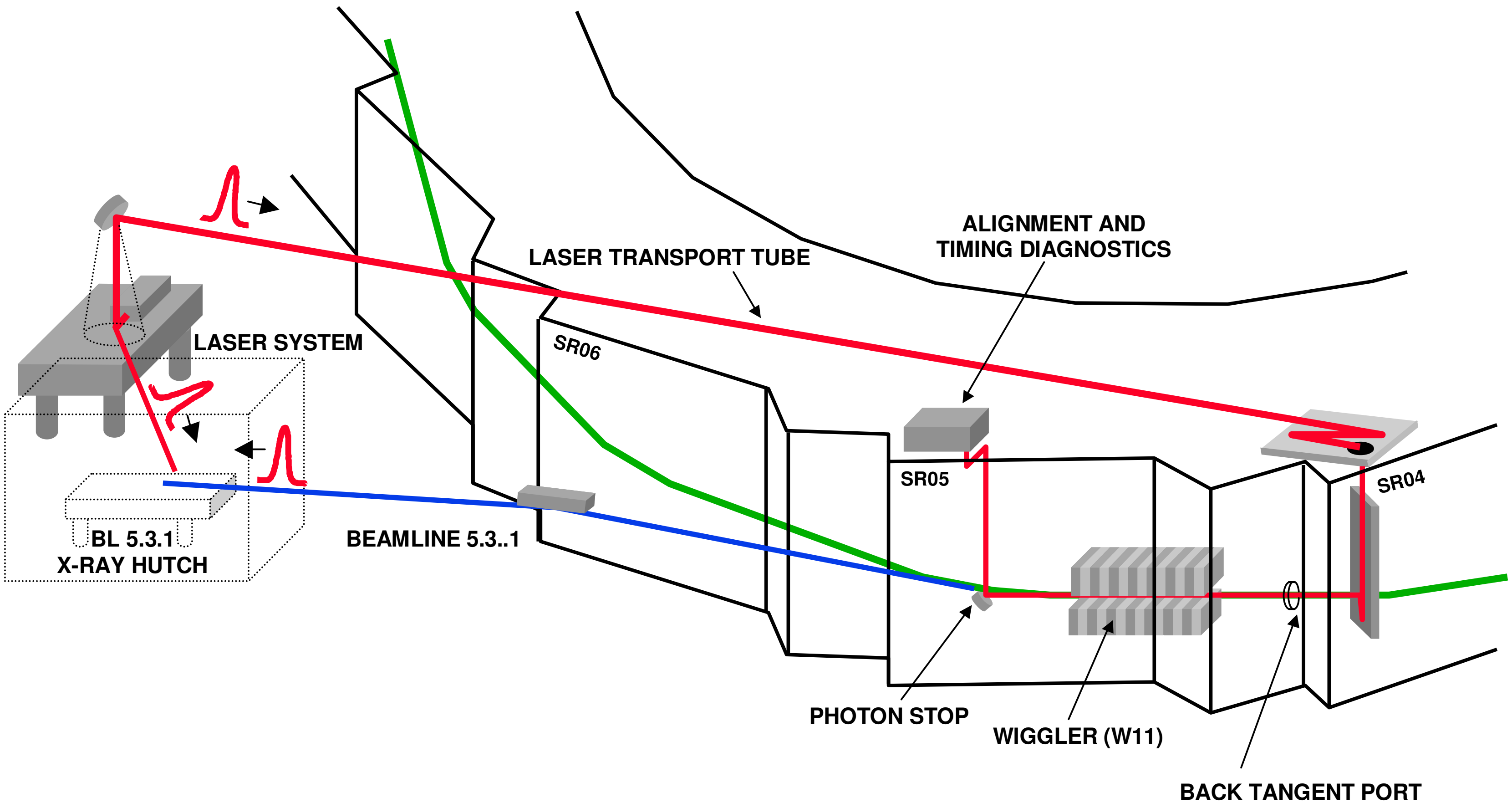


ALS Storage Ring Dispersion



$\sigma_{dispersion}$ ~ 85 fs per full arc
(200 fs FWHM)

Laser Synchrotron Beam Slicing - Layout



Laser Gain through Wiggler (FEL Gain)

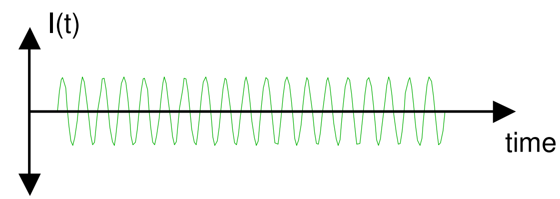
total optical field $\quad \mathcal{E}_{total}(\omega) = \mathcal{E}_{laser}(\omega) + e_s(\omega)$

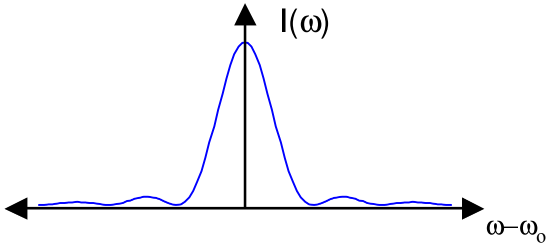
$$\mathcal{E}_{total}(\omega)^2 = \mathcal{E}_{laser}(\omega)^2 + e_s(\omega)^2 + 2\mathcal{E}_{laser}(\omega)e_s(\omega)$$

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

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

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

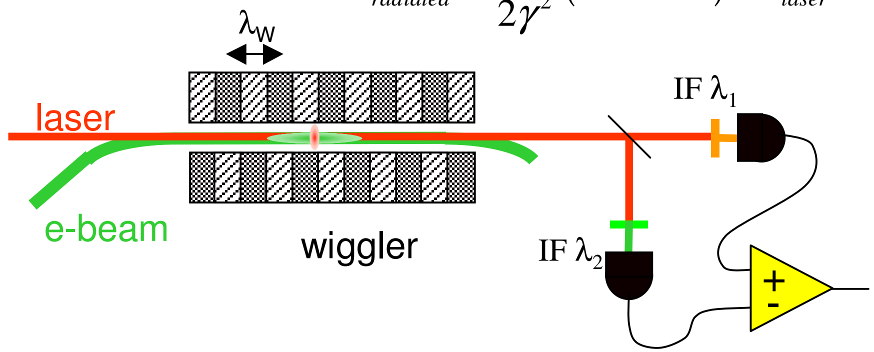


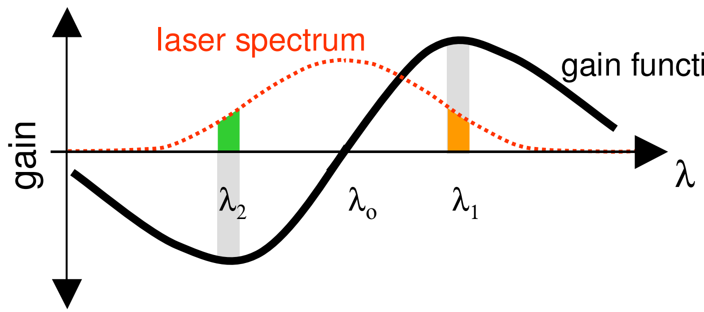


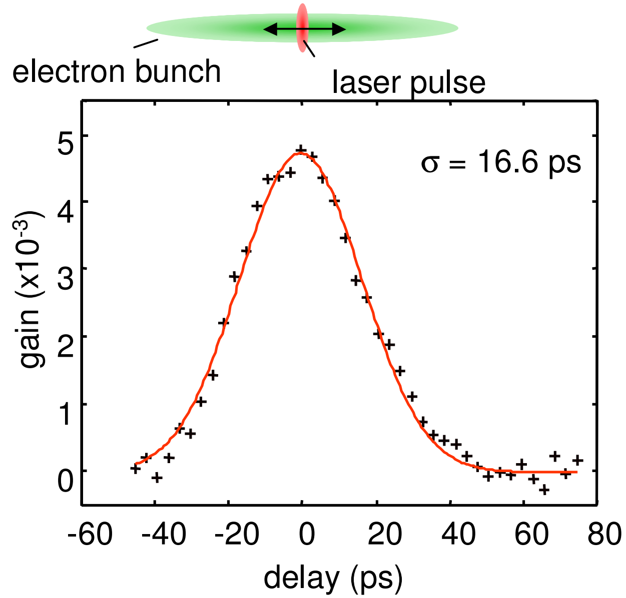
$$gain = \frac{(2\pi M)^2}{\gamma}\frac{\pi}{2}\frac{I_{peak}}{I_A}\frac{d}{dx}\left(\frac{\sin^2(x)}{x^2}\right)\eta \qquad x = \frac{\pi M(\omega - \omega_o)}{\omega_o}$$

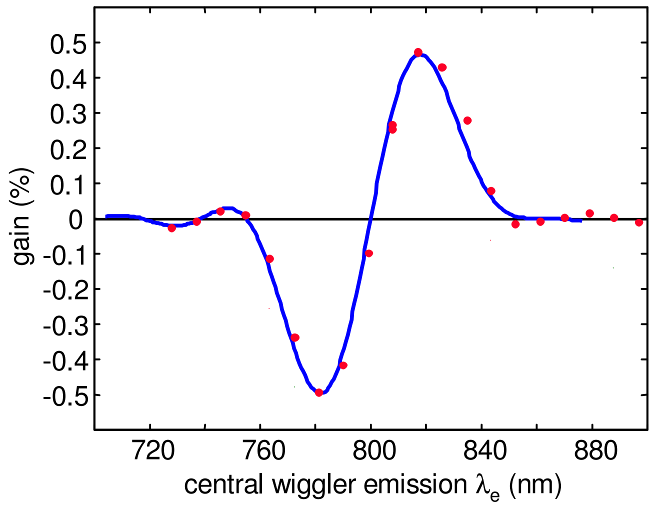
Measurement of Laser Gain through Wiggler

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

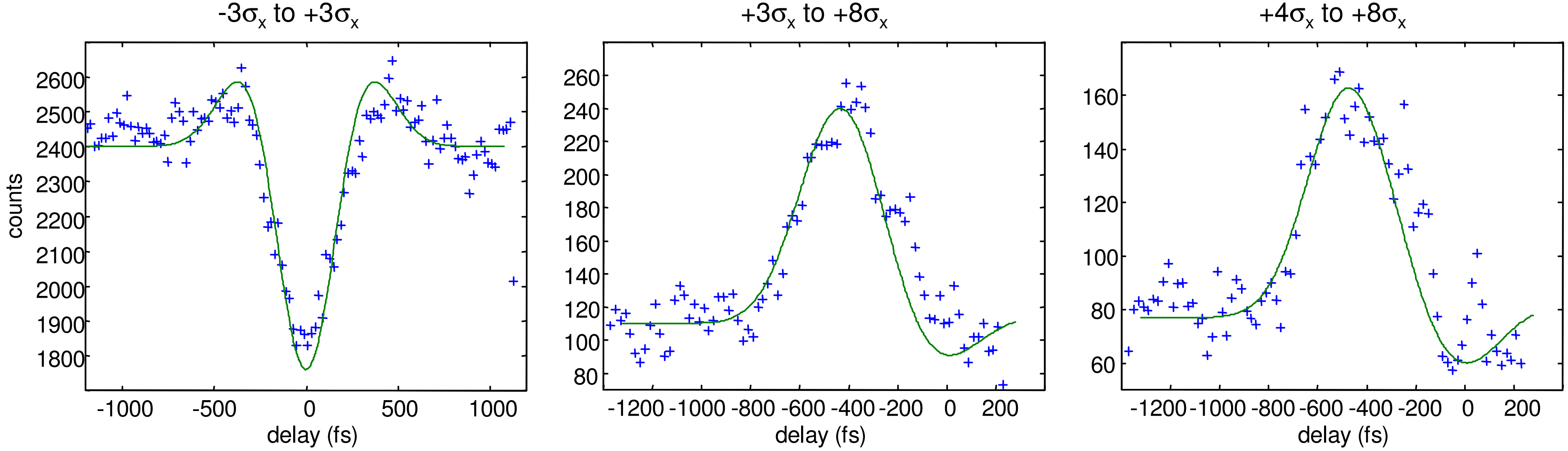




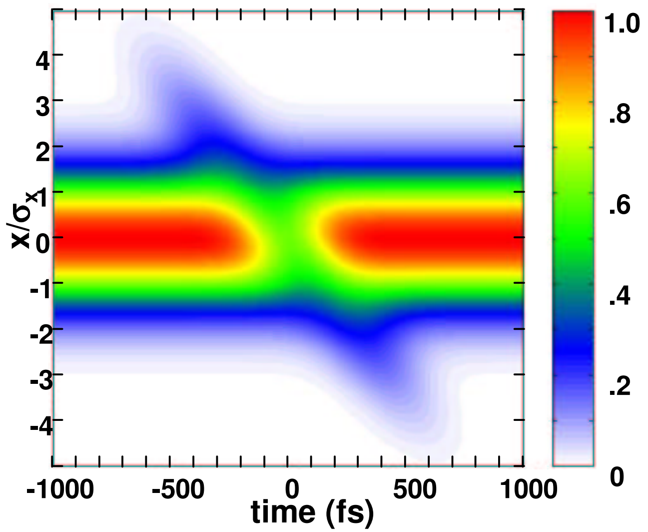


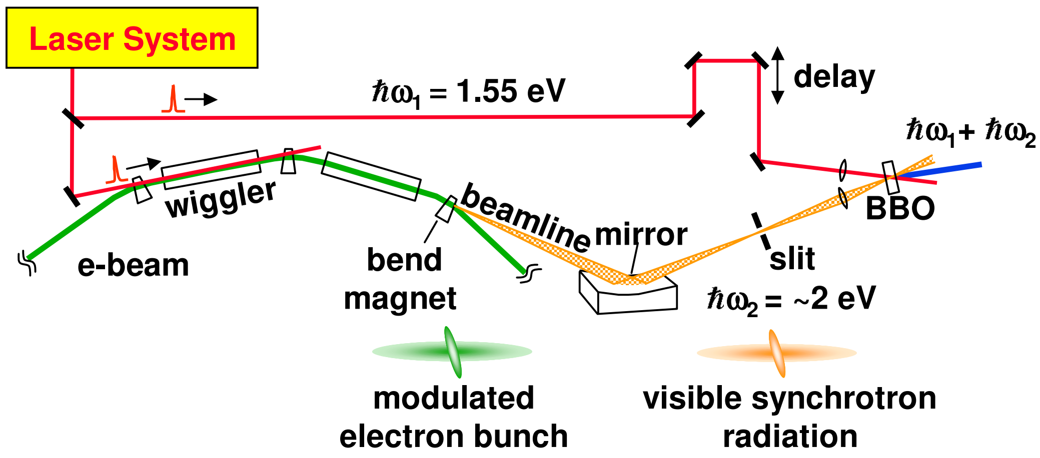


Femtosecond Pulses of Synchrotron Radiation



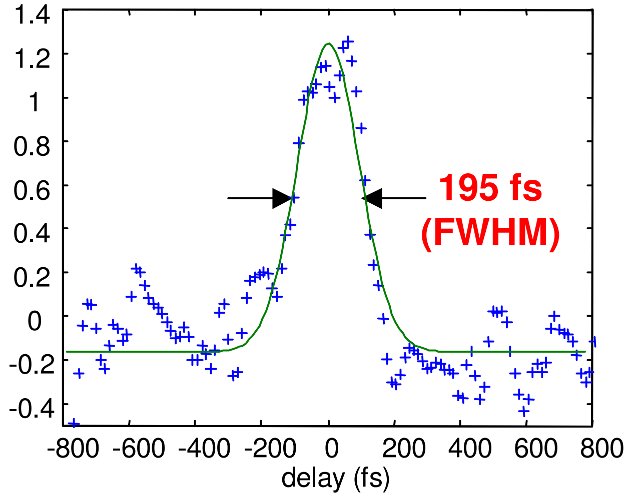
Calculated Electron Density Distribution



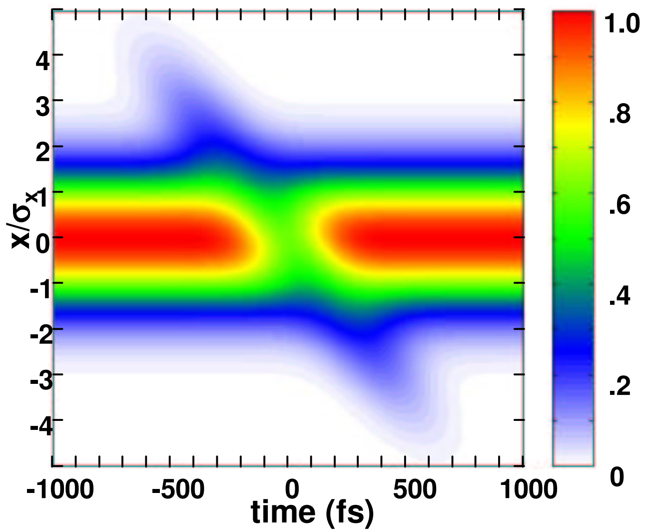


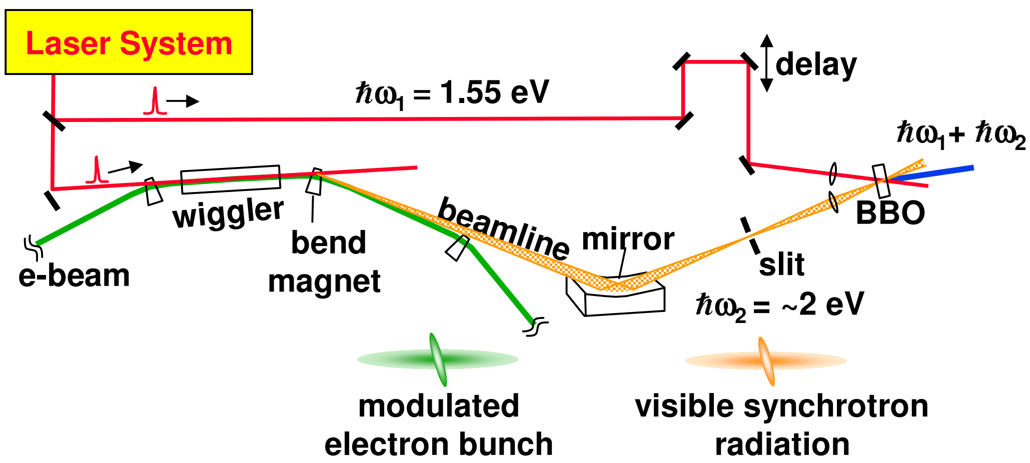
Schoenlein et al., *Science*, **287**, 2237 (2000)

Femtosecond Pulses of Synchrotron Radiation



Calculated Electron Density Distribution





Coherent Synchrotron Radiation

Coherent Infrared Radiation from Modulated Electron Bunch

- longitudinal modulation of electron density ~λ_{IR}
- vacuum chamber aperture must accommodate source divergence (formation length)

$$AB_{photon} = 2\rho\sin(\theta/2)$$

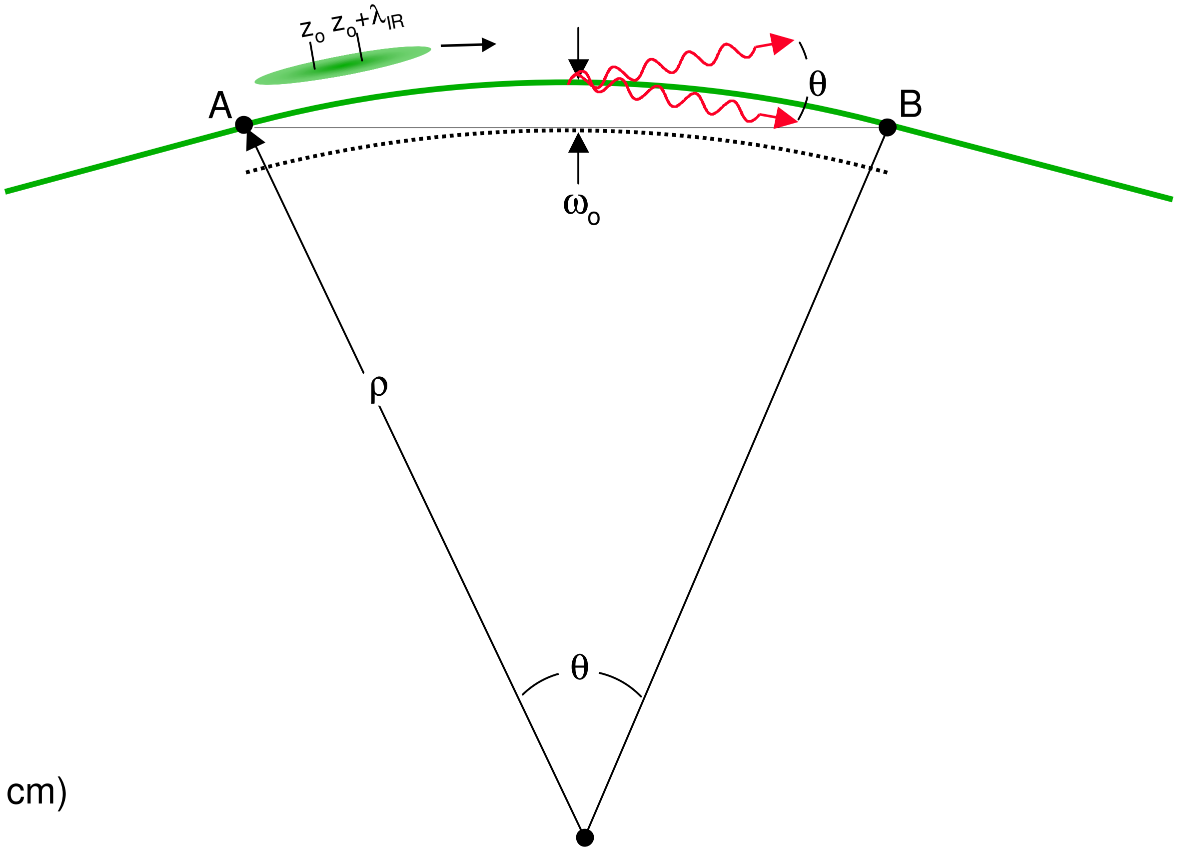
$$AB_e = \rho\theta$$

$$AB_e - AB_{photon} = \lambda_R \approx \rho\frac{\theta^3}{24}$$

$$L_{formation} = \rho\theta \cong Z_R = \frac{\pi\omega_o^{\ 2}}{\lambda_R}$$

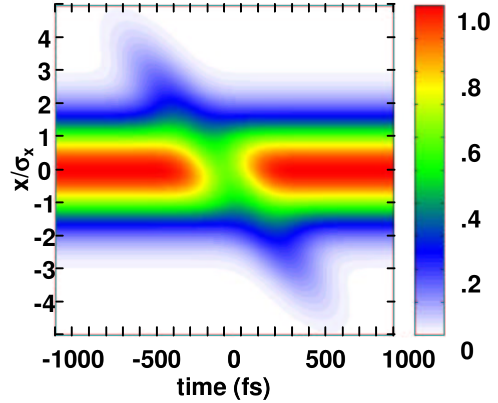
$$\lambda_R \le \rho^{-1/2}\omega_0^{\ 3/2}$$

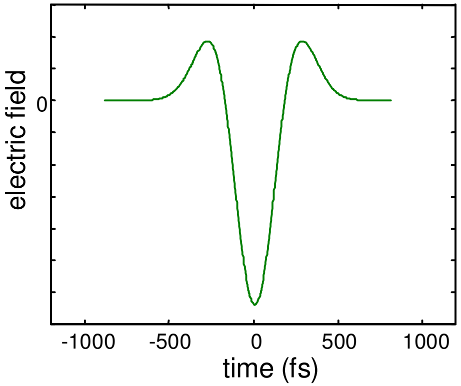
$\lambda_R \le$ 1.4 mm (ρ = 4 m, ω_o = 2 cm)

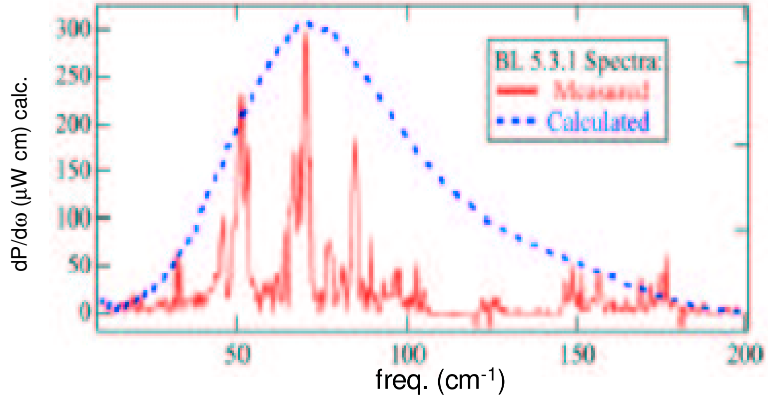


Coherent Infrared – Measurement Schematic

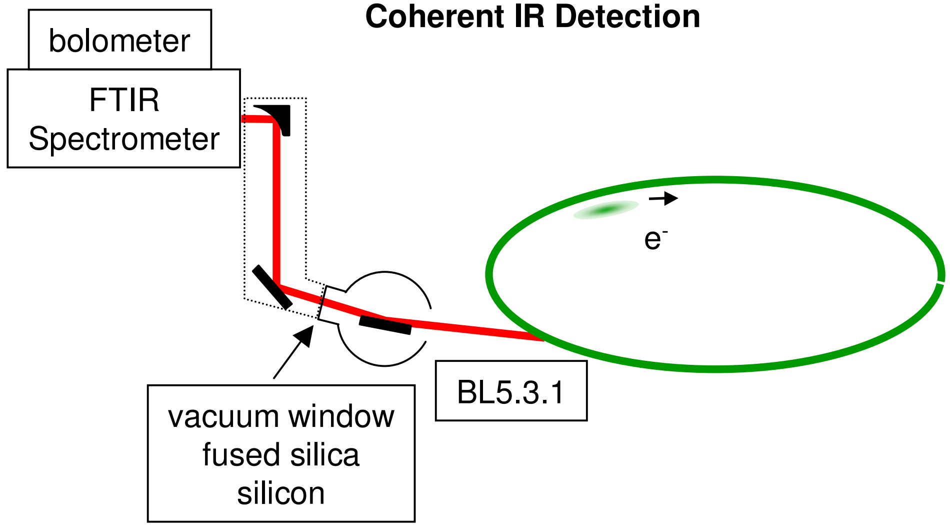
Coherent Synchrotron Radiation



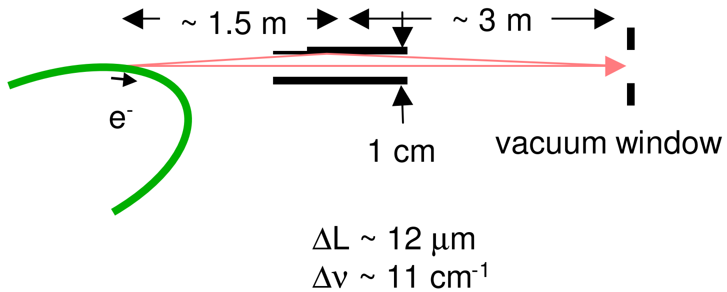




Coherent IR Detection



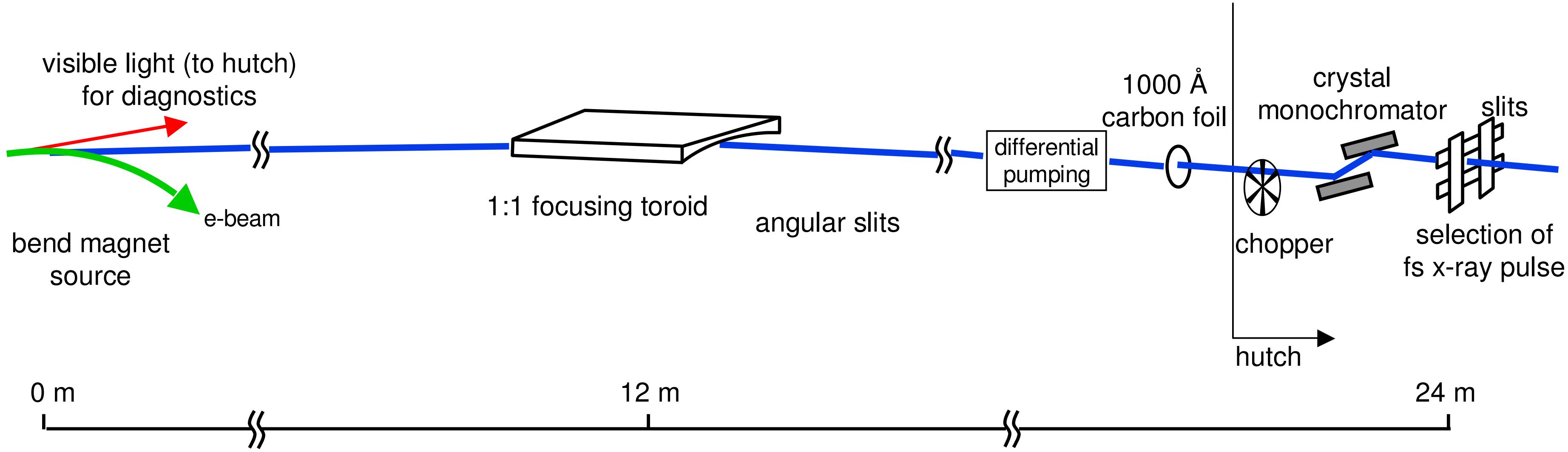
Waveguide Effects



$\Delta L \sim 12\ \mu m$
$\Delta \nu \sim 11\ cm^{-1}$

J. Byrd, Z. Hao, M. Martin, D. Robin, F. Sannibale, R. Schoenlein, A. Zholents, M. Zolotorev, *PRL*, **96**, 2006

Femtosecond x-ray Beamline 5.3.1

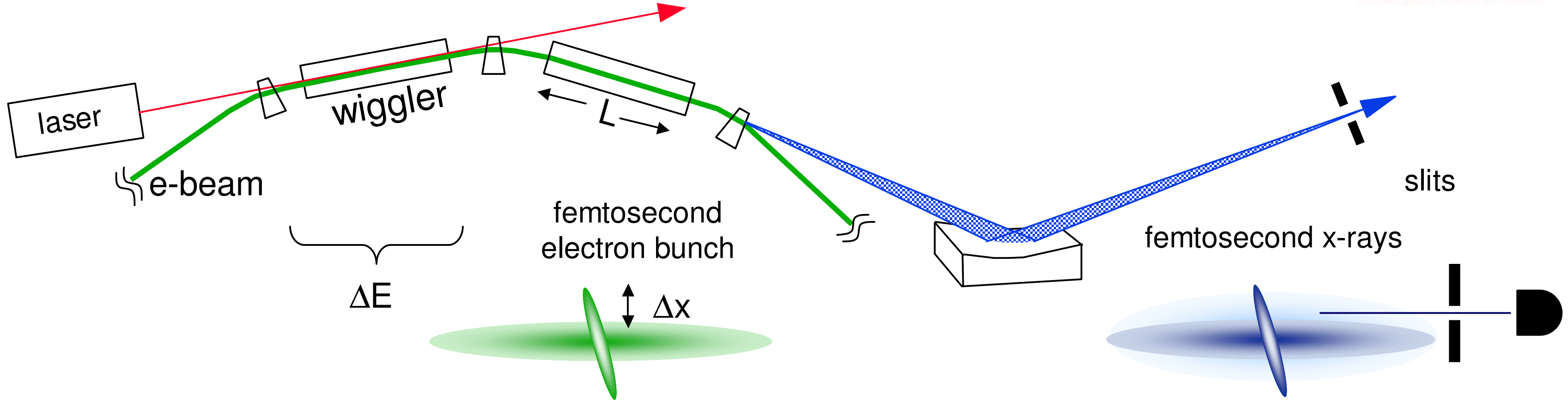


- 1:1 image of bend magnet source
  250 μm (H) x 50 μm (V)
- white beam, 0.1-12 keV
  (possibility for Laue diffraction)

- 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)

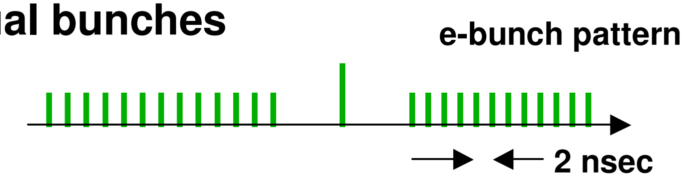
Separation of Femtosecond X-rays



- laser modulation of e-beam energy (ΔE)
  - laser power, wiggler matching
- storage ring dispersion (ΔE→Δx)
  - emittance and lifetime degradation
- beamline image quality
  - mirror scattering (non-specular)
  - depth of source effects

$$\frac{\text{Sig (fsec)}}{\text{background}} > 1$$

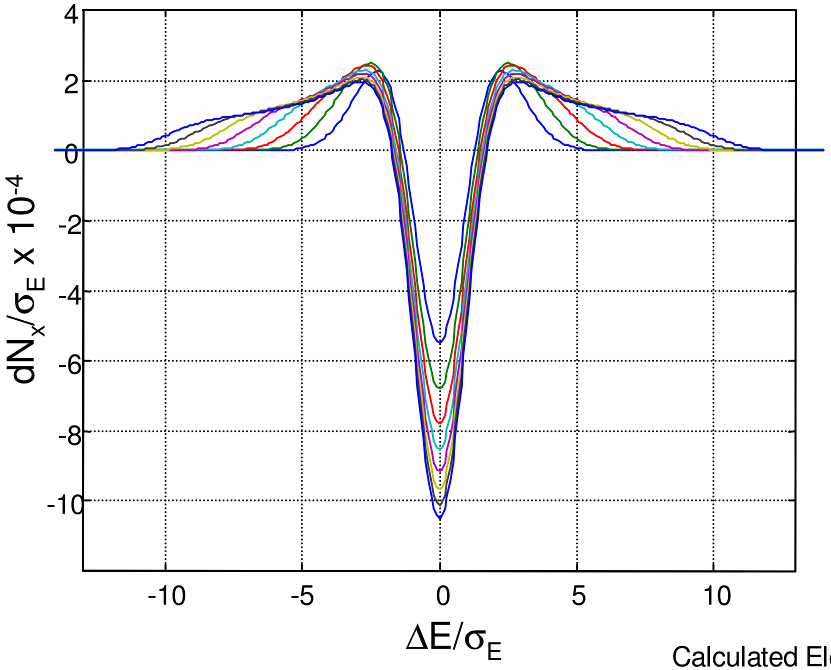
Requirement – gated detectors for isolating individual bunches
avalanche photodiodes
gated microchannel plates

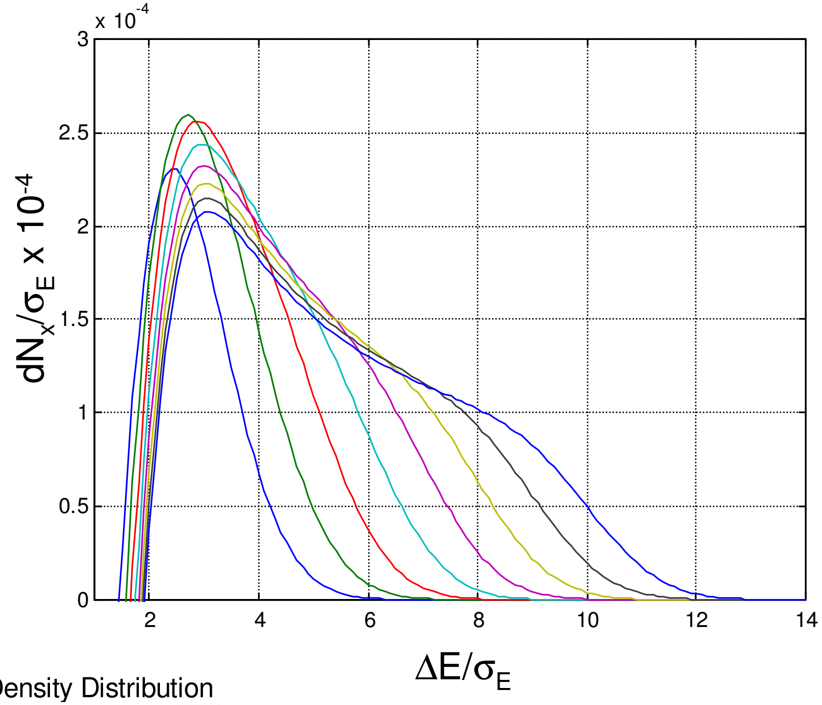


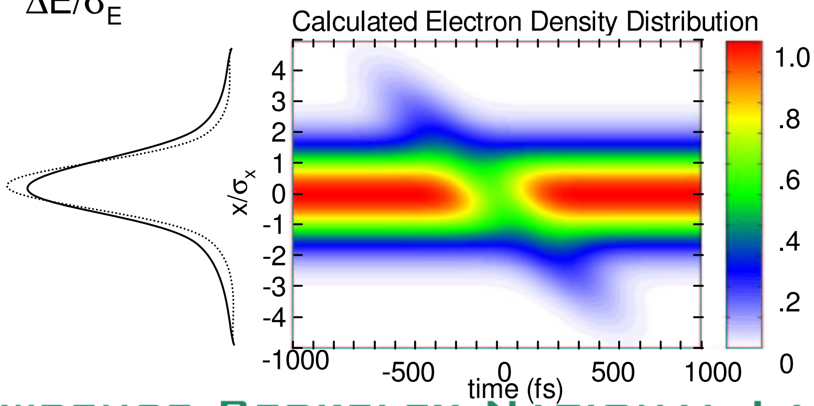
Differential Beam Profiles

$\Delta\rho(E) = \rho(E)_{laser\ on} - \rho(E)_{laser\ off}$

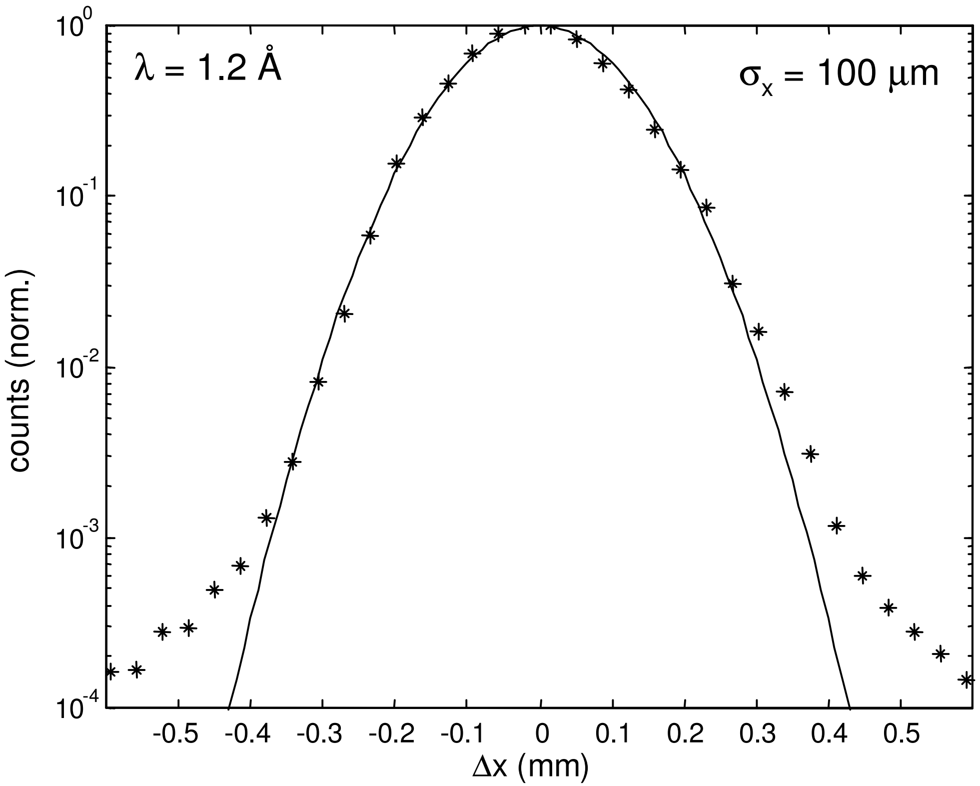
Modulation: $3\sigma_E$ to $10\sigma_E$ $\quad \tau_{laser}/\tau_e = 10^{-3}$

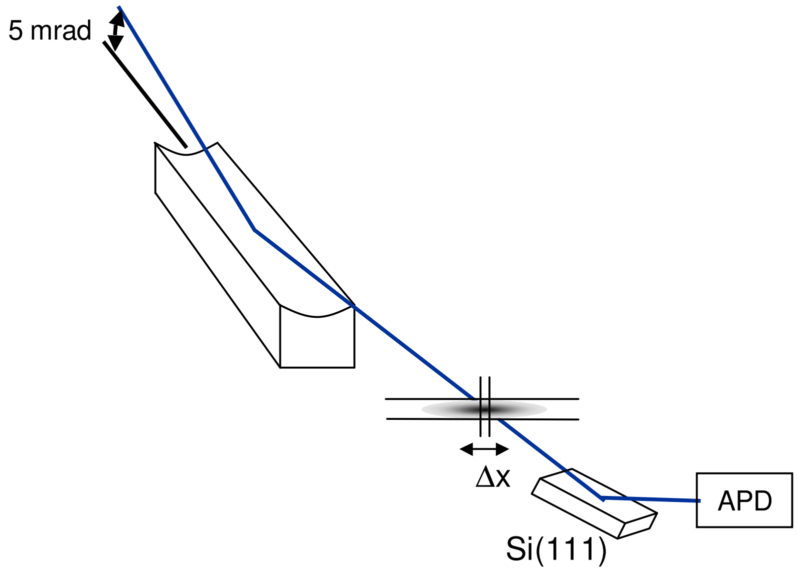






Beam Profile Measurements – BL5.3.1 Camshaft Bunch

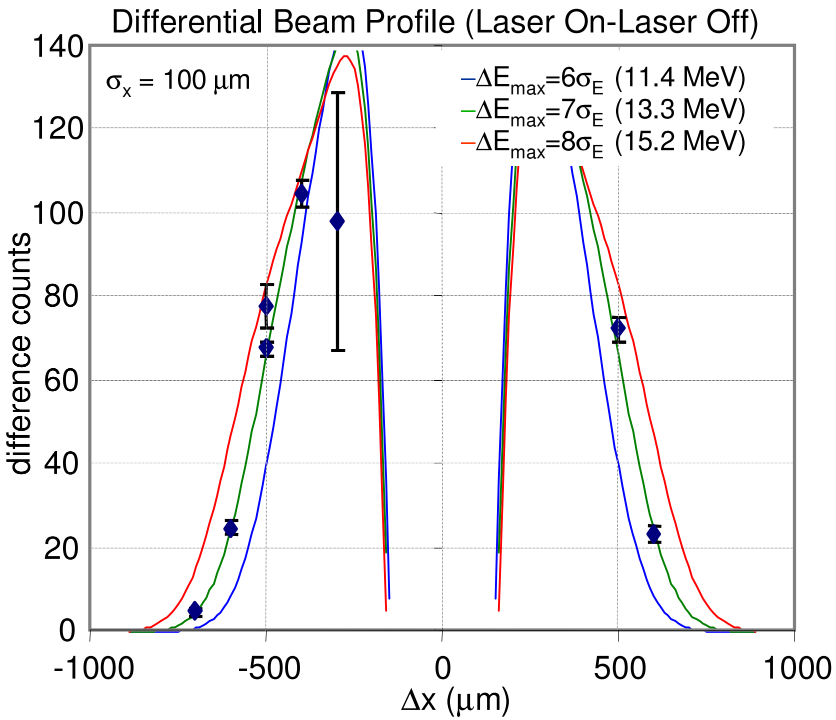


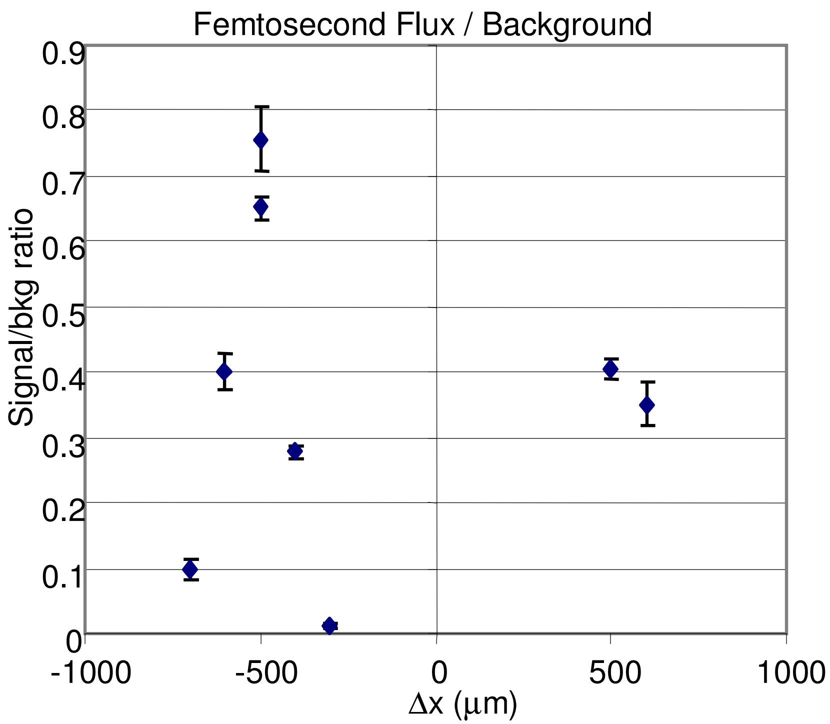


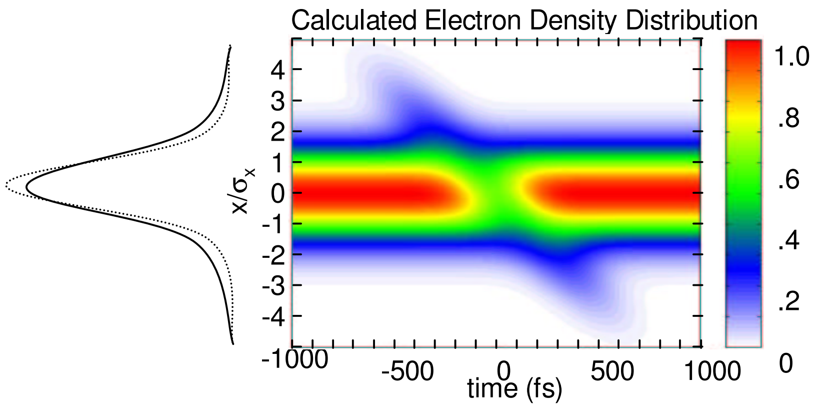
- fraction of e-beam that is modulated = 75 fs × 0.1 /75 psec = 10^{-4}

$\sigma_E/\Delta E$=1/10

Femtosecond X-ray Profile Measurements – BL5.3.1

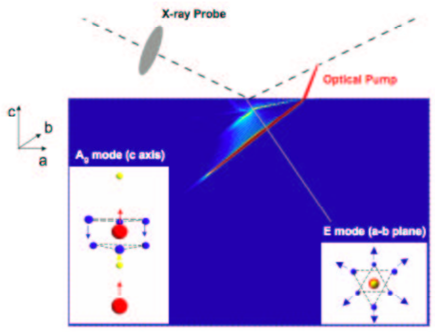




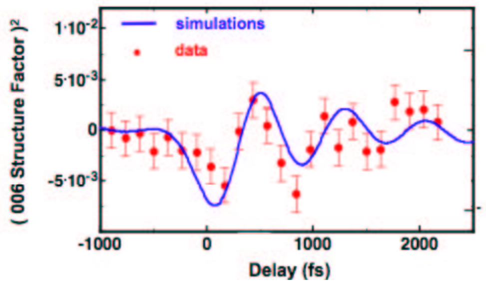


Peak Laser Power ~ 10 GW
(0.8 mJ, 75 fs)

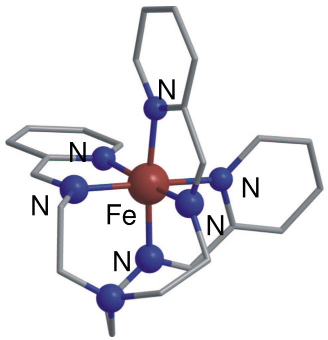
Science at time-resolved x-ray science beamline (ALS BL5.3.1)



- mÅ charge displacement – THz phonon-polariton mode in $LiTaO_3$
  A.Cavalleri et al., *Phys. Rev. Lett.,* **95**, 067405, (2005).
- Ultrafast insulator-metal transition in VO_2 - XANES
  A.Cavalleri et al., *Phys. Rev. Lett.,* **95**, 067405, (2005).
- 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
  S. L. Johnson et al., *Phys. Rev. Lett.,* **94**, 057407 (2005)
  S. L. Johnson et al., *Phys. Rev. Lett.* **91**, 157403 (2003)
- 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 $\eta_2 = \tau_{laser} / \tau_{synchrotron} = 10^{-3}$ ($\tau_{x\text{-}ray} \approx 170$ fs)
  (70 fs) (70 ps)
- repetition rate $\eta_3 = f_{laser} / f_{synchrotron} = 2\text{x}10^{-6}$
  (1 kHz) (500 MHz)

  $f_{laser} / f_{synchrotron}$ $\quad f_{limit} \approx 3\times\frac{\text{number of bunches}}{\tau_{damping}} = 150 \text{ kHz}$
  (40 kHz) (500 MHz)

Average Femtosecond X-ray Flux ~ Average Femtosecond Laser Power

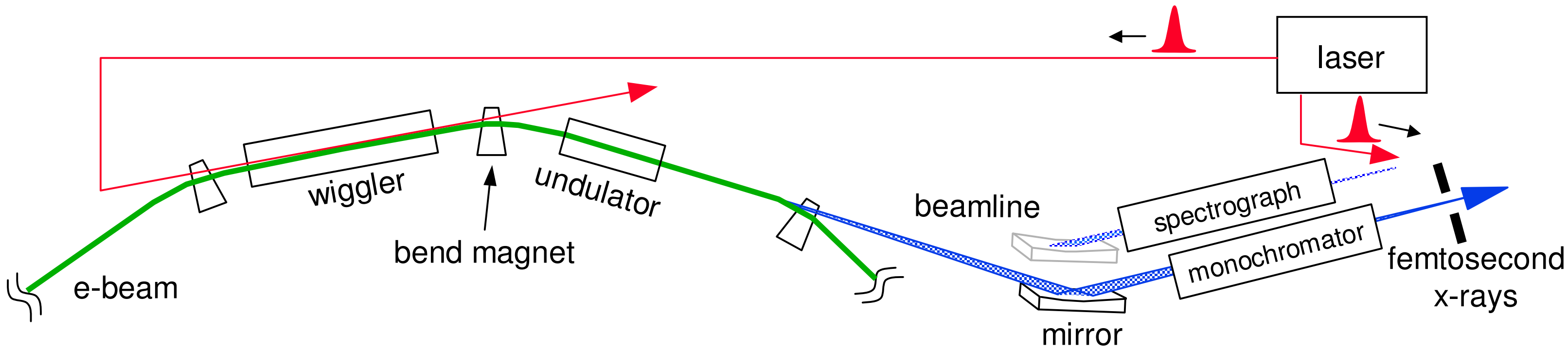
Bend Magnet

- flux ~10^{13} ph/sec/0.1% BW
- brightness ~10^{16} ph/sec/0.1% BW

Undulator

- flux ~10^{15} ph/sec/0.1% BW
- brightness ~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)

  x10^2 increase in flux, x10^3 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)

  x10 increase in flux

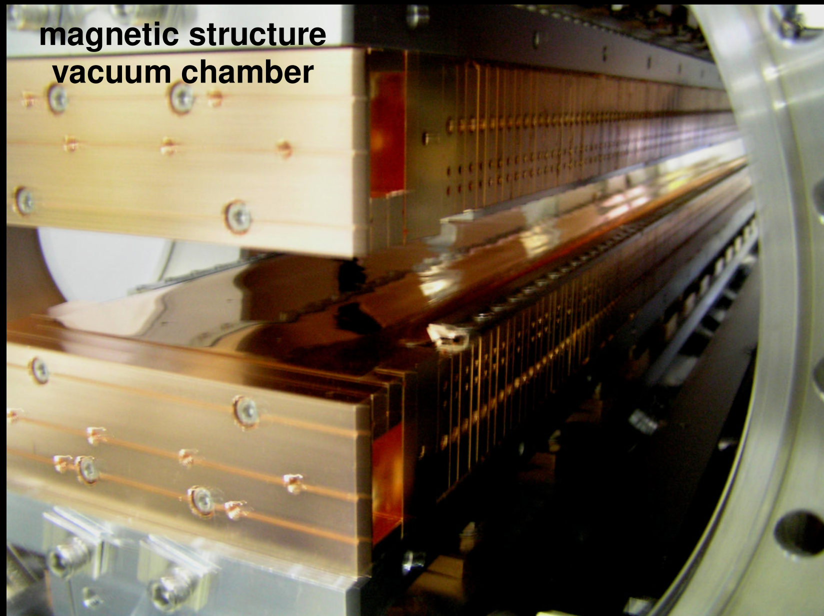
IV. Storage Ring Modifications

- local vertical dispersion bump – sector 6 and/or 5

In-Vacuum Undulator/Wiggler

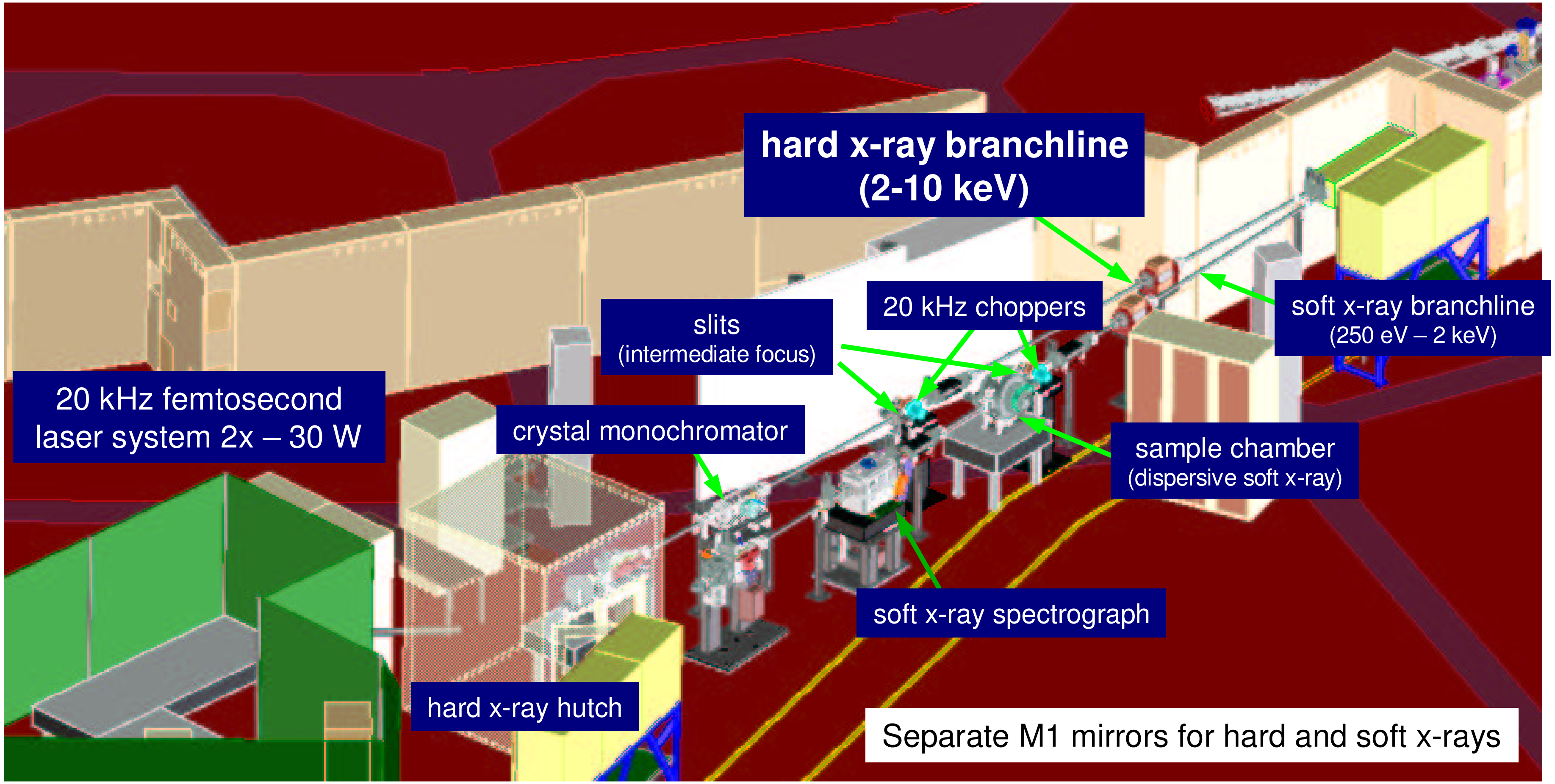
S. Marks et al.

Specifications	
Magnetic gap	5.5 mm
Period	30 mm
No. periods	50
Vacuum gap	>5 mm
B_o	1.45 T



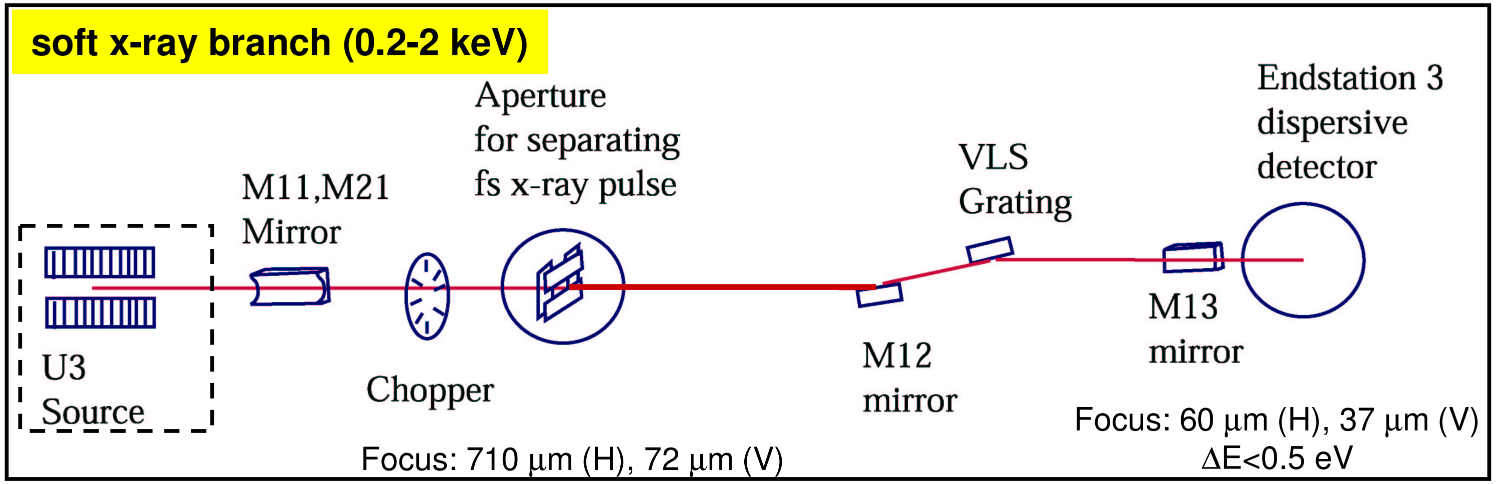
ALS Femtosecond Undulator Beamline 6.0 Layout

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

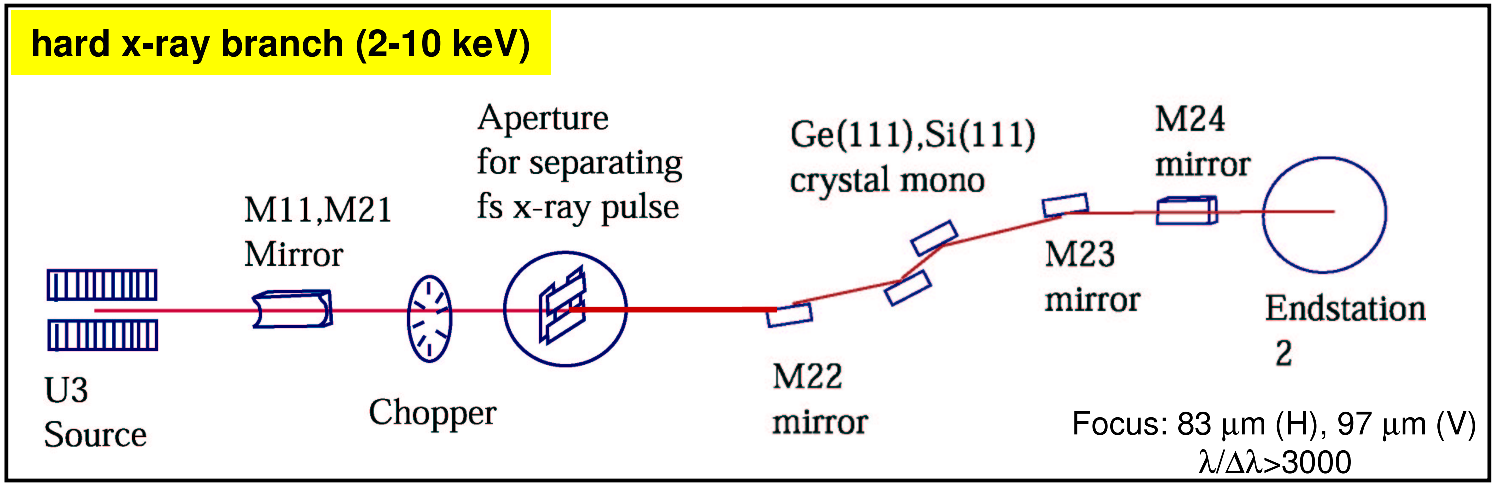


ALS Femtosecond Undulator Branchlines

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



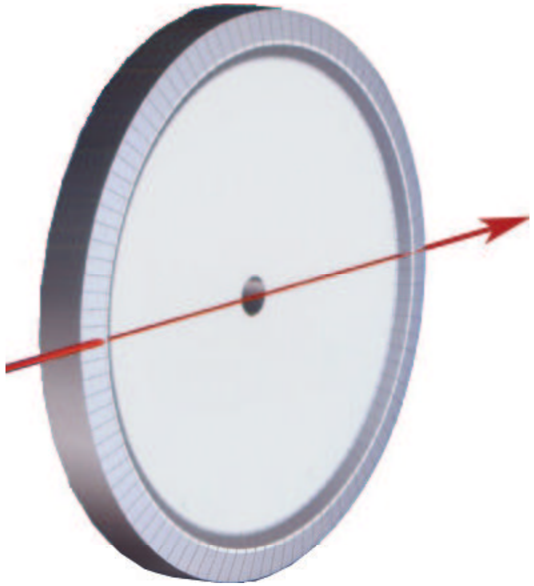
commission 2005



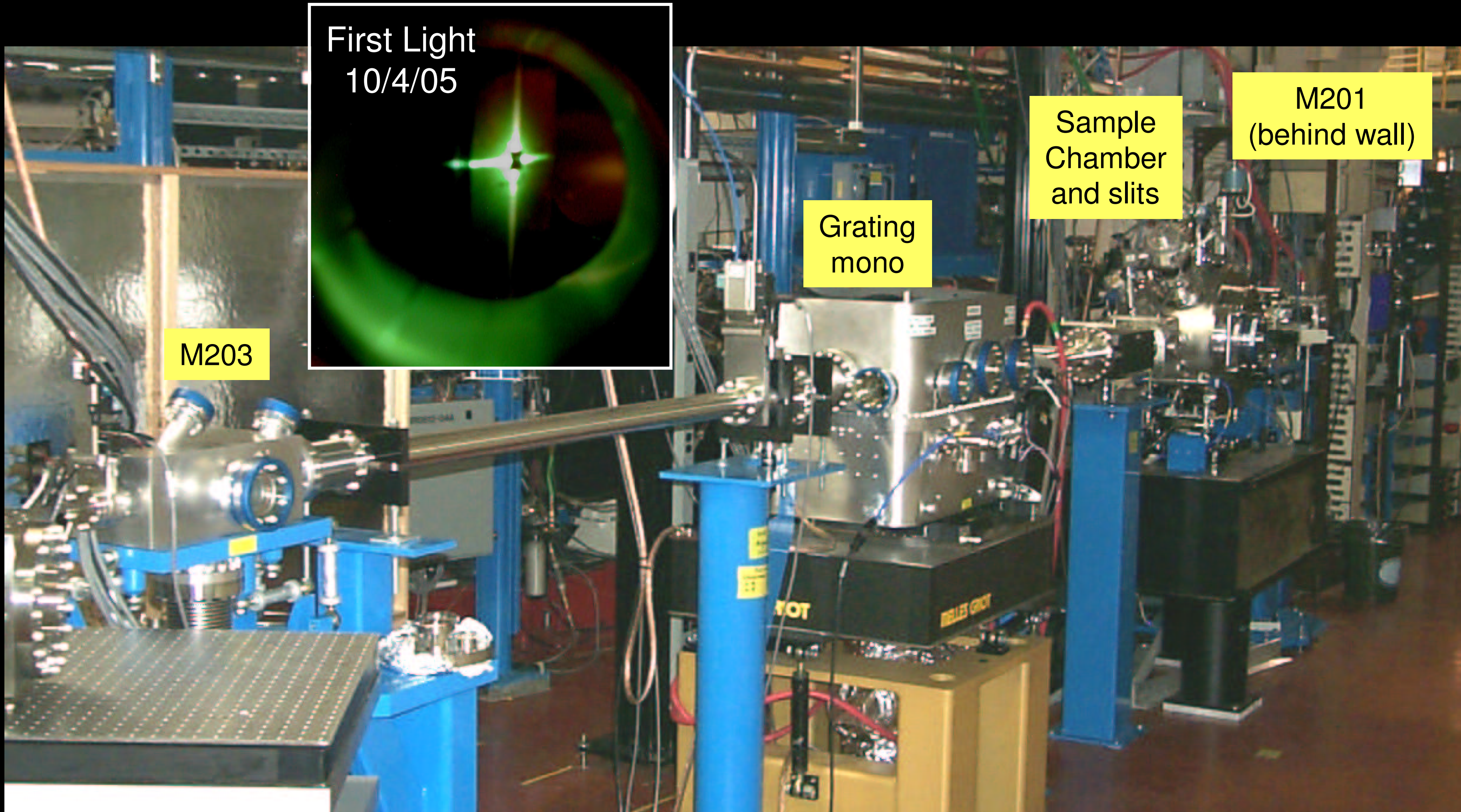
commission Fall 2006

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



Femtosecond Soft X-ray Beamline 6.0.1.2

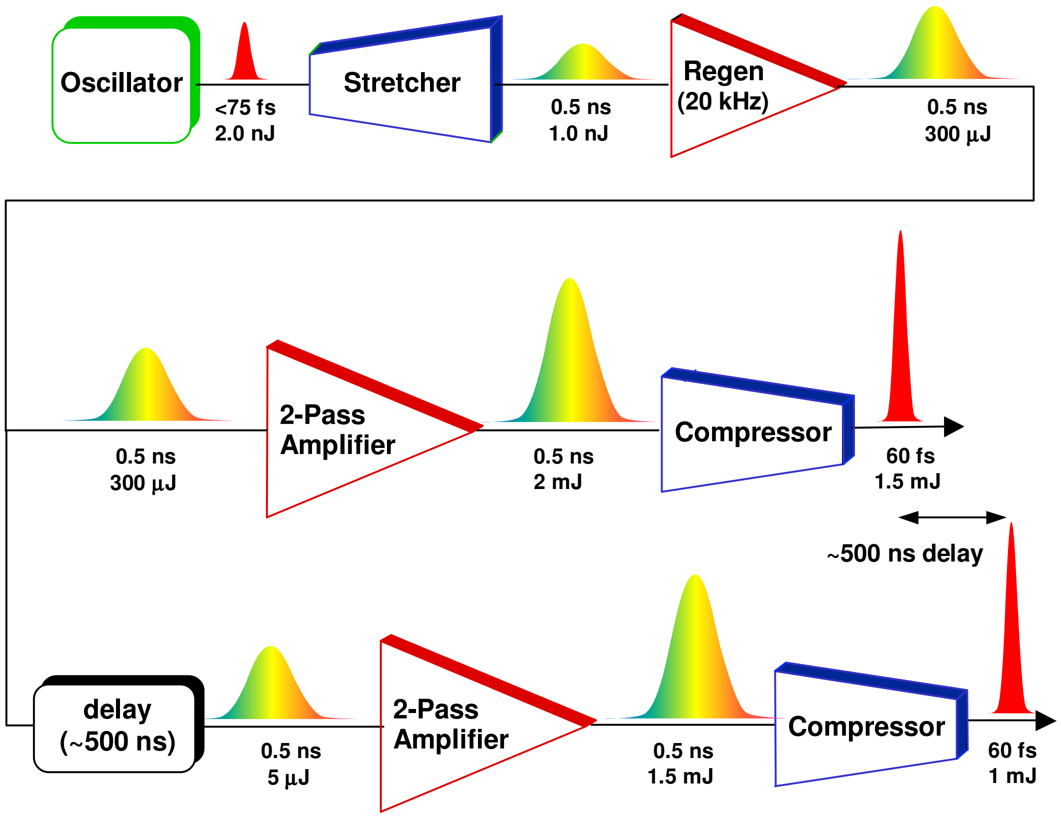


Femtosecond Laser System

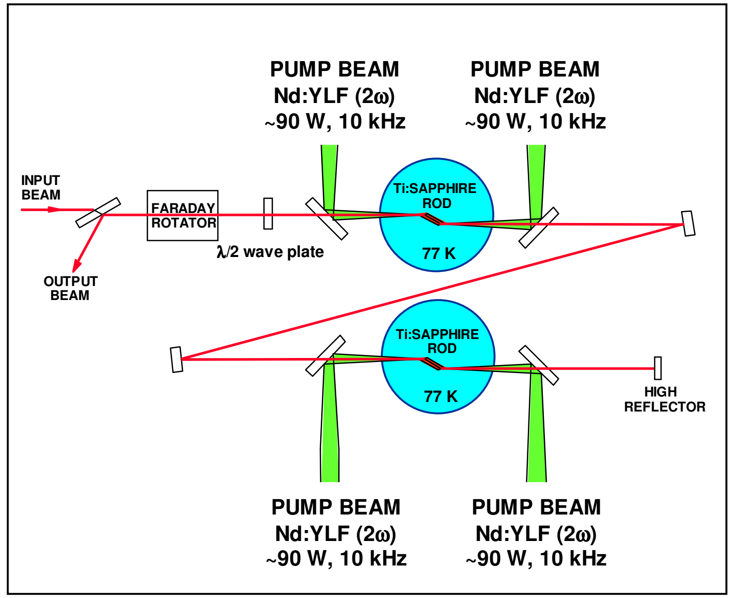
R. Wilcox, R. Schoenlein

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



cryogenic power amplifier



Femtosecond Laser System

R. Wilcox, R. Schoenlein

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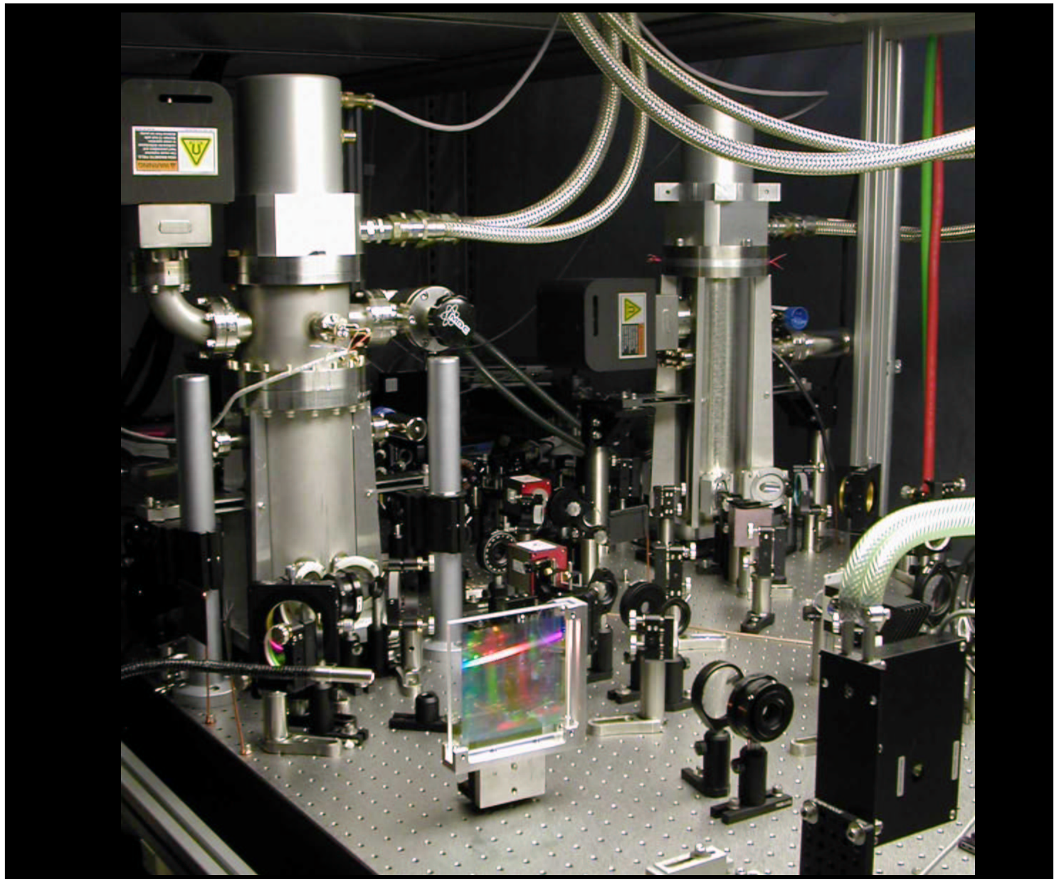
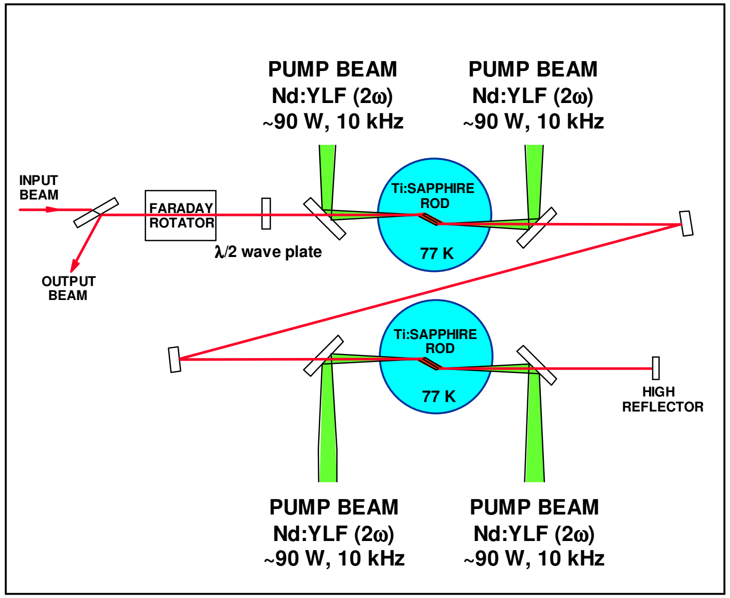
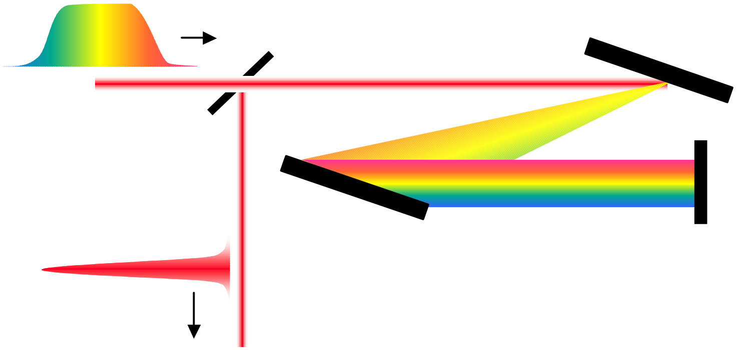


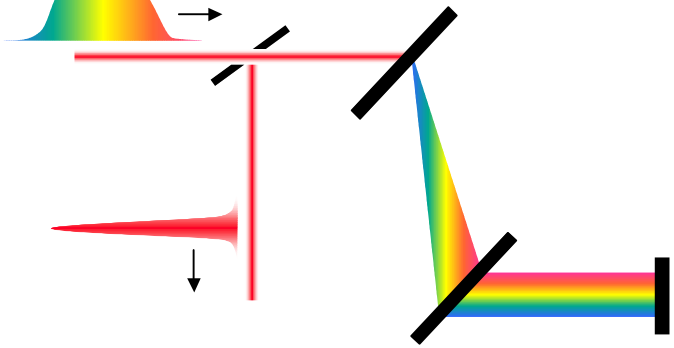
cryogenic power amplifier



Femtosecond Pulse Compression

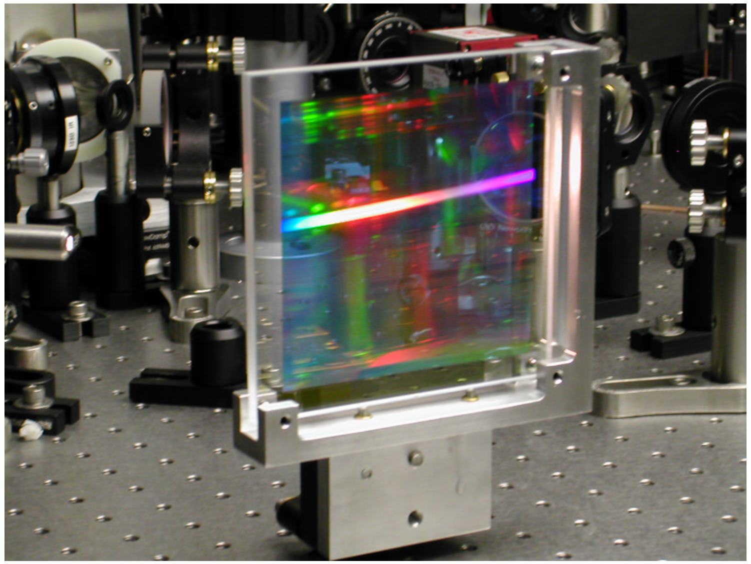


Diffraction Grating Compressor

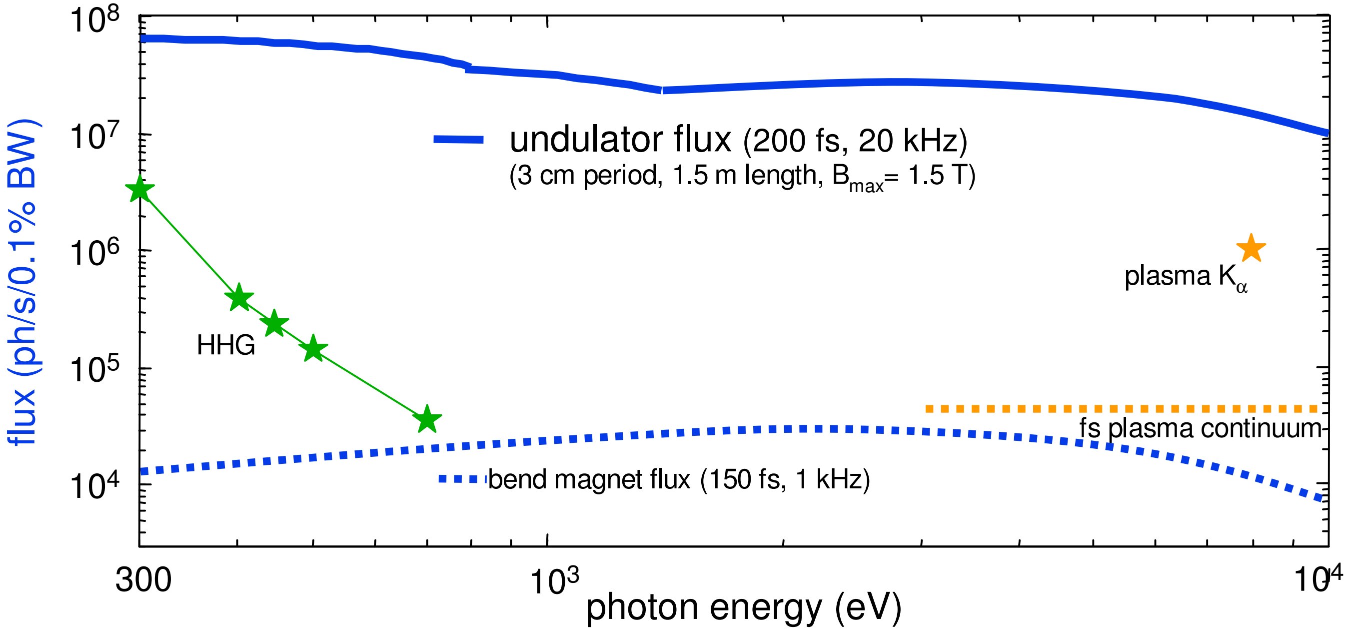


Transmission Grating Compressor

Power Densities: 10-100 W/cm^2



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_α - 10^{10} ph/s/4π (proj. 10^{12} with Hg target)
cont. 6×10^7 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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Femtosecond Undulator Beamline Development

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LBNL

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D. Cambie
Engineering Division
LBNL