Ultrafast X-ray Science

Lawrence Berkeley National Laboratory

KEK Japan, March 2005

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Will consider implementation to Synchrotrons

a) laser e-beam "slicing"

and

b) rf orbit deflection

previously published in:

1) A. Zholents, M. Zolotorev, *Femtosecond x-ray pulses of synchrotron radiation*, Phys. Rev. Lett. V76, N6, (1996), pp.912-915.
2) A. Zholents, P. Heimann, M. Zolotorev, J. Byrd, *Generation of subpicosecond X-ray pulses using RF orbit deflection*, Nuclear Instruments & Methods in Physics Research Sect. A (425)1-2 (1999) pp. 385-389.
3) R. W. Schoenlein, S. Chattopadhyay, H. H. W. Chong, T. E. Glover, P. A. Heimann, C. V. Shank, A. A. Zholents, and M. S. Zolotorev, *Generation of Femtosecond Pulses of Synchrotron Radiation,* Science, Mar 24, 2000: 2237-2240.
4) R. W. Schoenlein, S. Chattopadhyay, H. H. W. Chong, T. E. Glover, P. A. Heimann, C. V. Shank, A. A. Zholents, and M. S. Zolotorev, *Generation of Femtosecond X-ray Pulses via Laser-Electron Beam Interaction,* Appl. Phys. B, 1-10, 2000.

Outline

Scientific Motivation

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

Synchrotron X-ray Sources

- X-ray radiation characteristics

Generation of Femtosecond X-rays from Synchrotrons

- Manipulation of the stored electron beam with femtosecond laser pulses
- Results from proof-of-principle experiments at the ALS
- Future prospects, limitations, practical issues – experimental applications
- Future beamlines for femtosecond x-ray spectroscopy at the ALS (BESSY, SLS)

Generation of subpicosecond X-ray pulses using RF orbit deflection

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

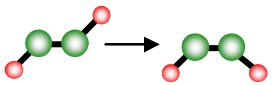
- Structural Transitions in VO_2 (Cavalleri et al.)
- Light-induced Spin-crossover transition in $Fe[tren(py)_3]^{2+}$ (Chong et al.)
- Charge Transfer in $[Ru(bpy)_3]^{2+}$ (Bressler, Chergui et al.)
- Photodissociation dynamics of solvated metal carbonyls (Khalil et al.)
- X-ray/laser ionization dynamics in atomic systems (Hertlein, Belkacem et al.)
- Bonding Properties of Liquid Carbon (Johnson, Falcone et al.)

Femtosecond X-ray Science at the ALS

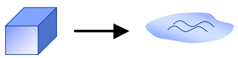
Structural Dynamics in Condensed Matter

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

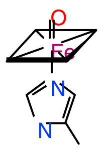
- ultrafast chemical reactions



- ultrafast phase transitions

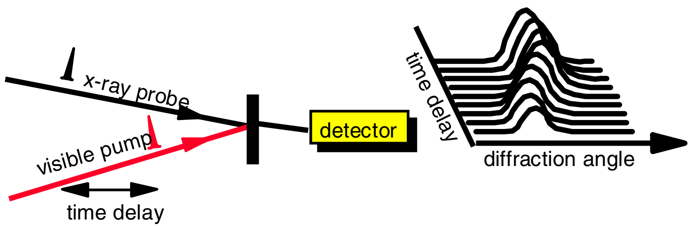


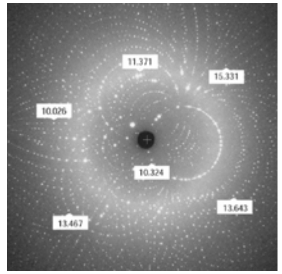
- surface dynamics
- ultrafast biological processes



Rapidly emerging field of research
Physics, Chemistry and Biology

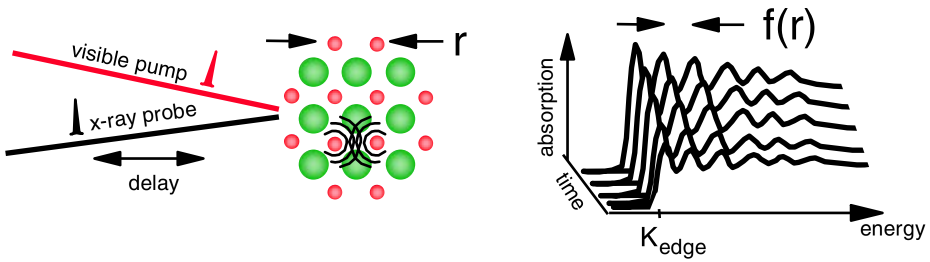
time-resolved x-ray diffraction





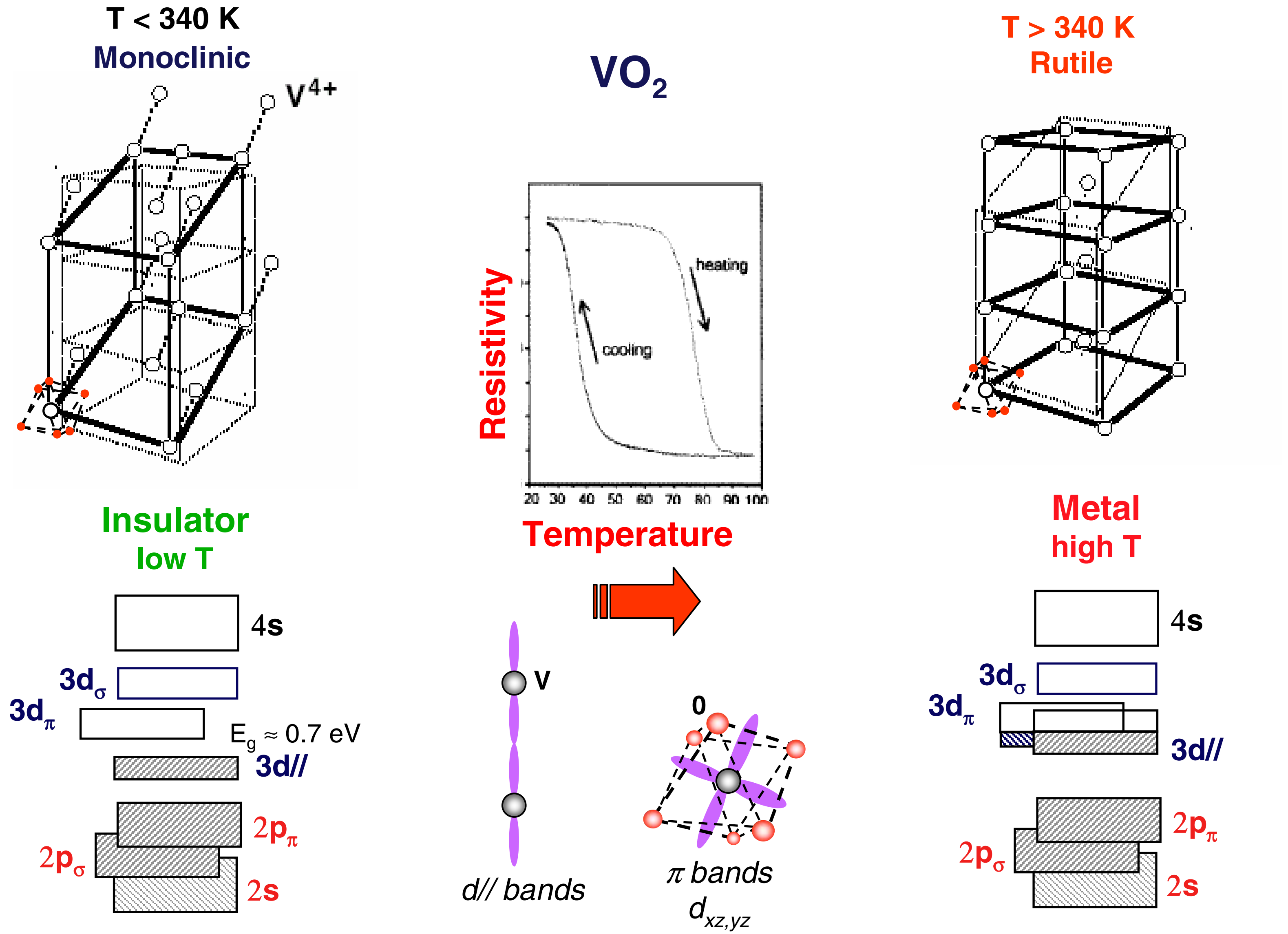
ordered crystals - phase transitions, coherent phonons

time-resolved EXAFS, NEXAS, surface EXAFS



complex/disordered materials - chemical reactions
surface dynamics
bonding geometry

Ultrafast Structural and Electronic Transitions in VO_2

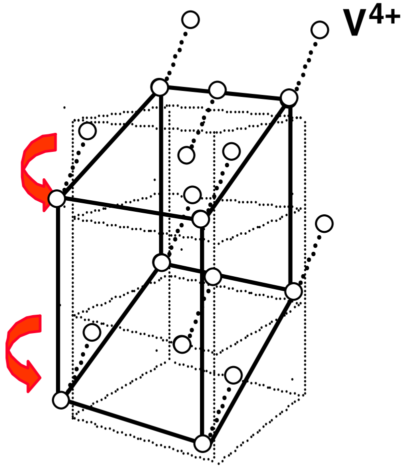


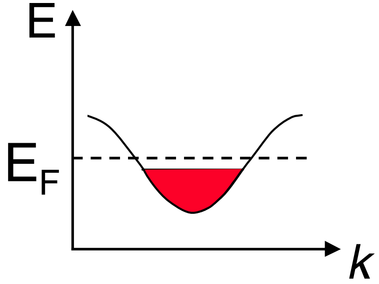
Origin of Insulating Phase of VO_2 ?

Band insulator – structural component ?
Mott-Hubbard insulator – e-e correlation ?

Cell doubling

Peierls transition ?



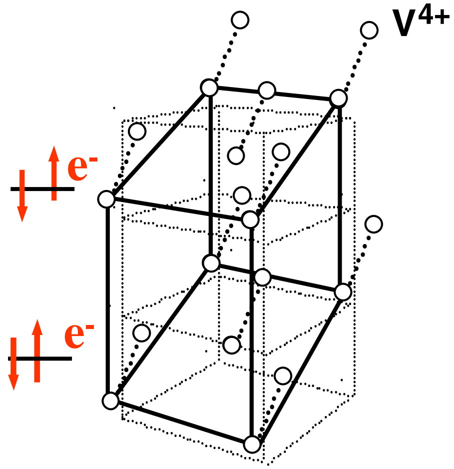


J.B. Goodenough *Phys. Rev.*, **117**, 1442 (1960)

Wentzcowitch et al. *Phys. Rev. Lett.*, **72**, 3389 (1994)

Localization

Mott-Hubbard insulator ?

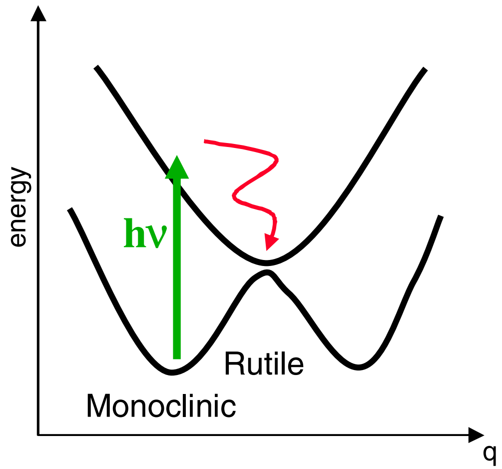


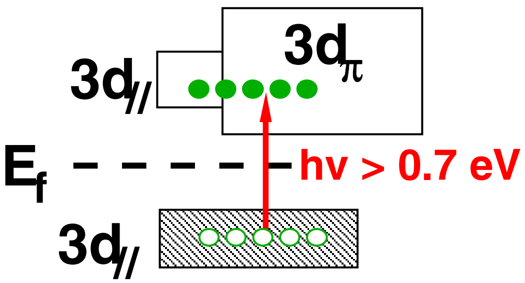
Zylbersztejn and N. Mott
Phys. Rev. B, **11**, 4383 (1975)

Pouget et al. *Phys. Rev., B* **10**, 801, (1974)
Phys. Rev. Lett., **35**, 873 (1975)

Structural Transitions in VO_2

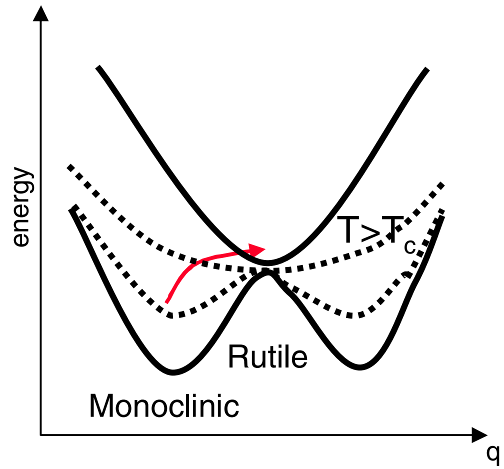
Optical Pumping – excited state





> 50% holes

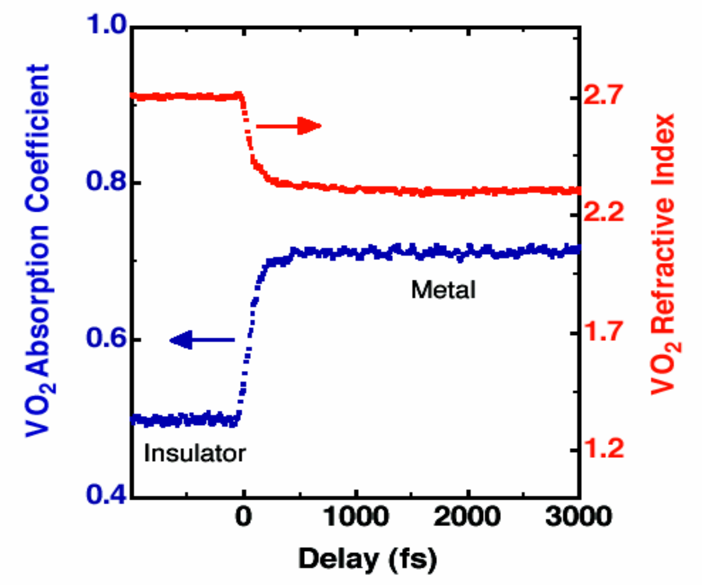
Ground-state vibrational pumping

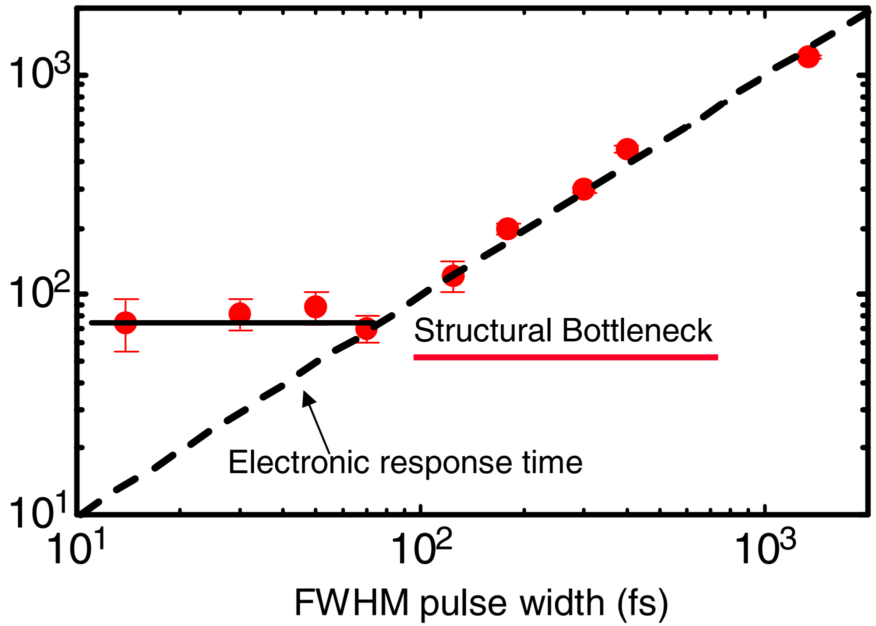


- thermal equilibrium – temperature
- non-thermal – femtosecond IR pulses

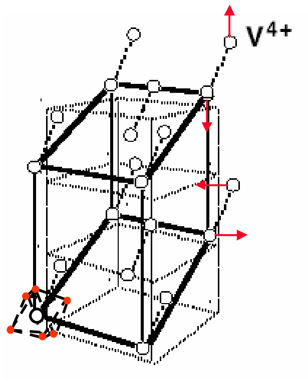
new information compared to adiabatic changes in doping, pressure, temperature, etc.

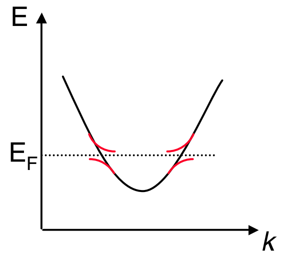
Optical Measurements of VO_2 Insulator-Metal Transition





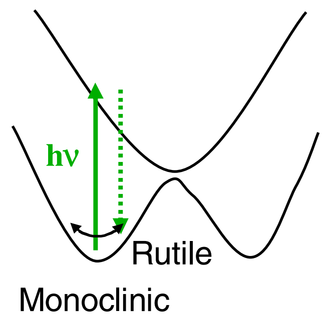
V-V distortion
($T_{optical\text{-}phonon}$~150 fs)





⇒ band (Peierls) type insulator

Low-excitation:
Coherent phonons at 6 THz (T_{vib}~150 fs)
Motion of V atoms (Ag symmetry)



Cavalleri, Dekorsy, Chong, Kieffer, Schoenlein, Shank, submitted to *Phys. Rev. Lett.* (2003).

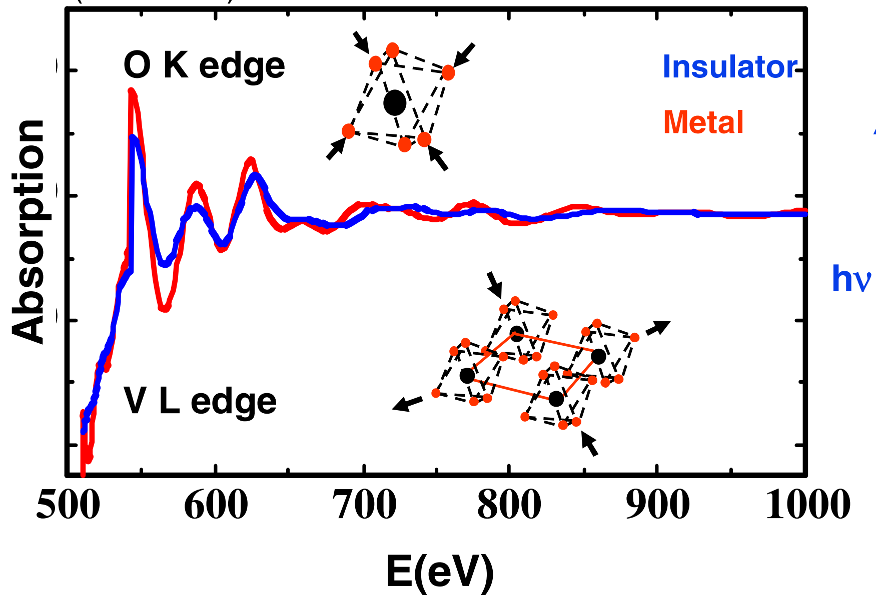
Ultrafast Structural and Electronic Transitions in VO_2

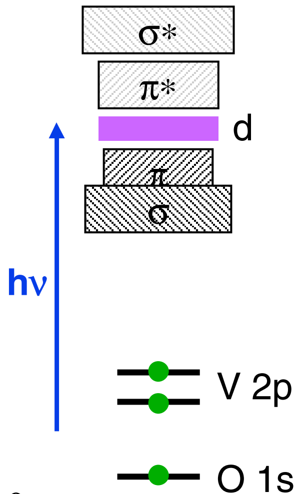
EXAFS
short-range atomic distortions

(sensitivity to V and O)

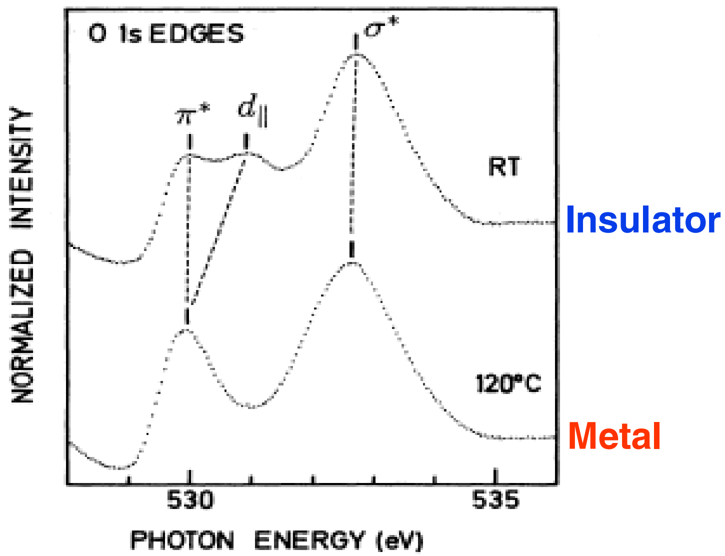
V K_{edge}~5.5 keV

(simulation)



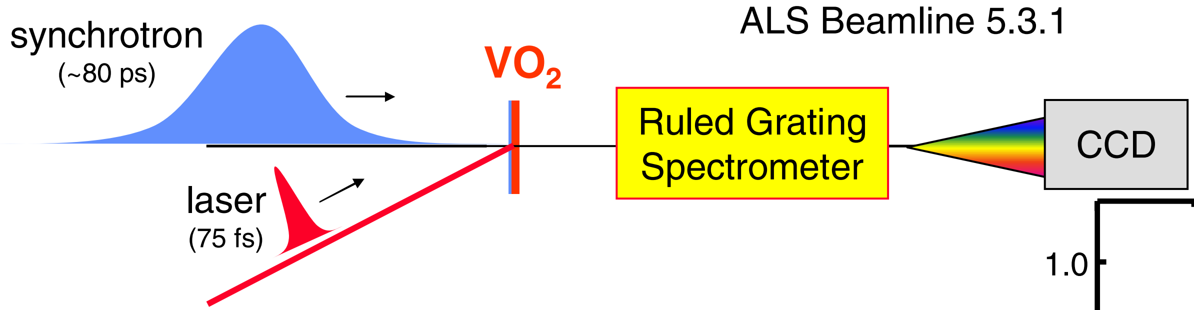


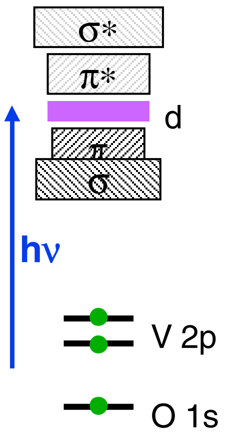
NEXAFS
electronic transition

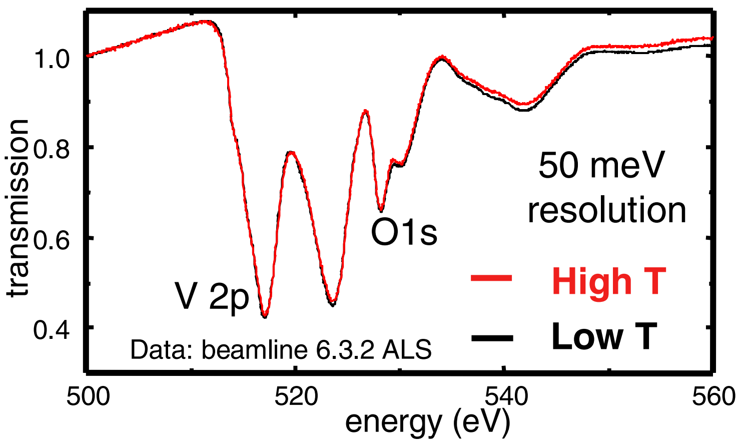


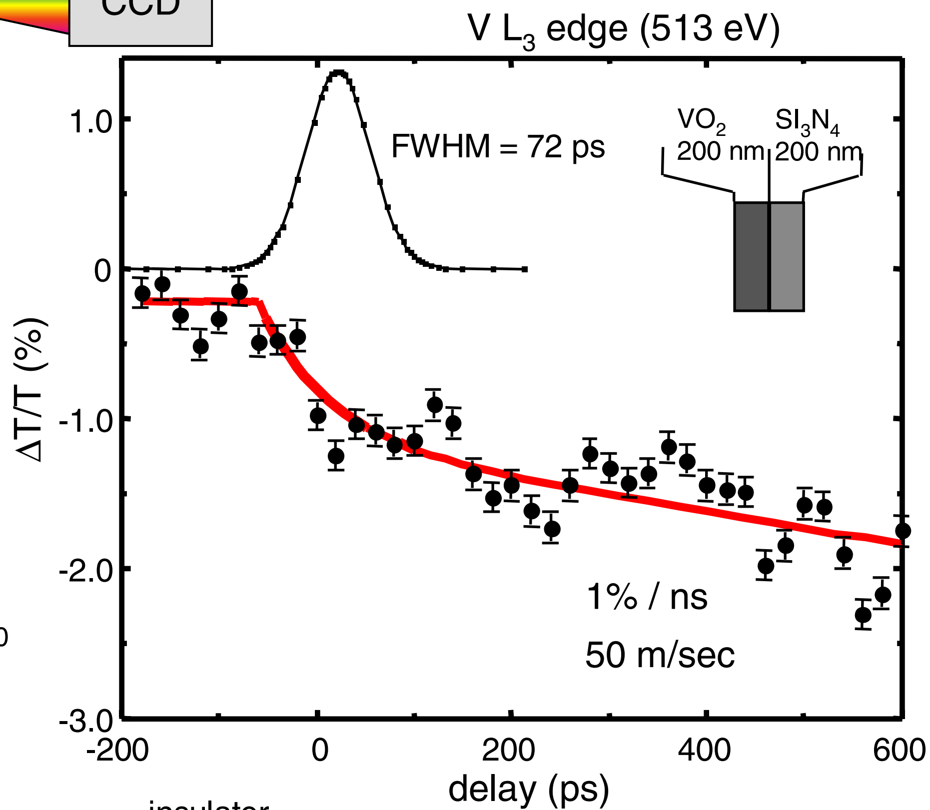
Abbate et al., *Phys. Rev. B*, **43**, (1991).

Time-resolved NEXAFS Measurements in VO_2







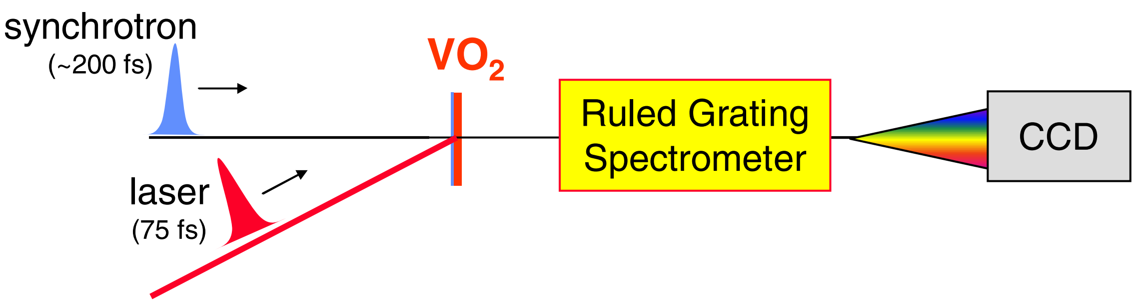


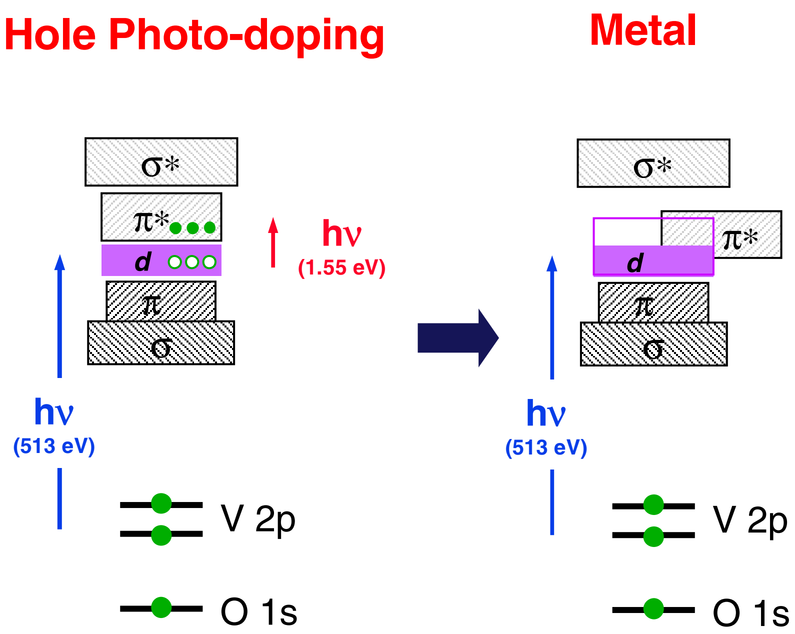
Signal on picosecond time scale dominated by growth of metallic phase

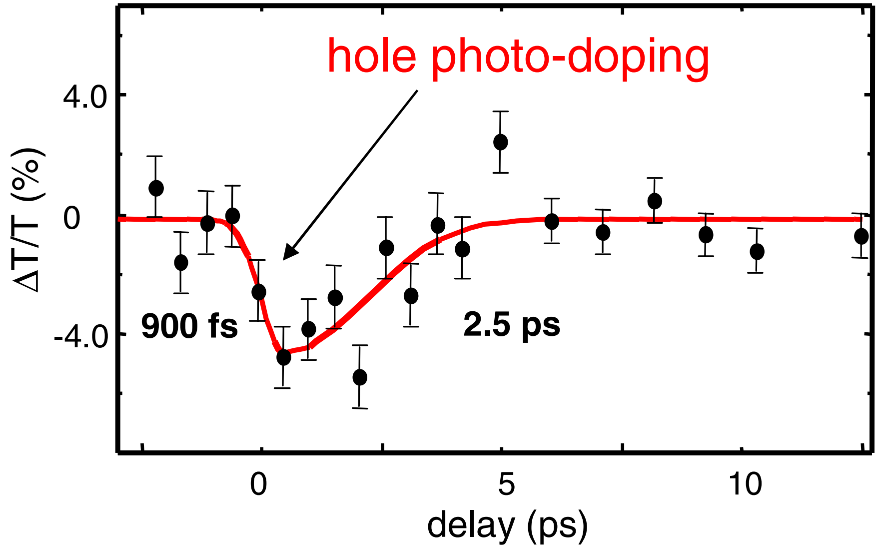
Cavalleri et al., Physical Review B 69, 153106 (2004)

Femtosecond NEXAFS Measurements of I-M Transition in VO_2

ALS Beamline 5.3.1

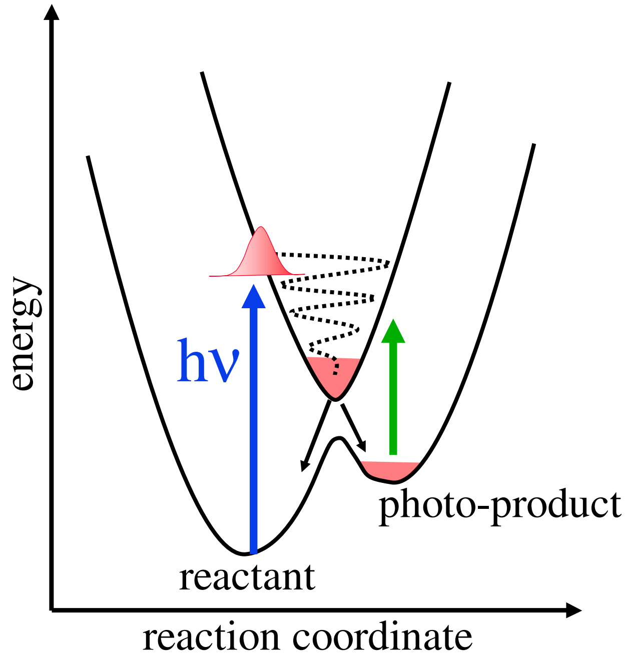




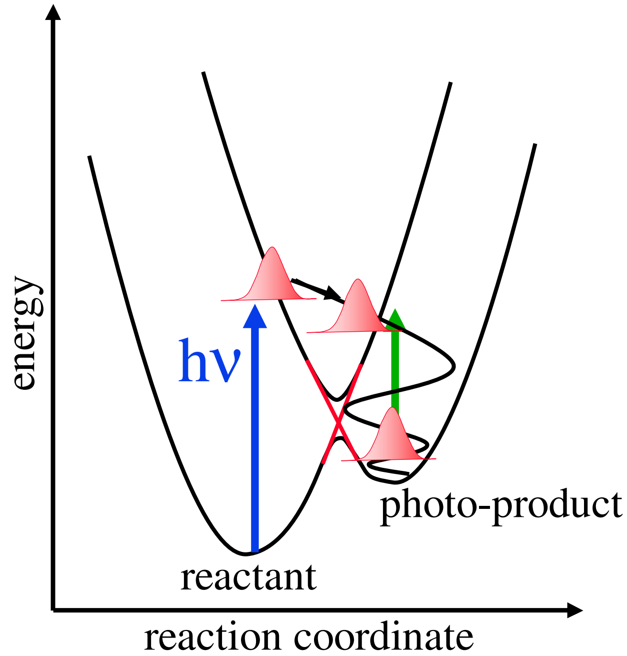


A. Cavalleri

Atomic and Electronic Structural Dynamics of the Transition State



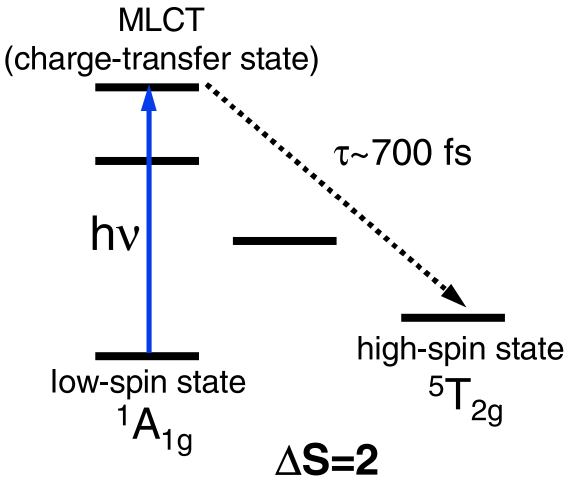
$\tau_{IVR} < \tau_{IC}$



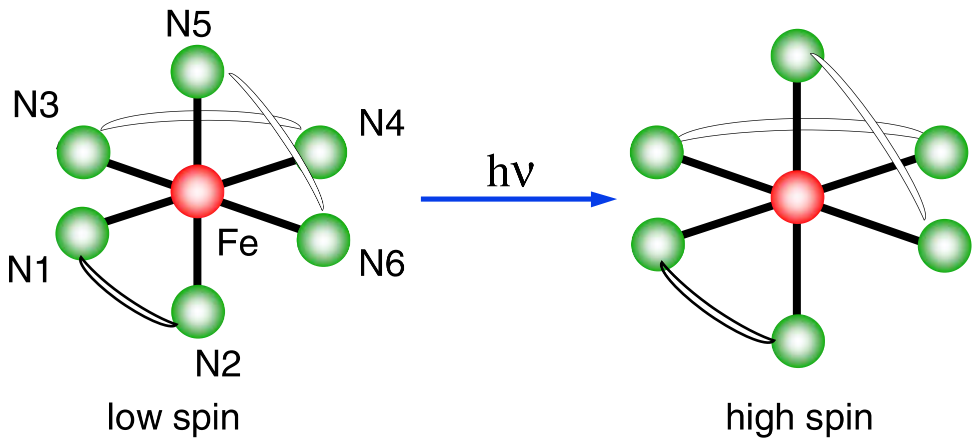
$\tau_{IC} < \tau_{IVR}$

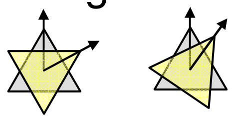
- intramolecular vibrational relaxation (IVR)
- internal conversion - IC

Fe^{II} Spin-Crossover Molecules



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



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

Motivation:

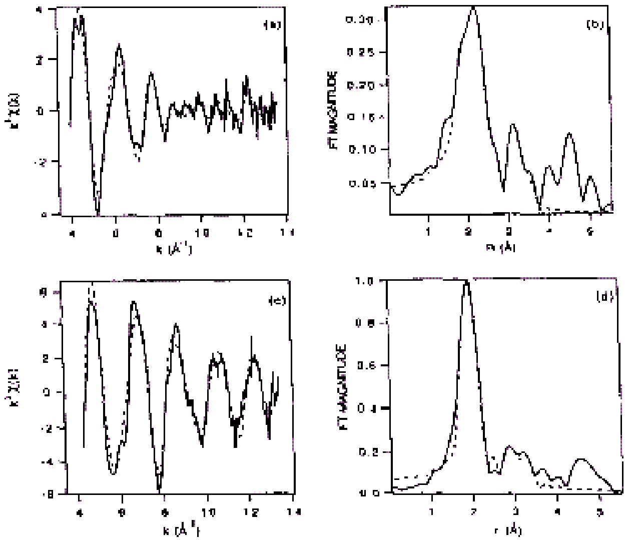
- relationship between structure, electronic, and magnetic properties

  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} Static Structure - EXAFS

trap high-spin state at low temperatures

EXAFS (Fe K-edge)



T (K)	Atom pair	N	R (Å)	σ^2	R_c (Å) [a]	Angle (°)
298	Fe–N	2	2.02	0.0055	2.06	
	Fe–N	4	2.19	0.0070	2.20	
	Fe–C	4	3.04	0.0050	3.02	159
	Fe–C	2	3.20	0.0080	3.20	130
	Fe–C	4	3.20	0.0080	3.20	126
90	Fe–N	1.9	1.96	0.0050	1.96	
	Fe–N	3.5	1.98	0.0050	2.01	
	Fe–C	3.5	2.88	0.0030	2.83	166
	Fe–C	1.9	3.00	0.0030	3.04	132
	Fe–C	3.5	3.00	0.0030	3.04	123

G. Sankar et al., *Chem. Phys. Lett.*, **251**, 79, 1996.

NEXFS (Fe L-edge)

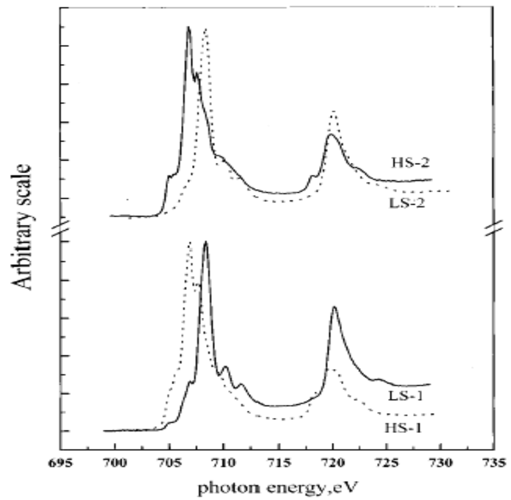


Figure 5. Iron $L_{II,III}$-absorption edge of (a, bottom) HS-1/LS-1 (17 K) and (b, top) HS-2 (17 K)/LS-2 (70 K) for the extraction sample.

J.-J. Lee et al., *JACS*, **122**, 5742, 2000.

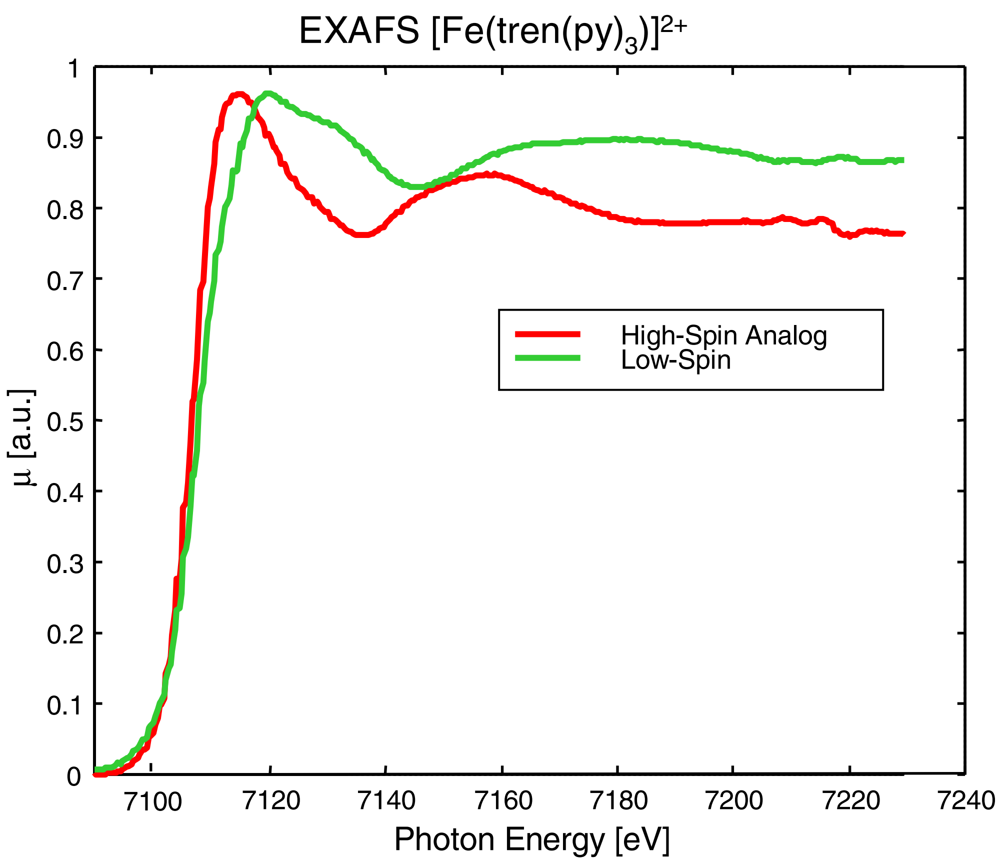
High-spin vs. Low-spin:

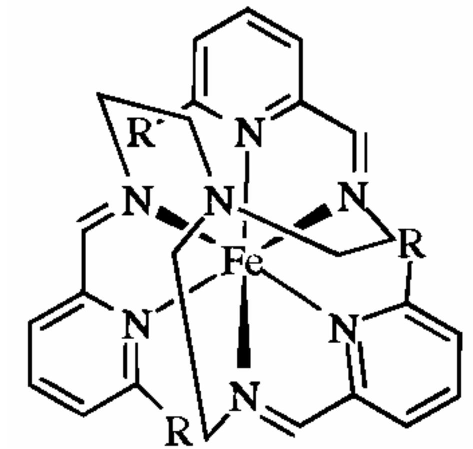
Electronic structure – NEXFS Fe L-edge

Atomic structure – EXAFS Fe K-edge

EXAFS Measurements - Fe^{II} Spin-Crossover Molecules

(ALS Beamline 5.3.1)





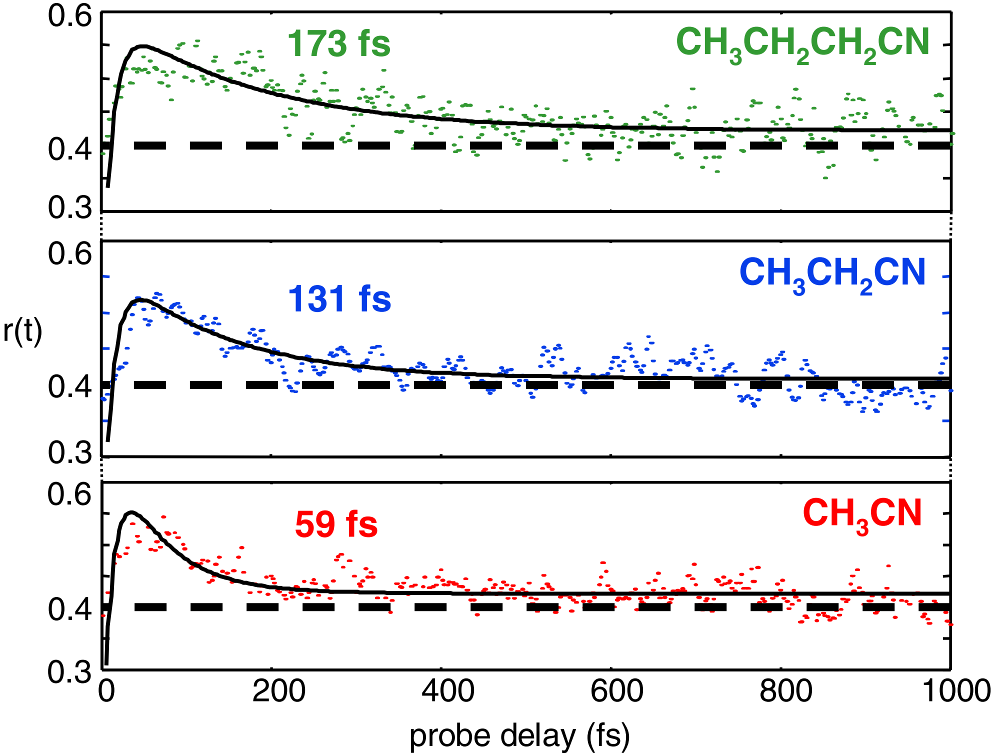
R = H (low spin)

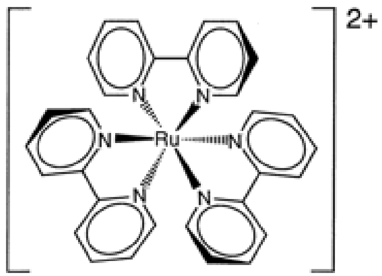
R = CH_3 (high spin)

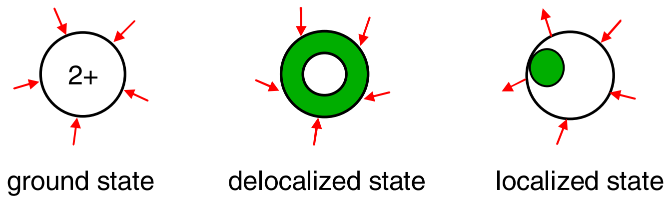
Ultrafast Chemical Reactions - Solvent Dependence

Ru – charge-transfer complex $[Ru(bpy)_3]^{2+}$

Femtosecond Polarization Anisotropy Measurements (visible)







solvent	τ_{exp}	R_τ	R_I	I (amu Å^2)
$CH_3CH_2CH_2CN$:	173 fs	2.9	3.2	142
CH_3CH_2CN:	131 fs	2.2	1.8	78.6
CH_3CN:	59 fs	1	1	44.4

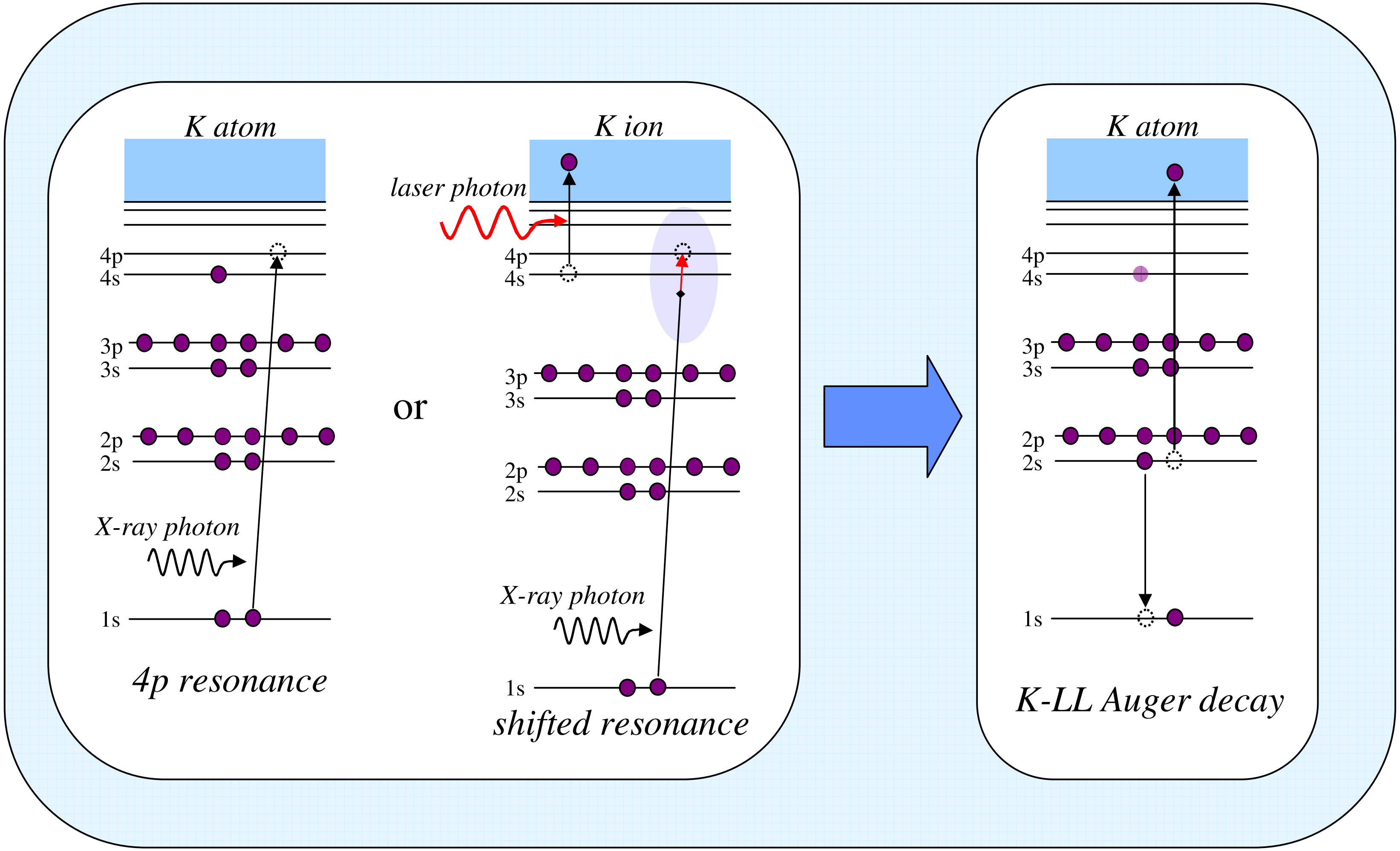
Yeh, Shank, and McCusker, *Science*, **289** (2000)

Femtosecond dynamics of excited-state evolution in [Ru(bpy)3]$^{2+}$
Damrauer, Cerullo, Yeh, Boussie, Shank, and McCusker, *Science*, **275**, 54, (1997).

Laser Modification of 1s Electron Binding Energy in Potassium

ALS Beamline 5.3.1

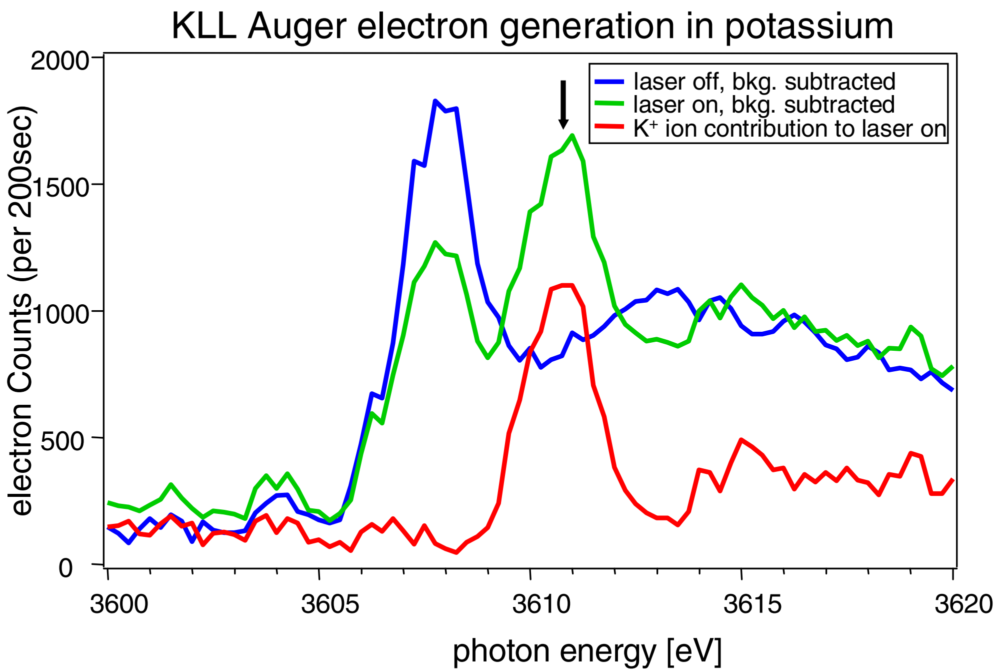
M. Hertlein, B. Feinberg, N. Neumann, K. Cole, H. Adaniya, J. Maddi, M. Prior, T. Osipov, and A. Belkacem

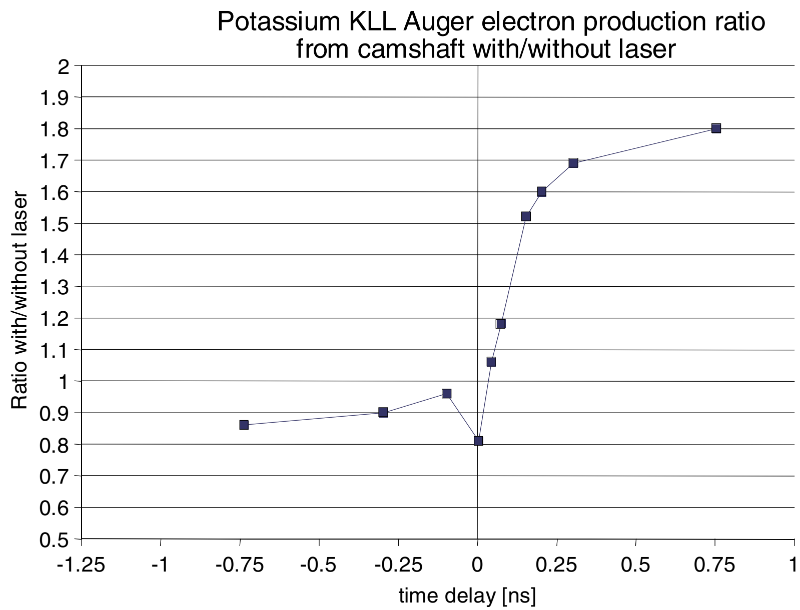


Potassium K-edge Shift after Femtosecond Laser Excitation

ALS Beamline 5.3.1

M. Hertlein, B. Feinberg, N. Neumann, K. Cole, H. Adaniya, J. Maddi, M. Prior, T. Osipov, and A. Belkacem





X-ray photon energy: 3610eV (at maximum of shifted resonance)

- Auger processes – high charge states
- excitation energy → charge state distribution
  (2 photon process: x-ray + laser)
- dynamics of post collision interactions and Auger decay

X-rays for Ultrafast Structural Dynamics

Characteristics for Ideal Source

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

(2) high average flux >10^8 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)

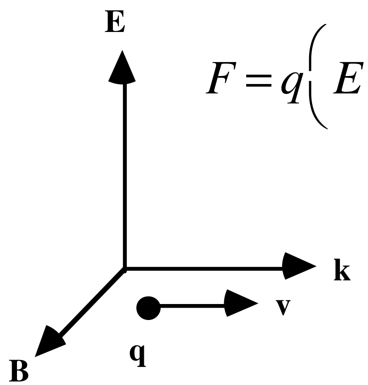
(4) rep. rate: 100 Hz - 10 kHz
- signal averaging, sample damage, sample replacement

stability – pulse amplitude, alignment

variable polarization – x-ray dichroism (magnetic materials, chiral molecules)

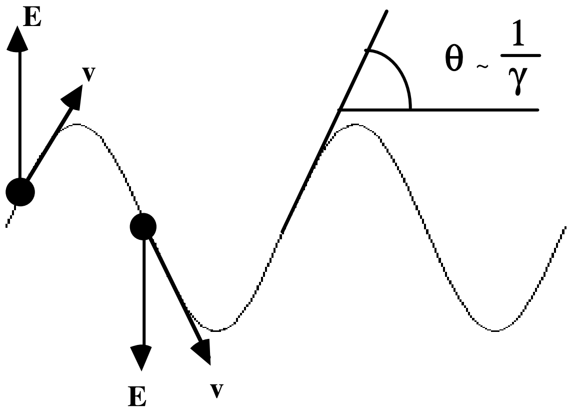
Wiggler as a Kicker

In slicing, transverse kickers are not effective
due to the cancellation of the transverse forces from
electric and magnetic fields.



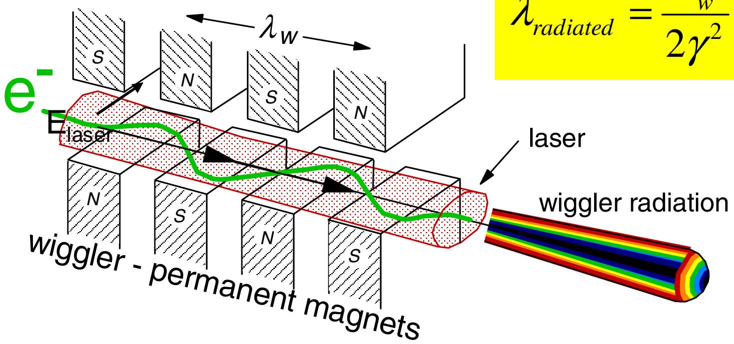
$$F = q\left(E - \frac{V}{C} \times B\right) \Rightarrow \frac{qE}{2\gamma^2}$$

Wiggler gives the energy "kick"



$$\lambda = l\,\frac{1}{2\gamma^2}\left(1 + \gamma^2\theta^2\right)$$

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:

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

wiggler radiated energy:

$$A_R \cong \frac{K^2}{1+K^2} 4\alpha\hbar\omega_R$$

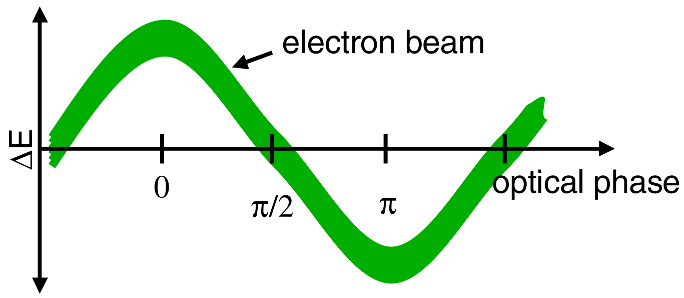
Laser requirements:

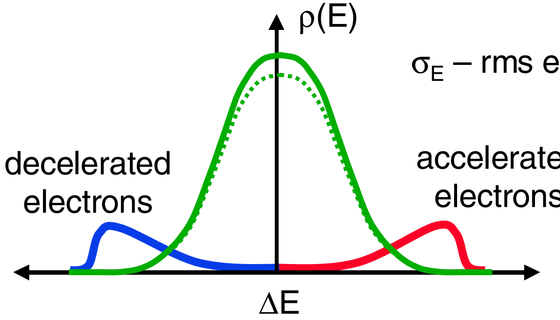
$h\omega_L = 1.55$ eV

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

$A_L = 610$ μJ

$\Delta E \cong 17$ MeV



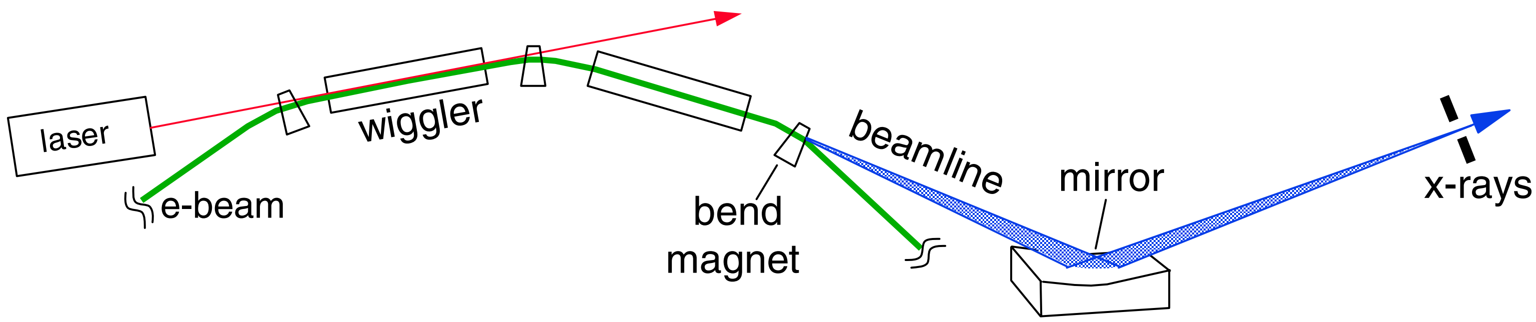


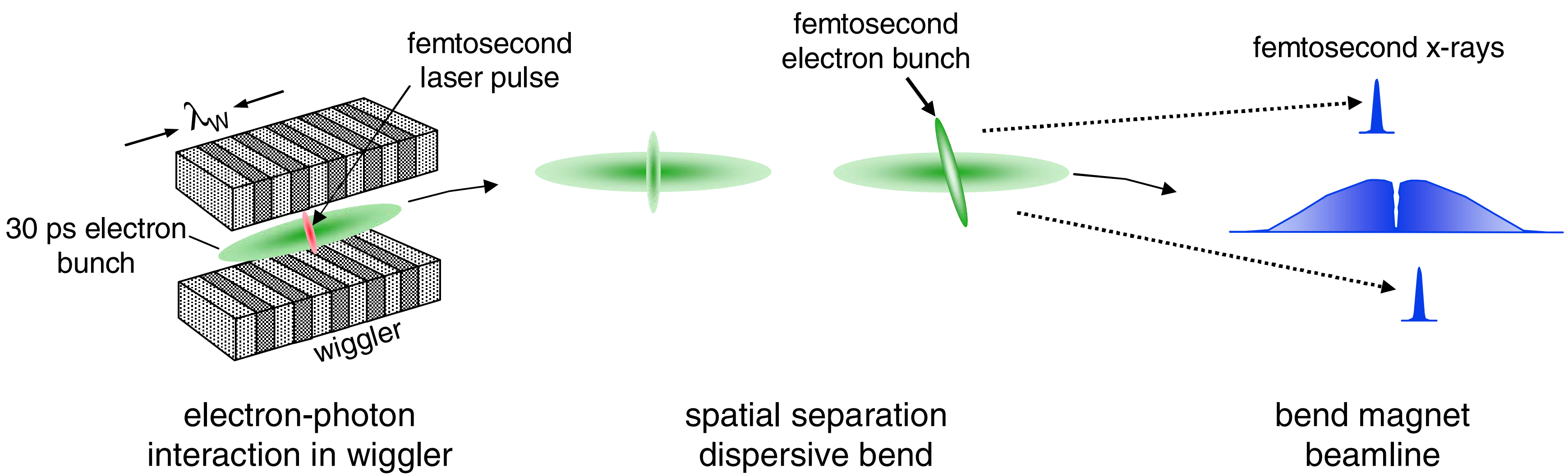
ALS beam energy spread σ_E ~1.9 MeV E_o =1.9 GeV

since $\sigma_E \sim E_o^2$ and we want $\Delta E \sim \sigma_E$
then the required laser pulse energy
scales as: $A_L \sim E_o^4$

Generation of Femtosecond X-rays from the ALS

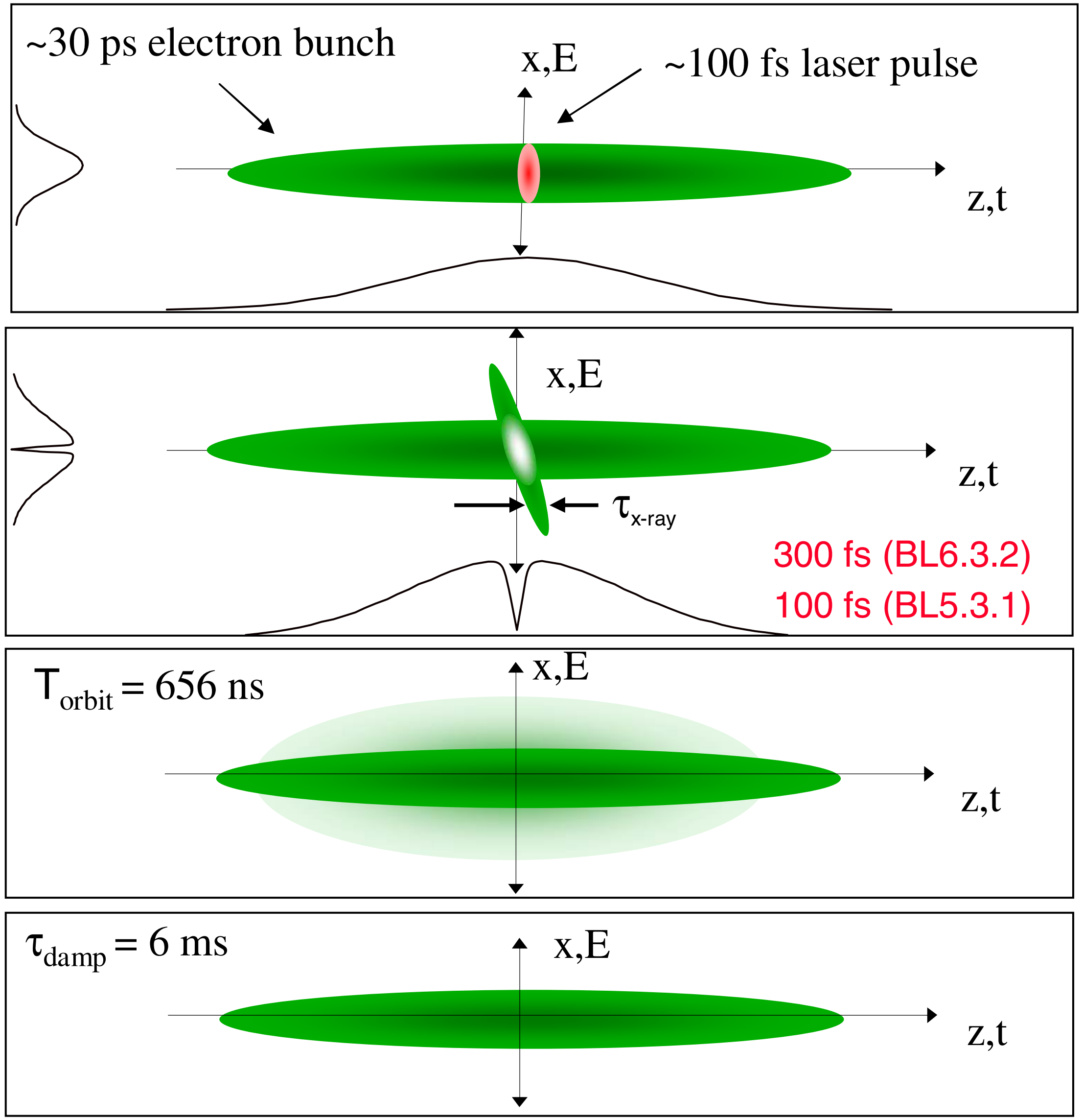
Zholents and Zolotorev, *Phys. Rev. Lett.*, **76**, 916,1996.



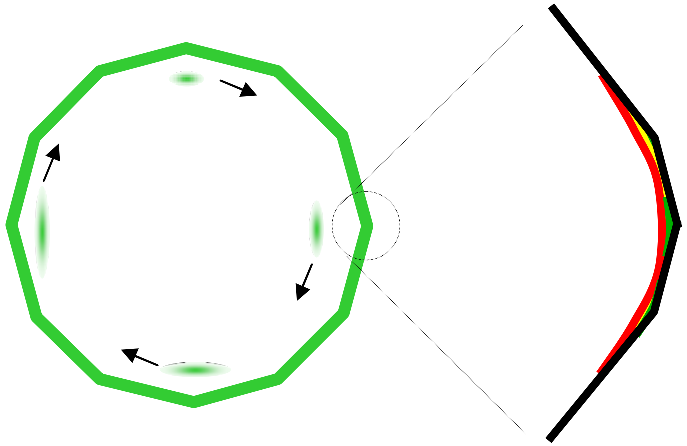


R. W. Schoenlein, S. Chattopadhyay, H. H. W. Chong, T. E. Glover, P. A. Heimann,C. V. Shank, A. A. Zholents, and M. S. Zolotorev, *Generation of Femtosecond Pulses of Synchrotron Radiation,* Science, Mar 24, 2000: 2237-2240.

Dynamics of Modulated Electron Beam

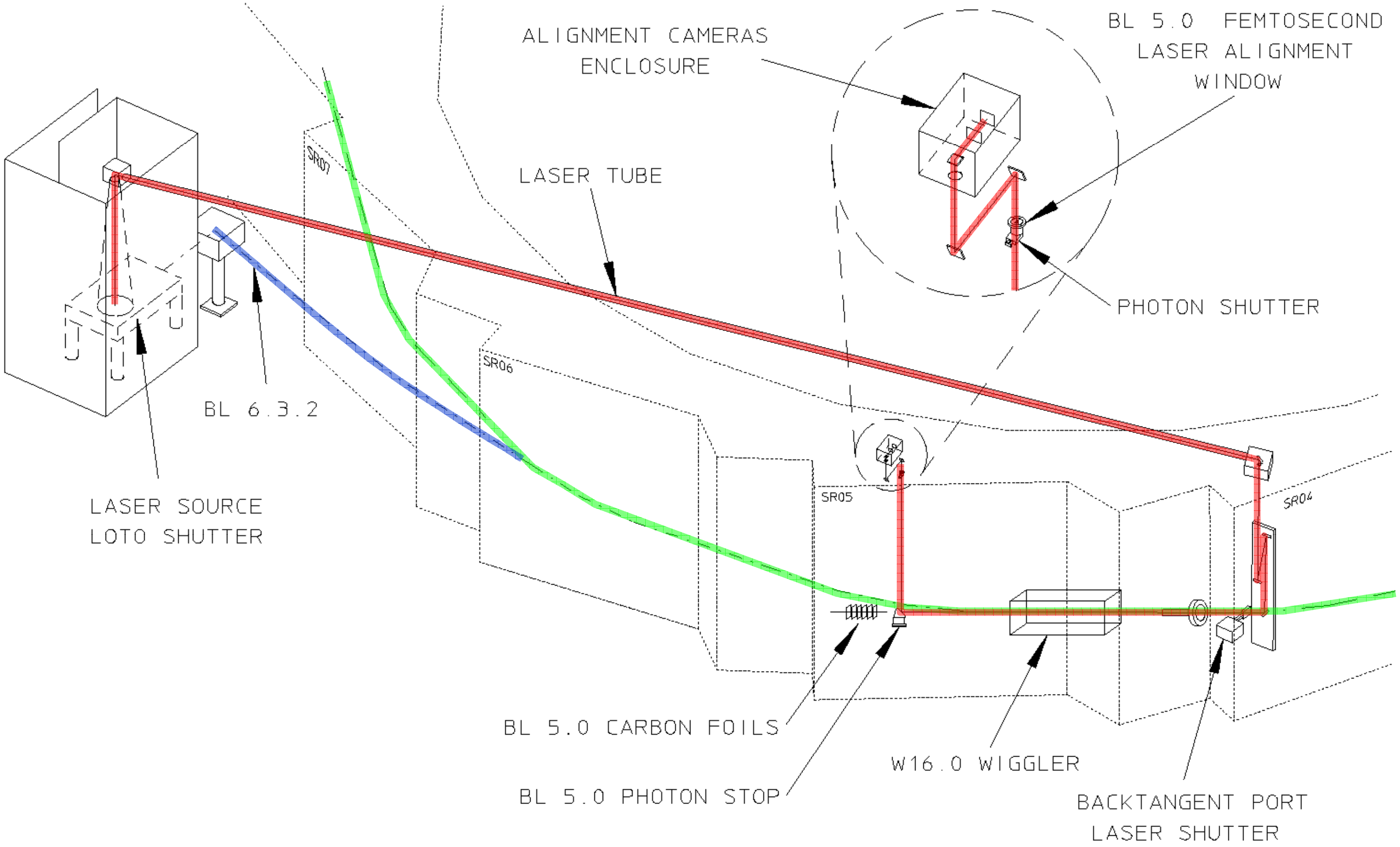


ALS Storage Ring Dispersion

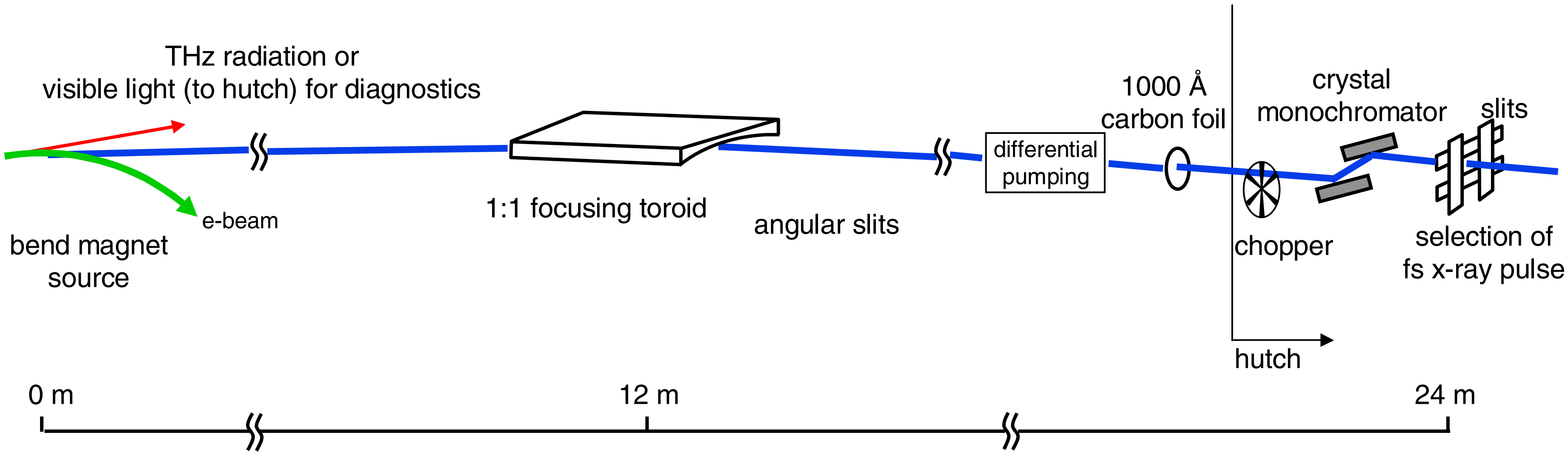


$\sigma_{dispersion}$ ~ 85 fs per full arc
(200 fs FWHM)

Synchrotron Beam Slicing - Layout



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

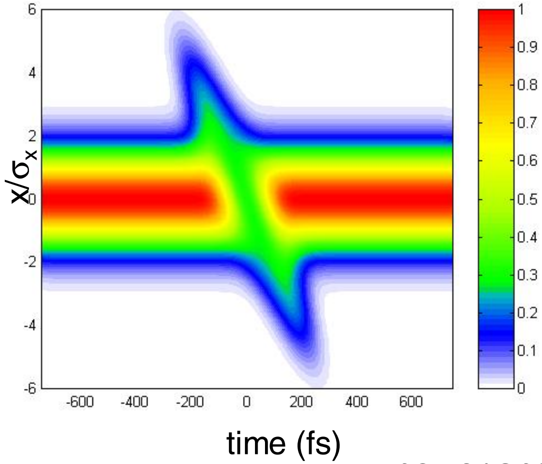
brightness ~10^8 ph/s/mm^2/mrad^2/0.1% BW

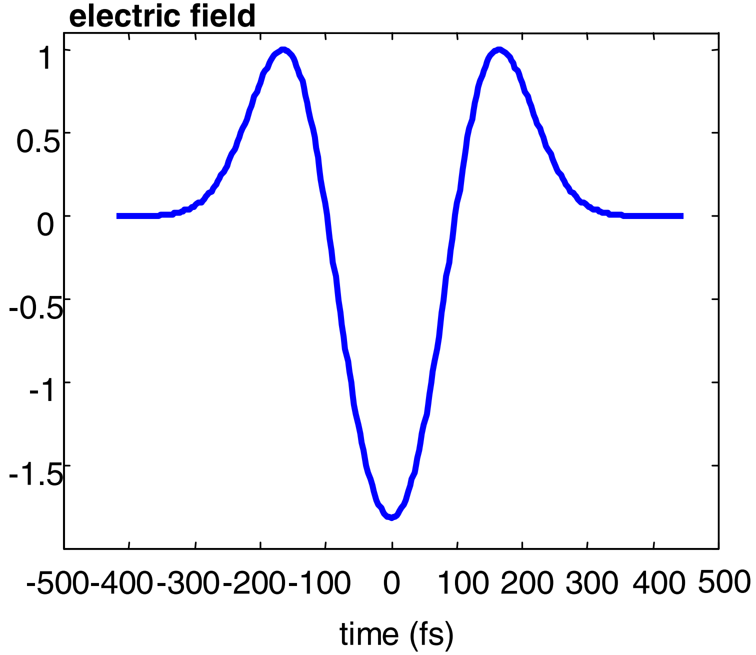
100 fs pulse duration

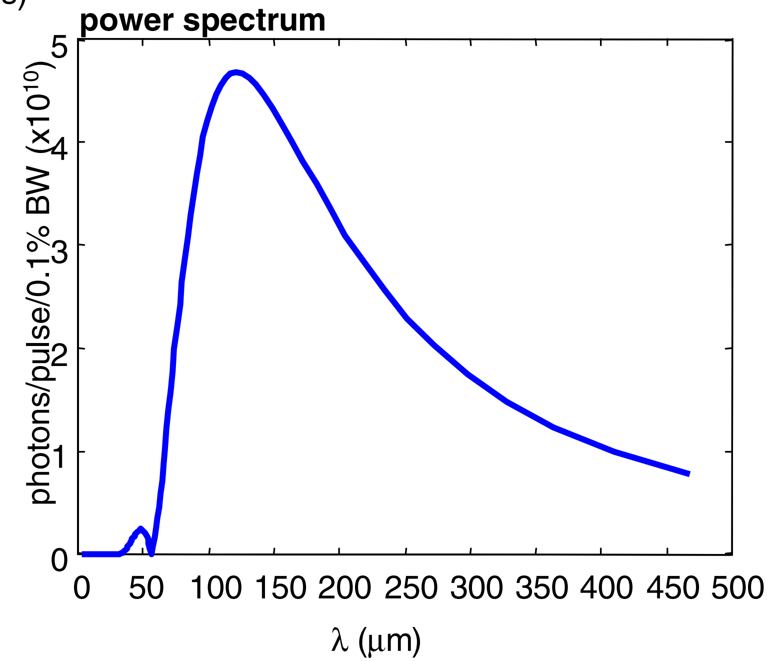
(5 kHz repetition rate)

Coherent Infrared Synchrotron Radiation

laser modulated e-bunch distribution

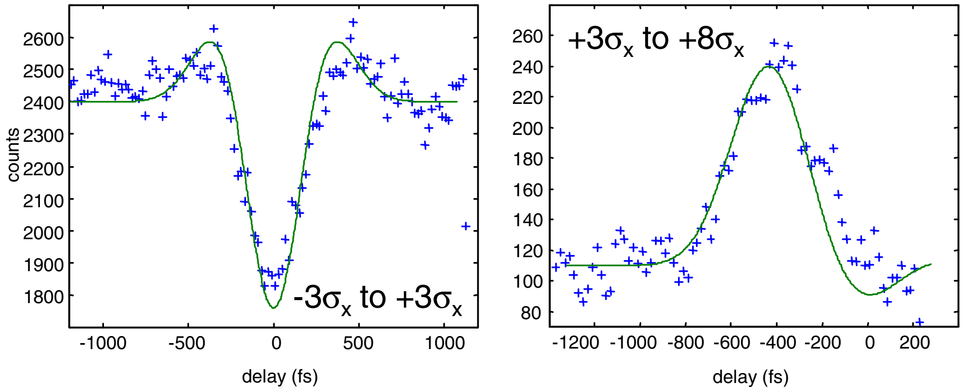




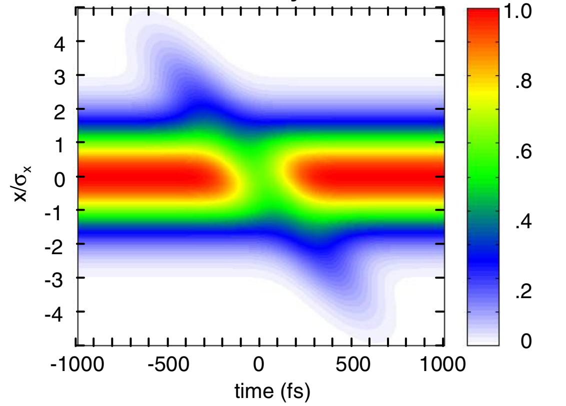


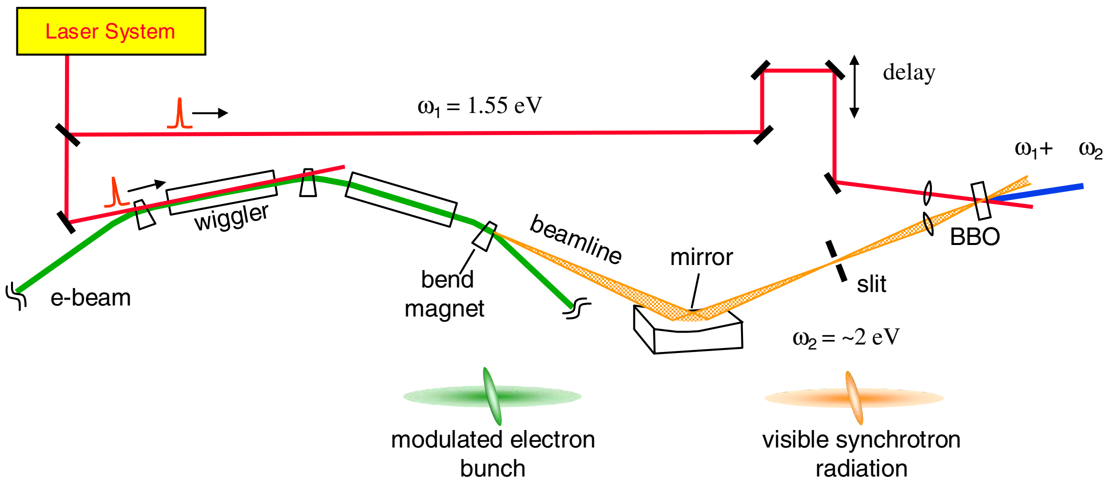
Femtosecond Pulses of Synchrotron Radiation

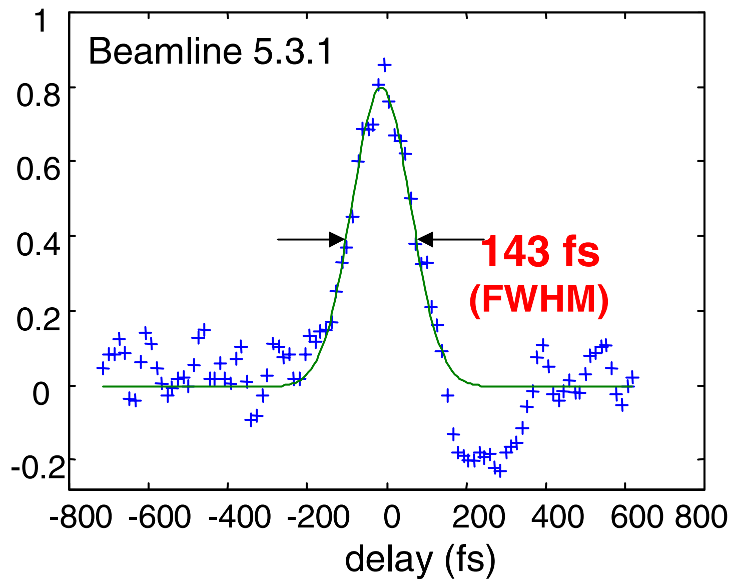
Demonstration Measurements – Beamline 6.3.2



Calculated
Electron Density Distribution

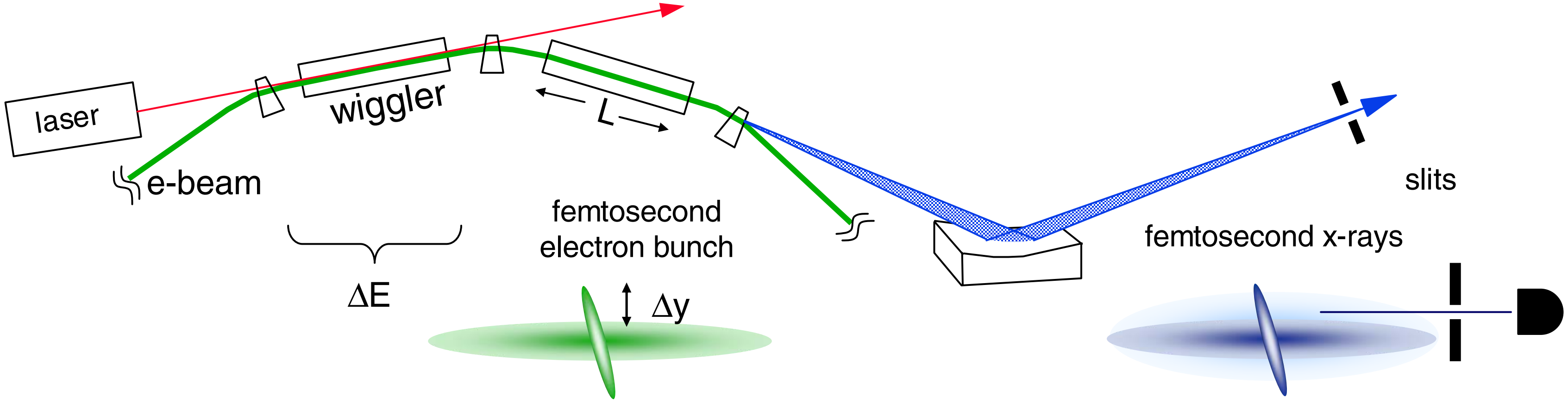






Schoenlein et al., *Science*, **287**, 2237 (2000)

Separation of Femtosecond X-rays



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

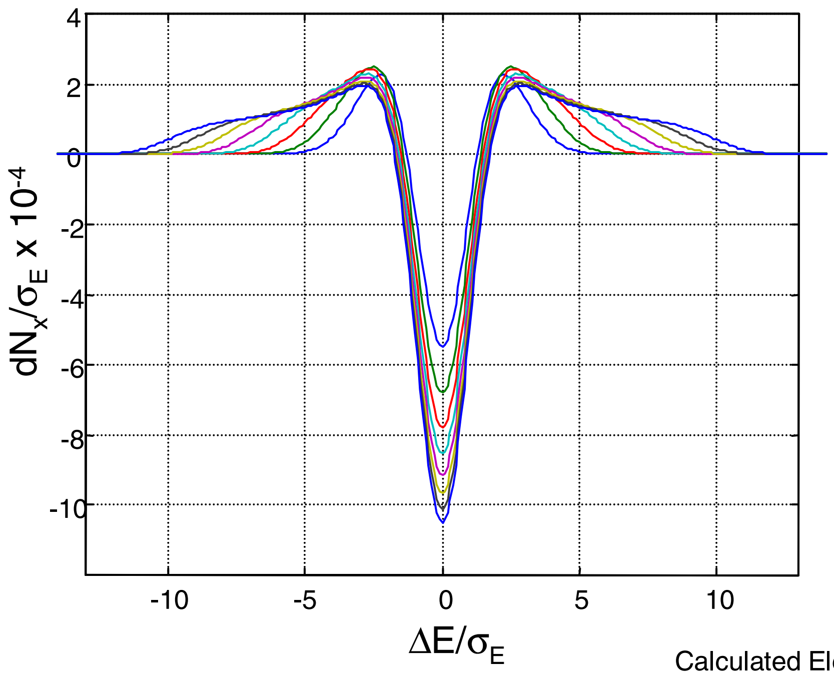
$$\frac{\text{Sig (fsec)}}{\text{background}} > 1$$

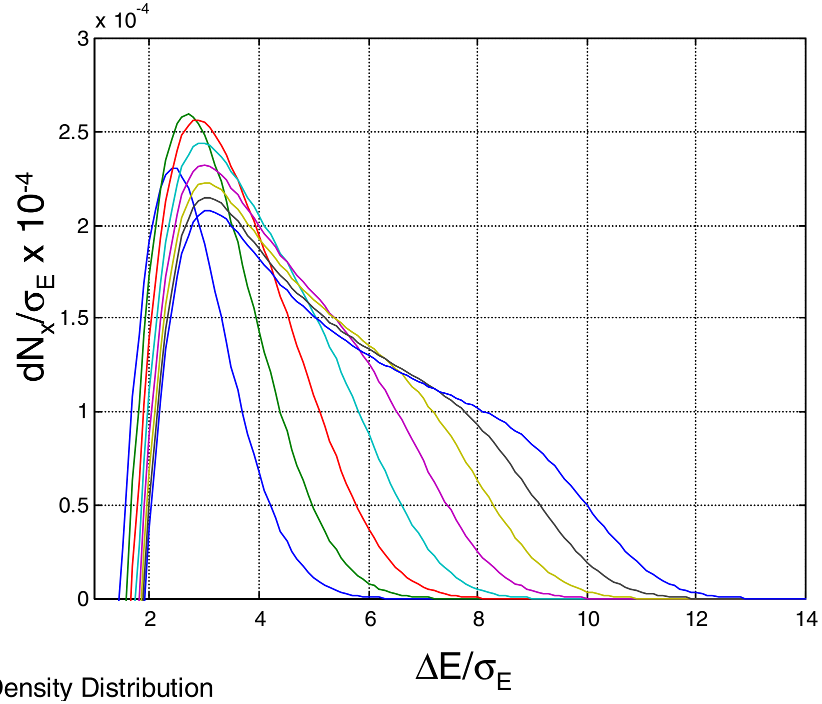
Requirement – gated detectors (2 nsec) for isolating individual bunches
avalanche photodiodes
gated microchannel plates

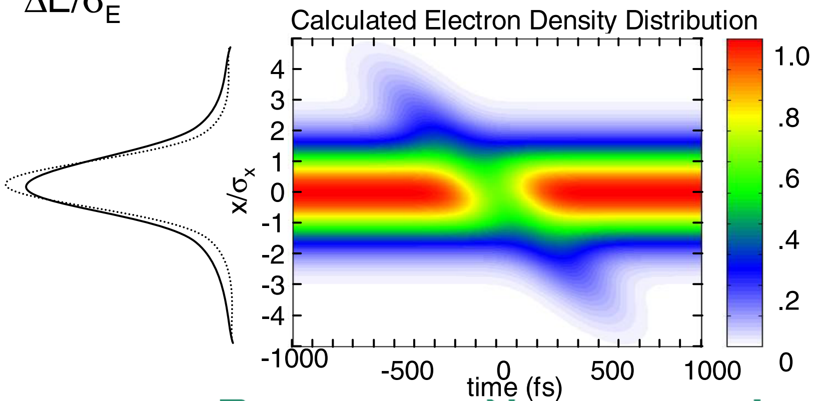
Differential Beam Profiles

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

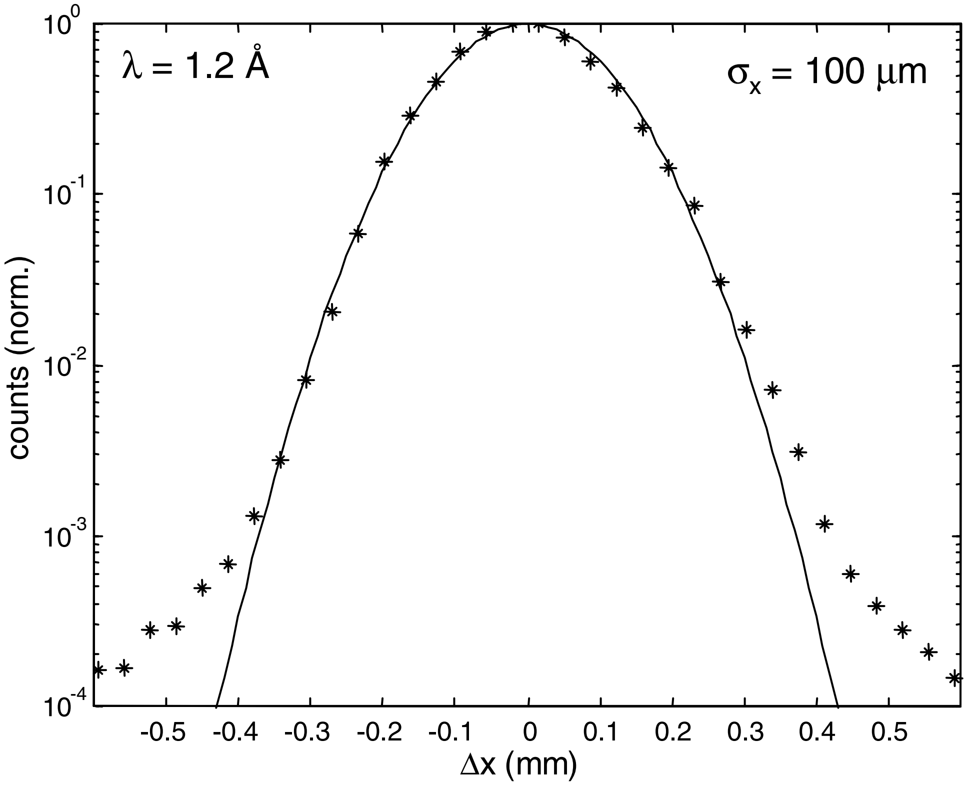
Modulation: $3\sigma_E$ to $10\sigma_E$ $\quad \tau_{laser}/\tau_e = 10^{-3}$

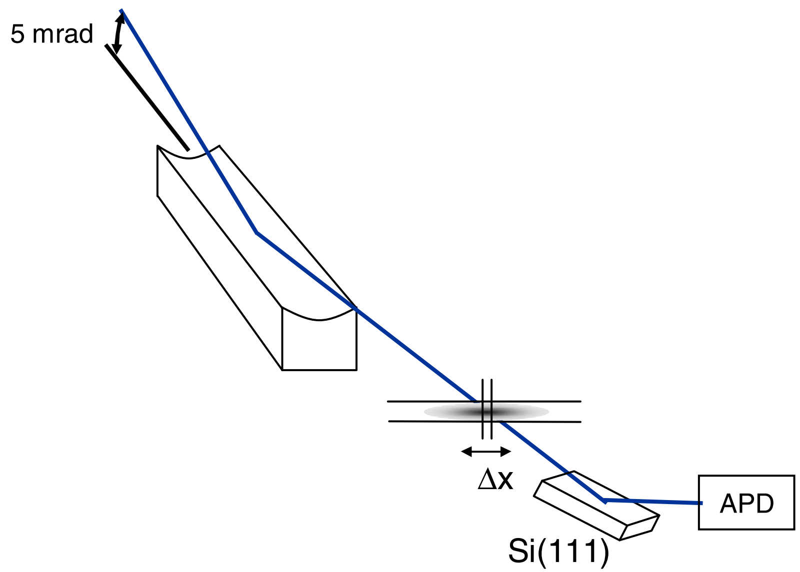






Beam Profile Measurements – BL5.3.1 Camshaft Bunch

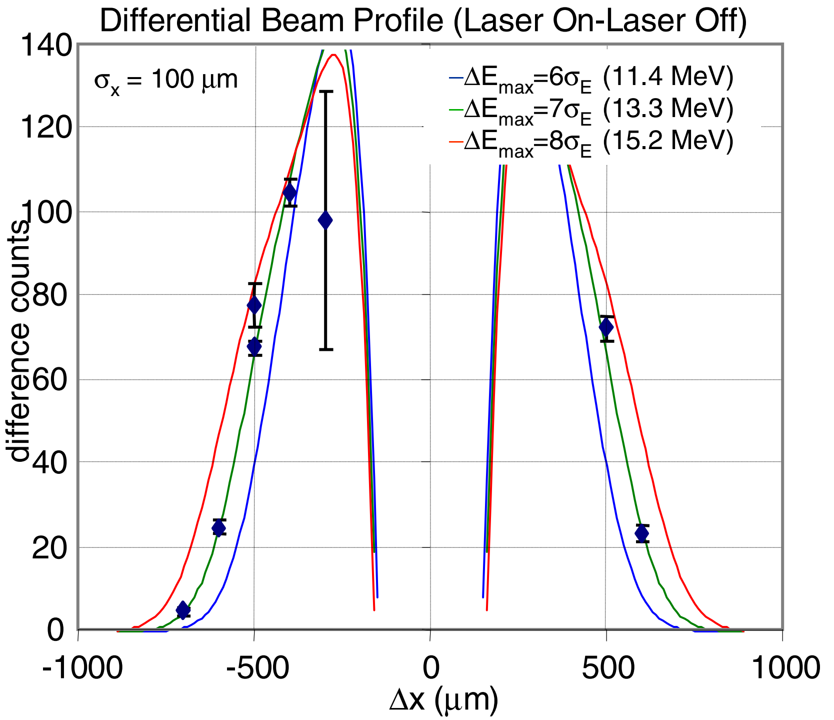


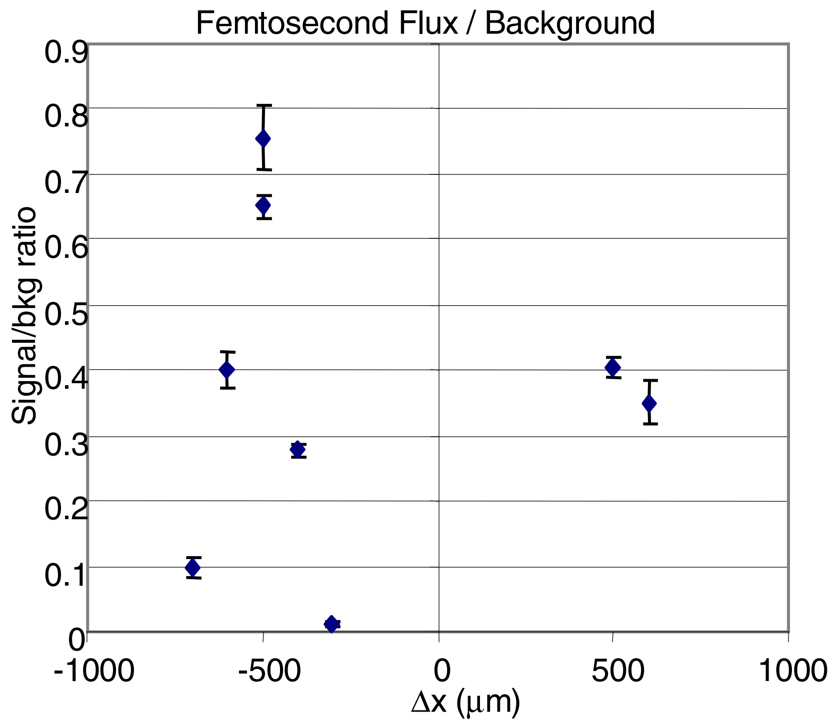


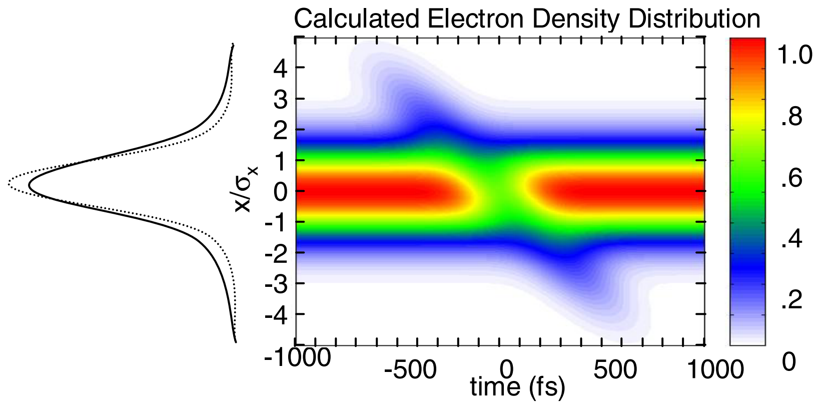
- fraction of e-beam that is modulated = 75 fs $\times$ 0.1 /75 psec = 10^{-4}

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

Femtosecond X-ray Profile Measurements – BL5.3.1







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

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)

  $f_{laser} / f_{synchrotron}$
  (40 kHz) (500 MHz)

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

Average Femtosecond X-ray Flux ~ Average Femtosecond Laser Power

Bend Magnet

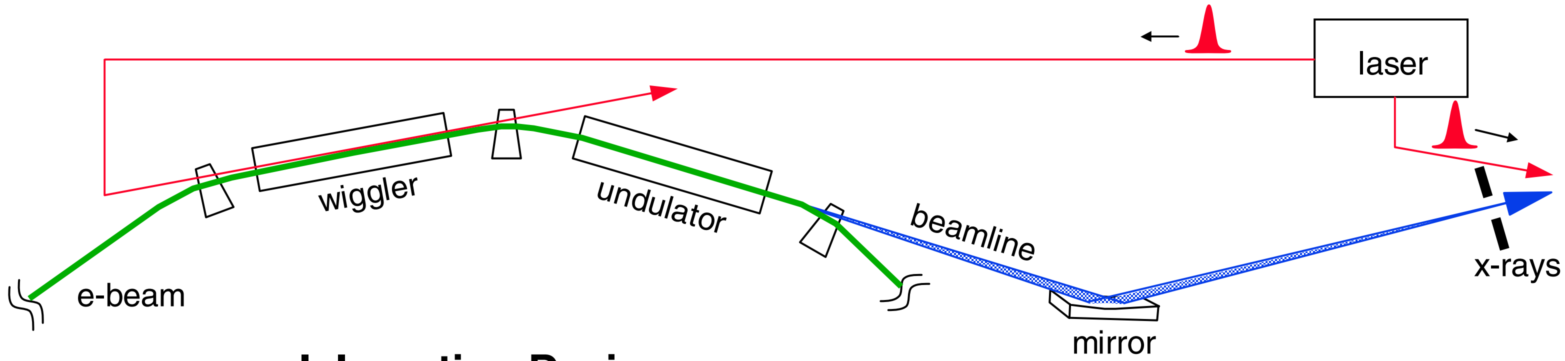
- flux ~10^{13} ph/sec/0.1% BW
- brightness ~10^{16} ph/sec/0.1% BW

Undulator

- flux ~10^{15} ph/sec/0.1% BW
- brightness ~10^{19} ph/sec/0.1% BW

Femtosecond Undulator Beamline – Overview

New beamline(s) – funded by DOE Basic Energy Sciences
Operation with femtosecond x-rays – early 2005



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

- 60 W (1.5 mJ per pulse, 40 kHz)

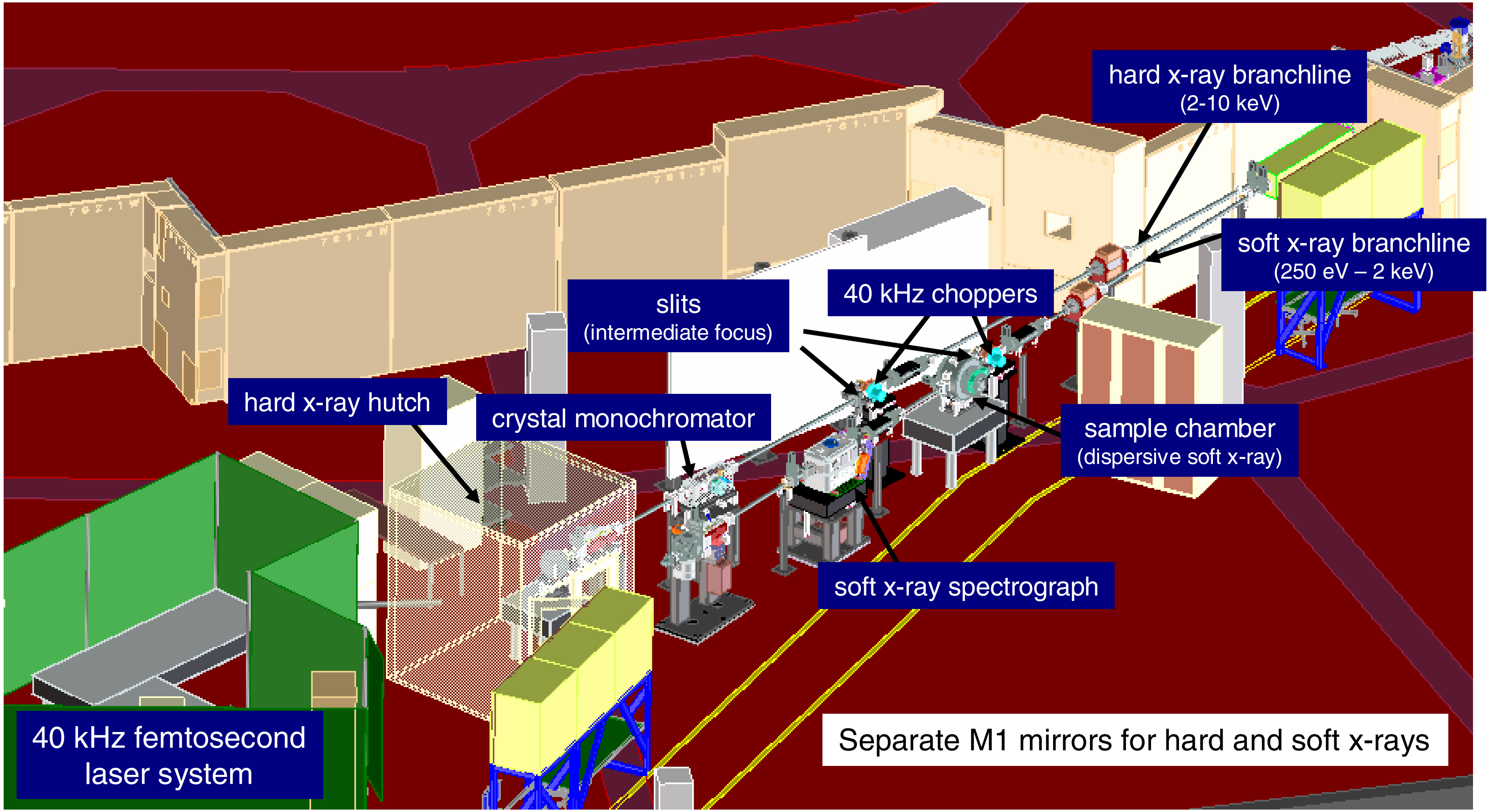
  x10 increase in flux

IV. Storage Ring Modifications

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

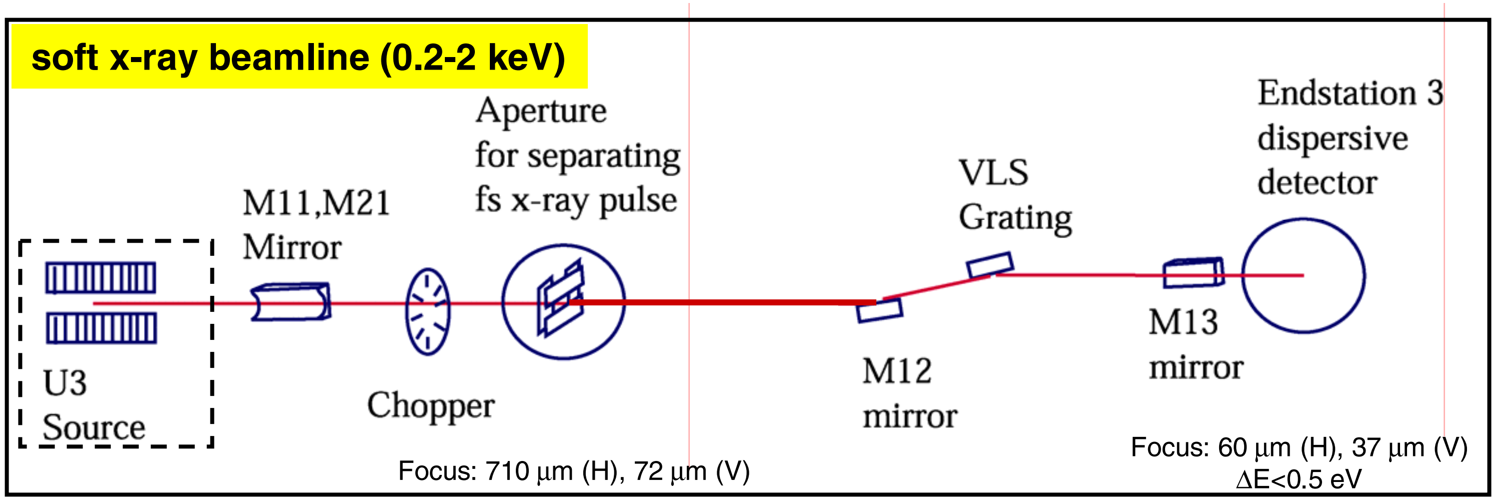
Femtosecond Undulator Beamline 6.0 Layout

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

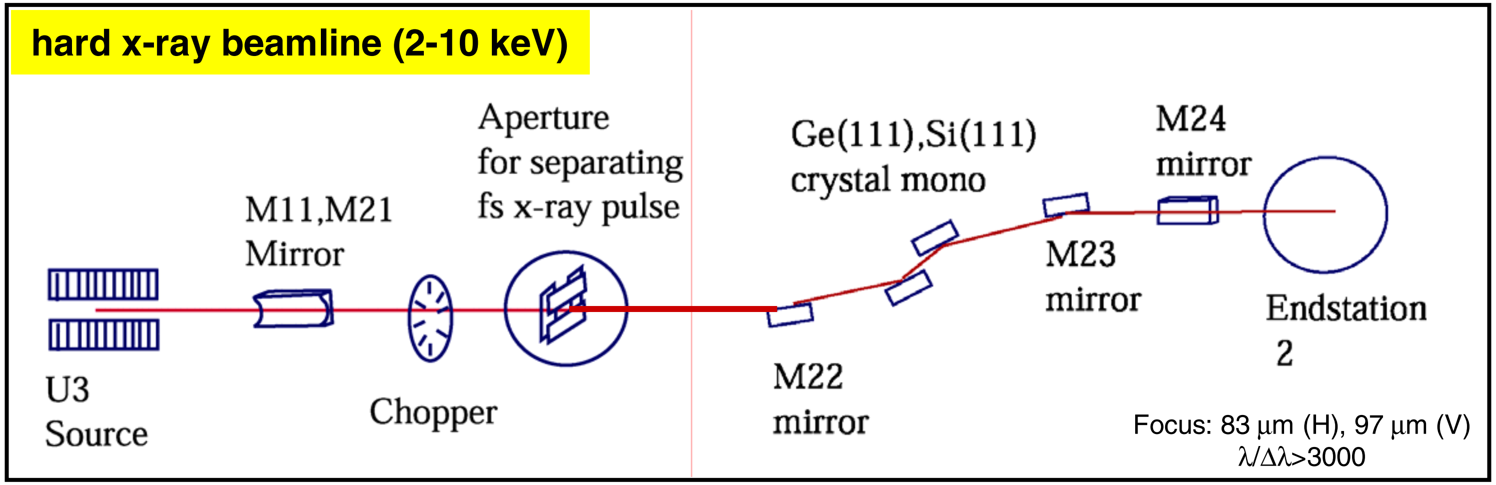


ALS Femtosecond Undulator Beamlines

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



commission April 2005

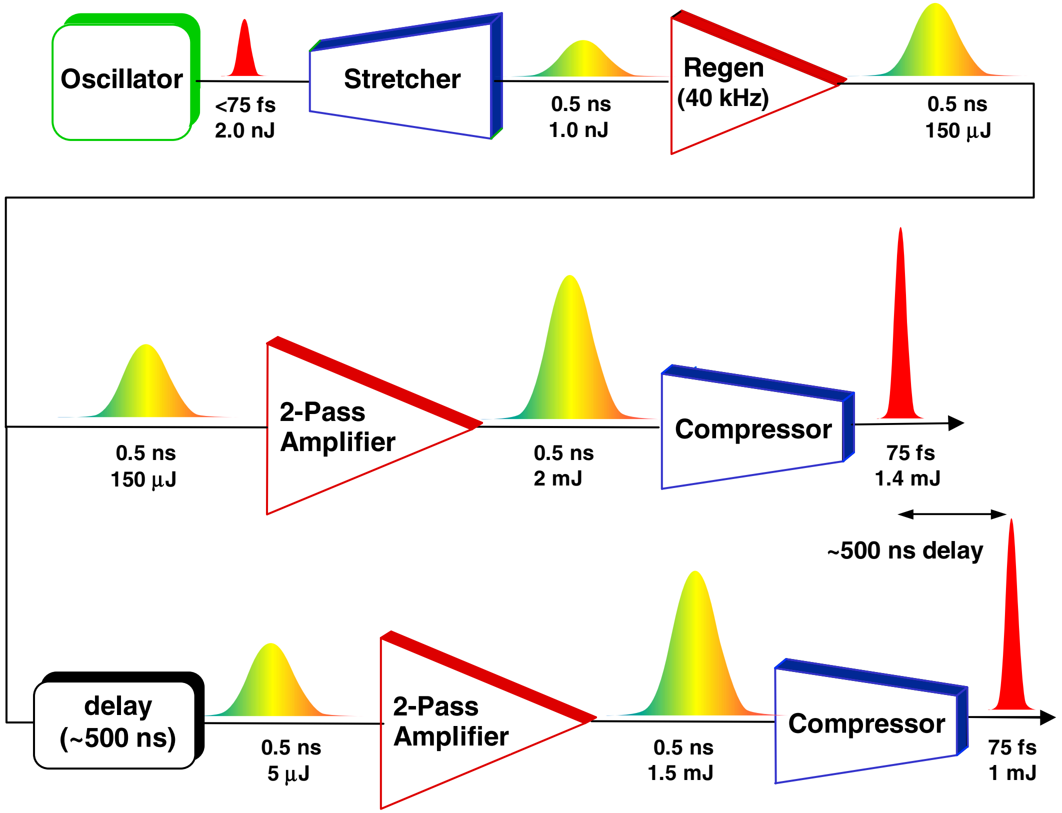


commission Sept. 2005

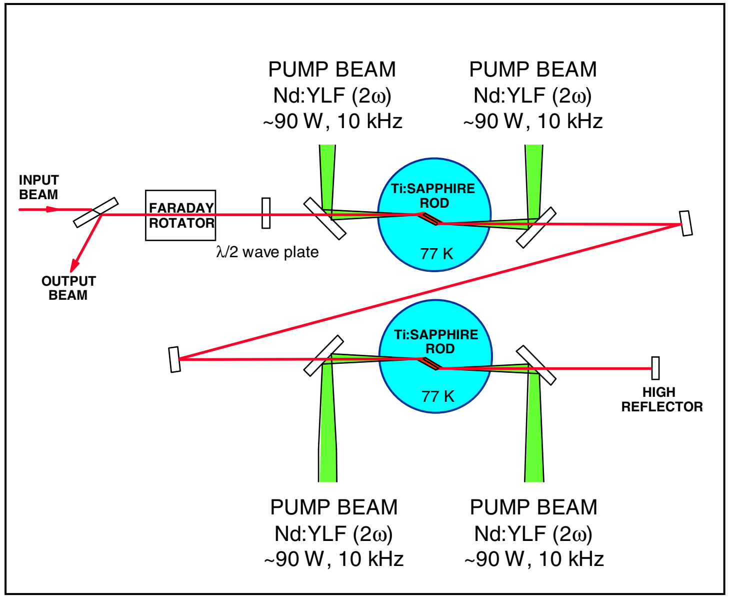
Femtosecond Laser System

Electron beam interaction requirements:
~1.5 mJ pulse energy, 50 fs FWHM, at ~800 nm
40 kHz repetition rate, 60 W average power
diffraction limited focusing, beam parameter: M^2~1.1

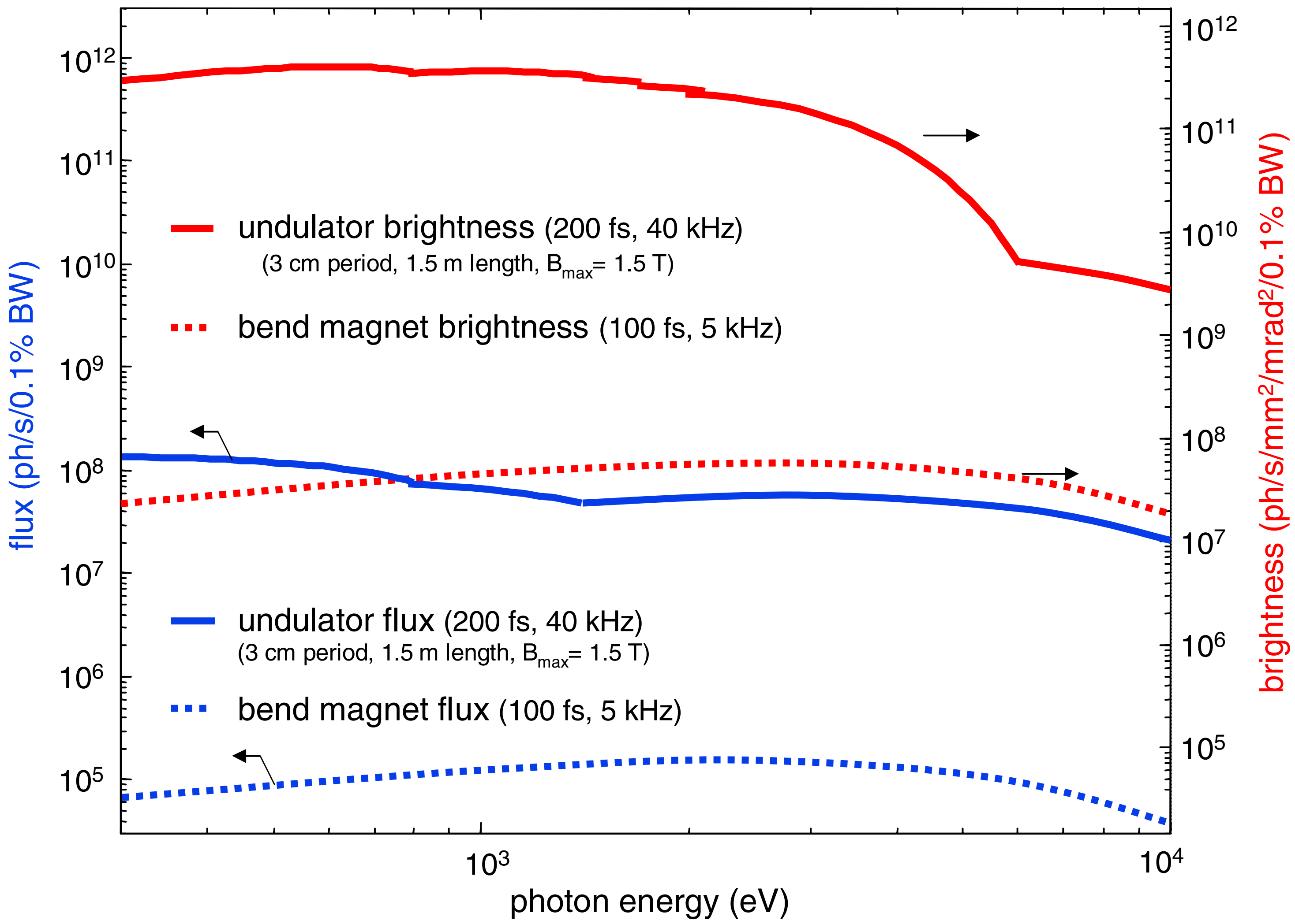
Excitation pump pulses for time-resolved experiments:
~1 mJ pulse energy at 800 nm (OPA)
50 fs pulse duration, 40 kHz repetition rate
~500 ns relative delay



cryogenic power amplifier

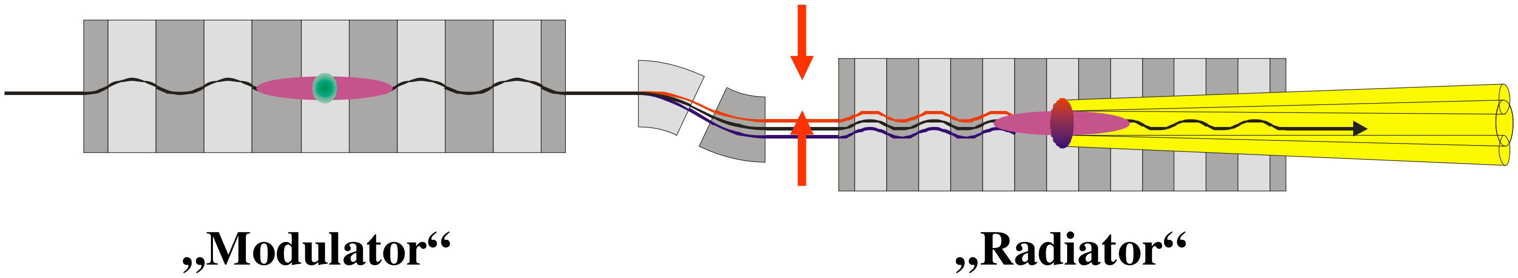


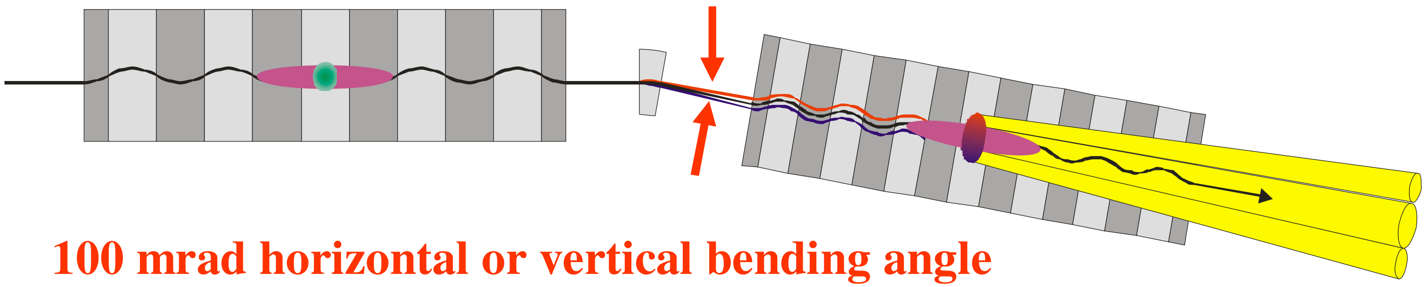
Femtosecond X-ray Flux and Brightness



Femtosecond X-ray Beamline at BESSY

S. Khan et al., PAC 1997, p.1810; EPAC 2002, p.700.



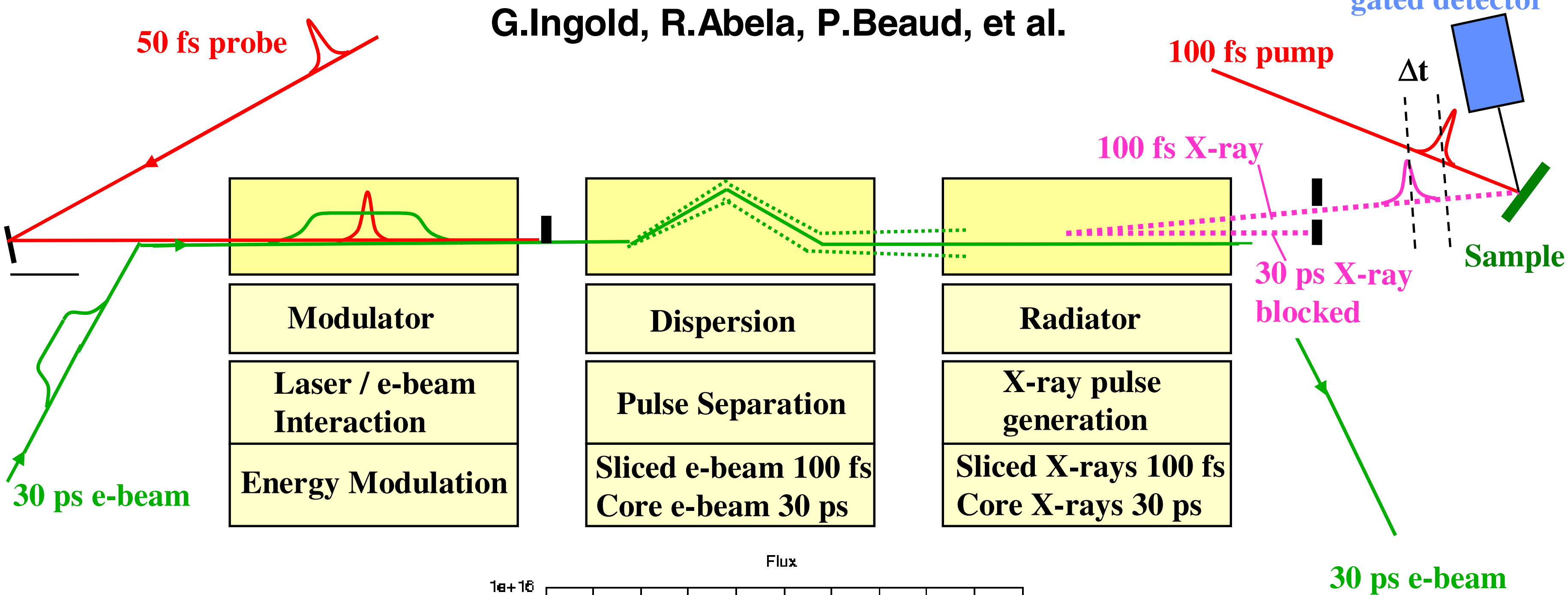


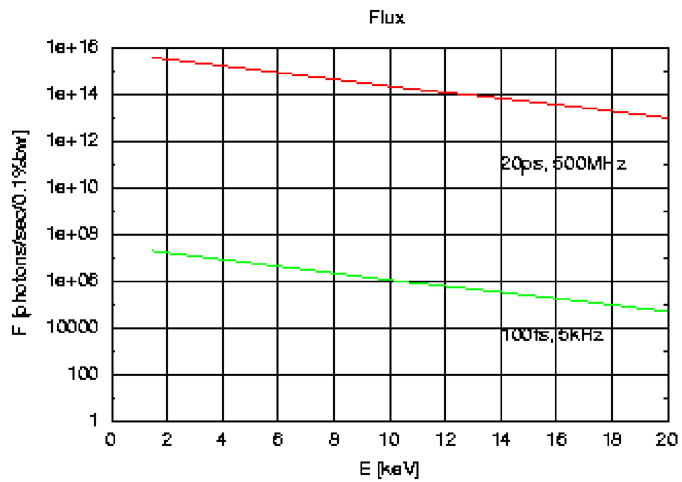
100 mrad horizontal or vertical bending angle

- angular separation → minimum background (~10:1)
- *one* straight section → minimum pulse lengthening (~50 fs)
- elliptical undulator → linear/circular polarization
- expected flux: ~10^6 photons/sec/0.1% BW (5 kHz, <2 keV)

SLS FEMTO Project

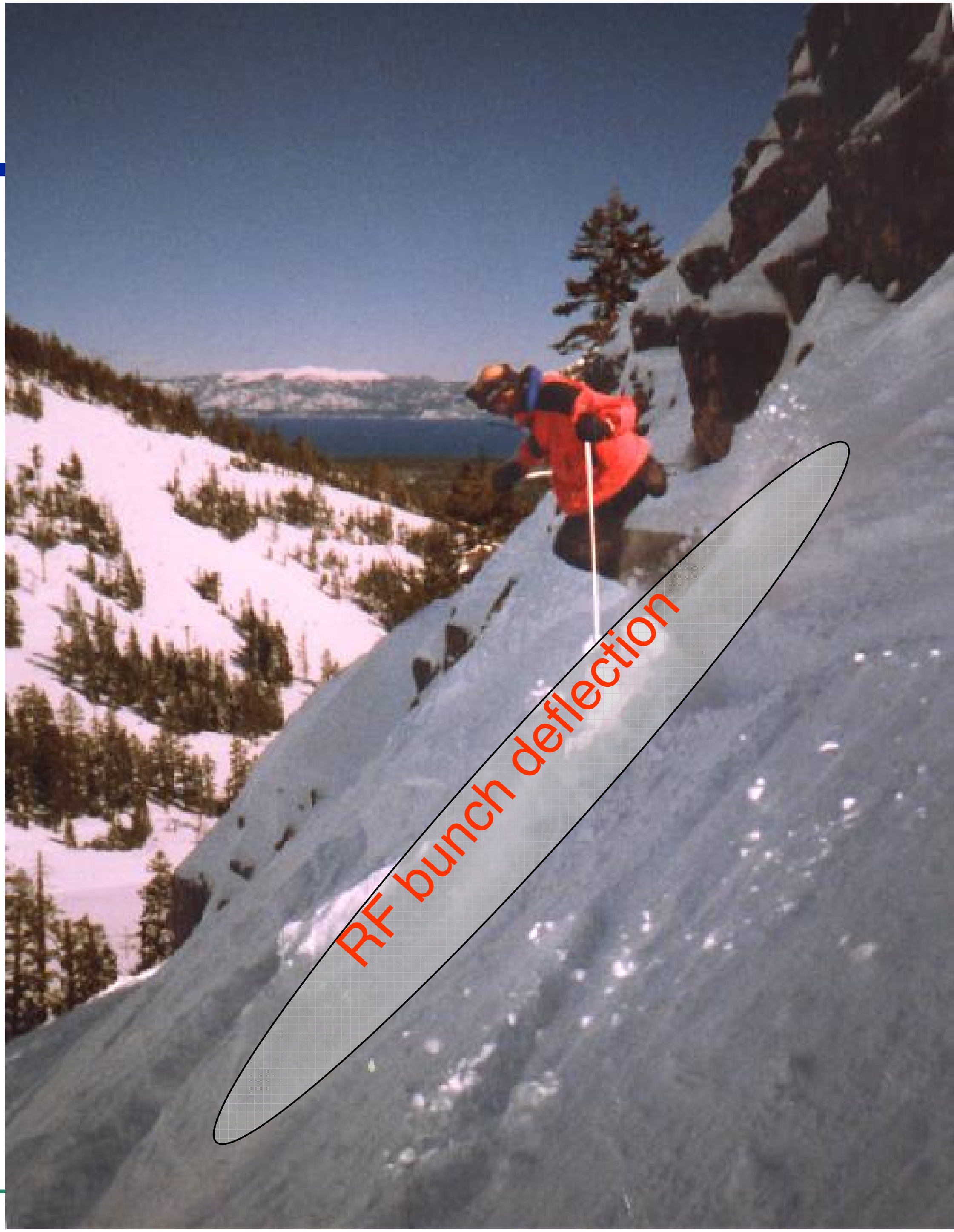
G.Ingold, R.Abela, P.Beaud, et al.



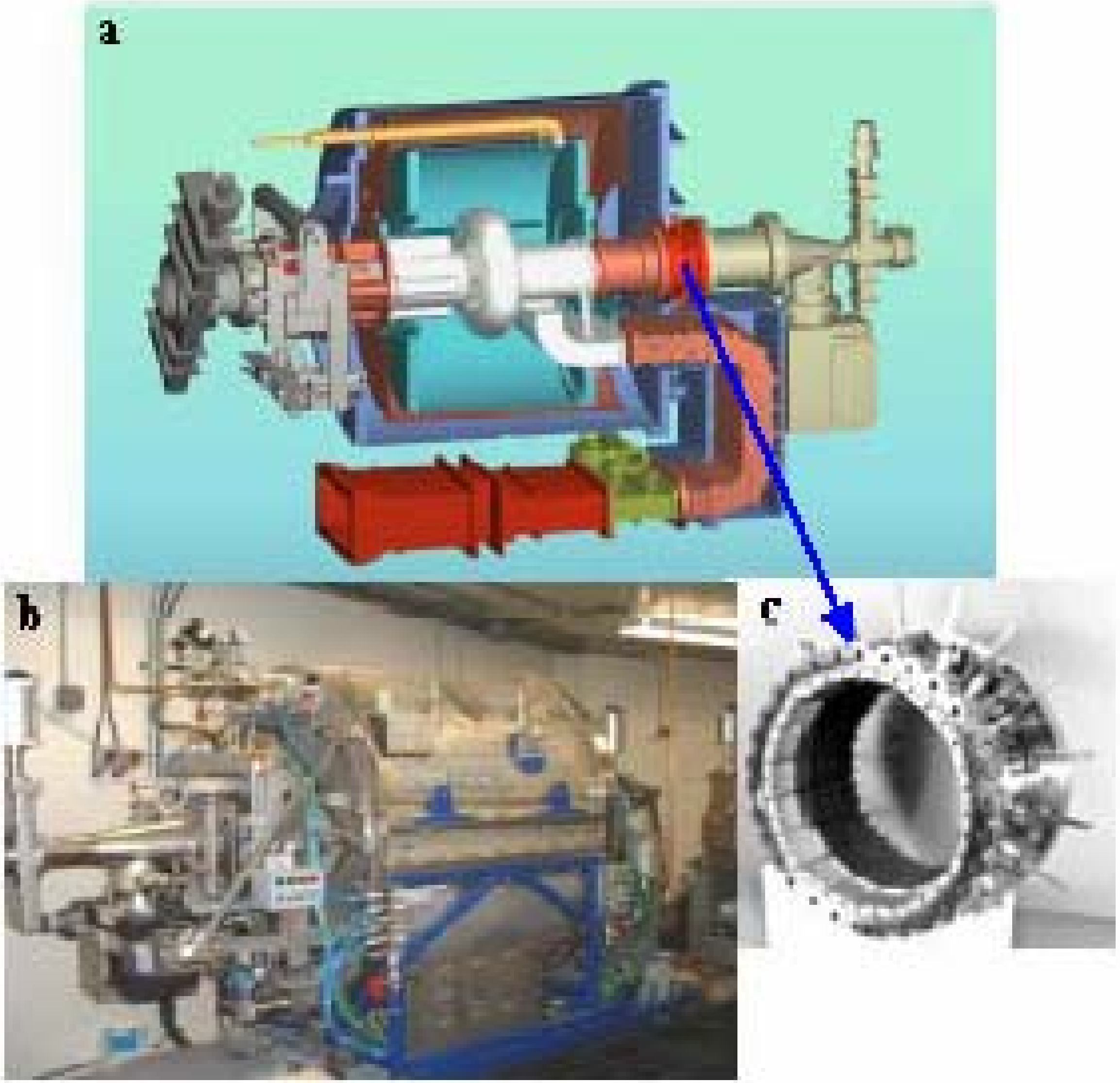


A. Zholents, P. Heimann, M. Zolotorev, J. Byrd, NIM. A (425)1-2 (1999) pp.385-389.

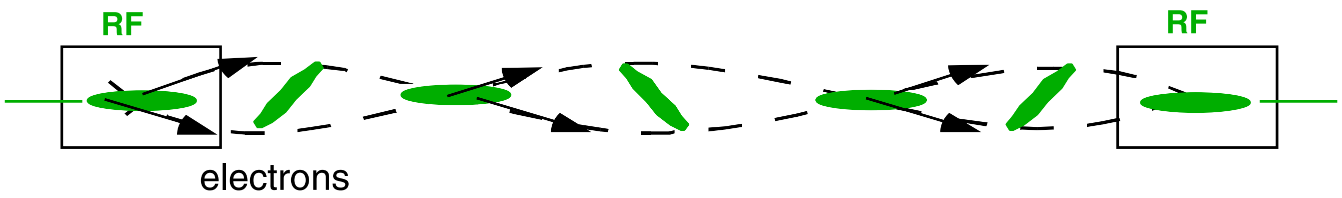
Generation of subpicosecond X-ray pulses using RF orbit deflection

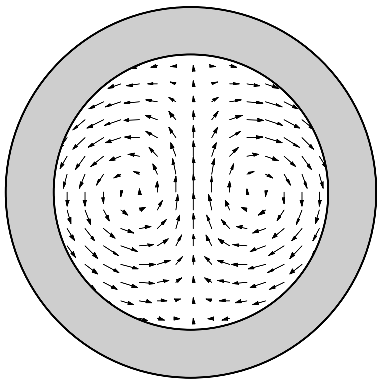

RF bunch deflection

KEK supeconducting deflecting cavity at 500 MHz reaching 1.5 MV/m

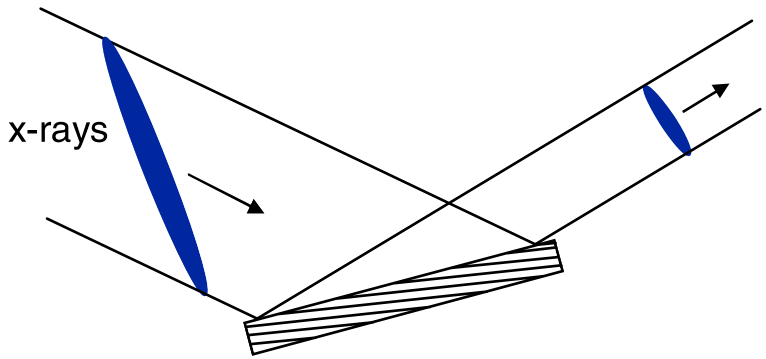


RF Electron Orbit Deflection and X-ray Pulse Compression

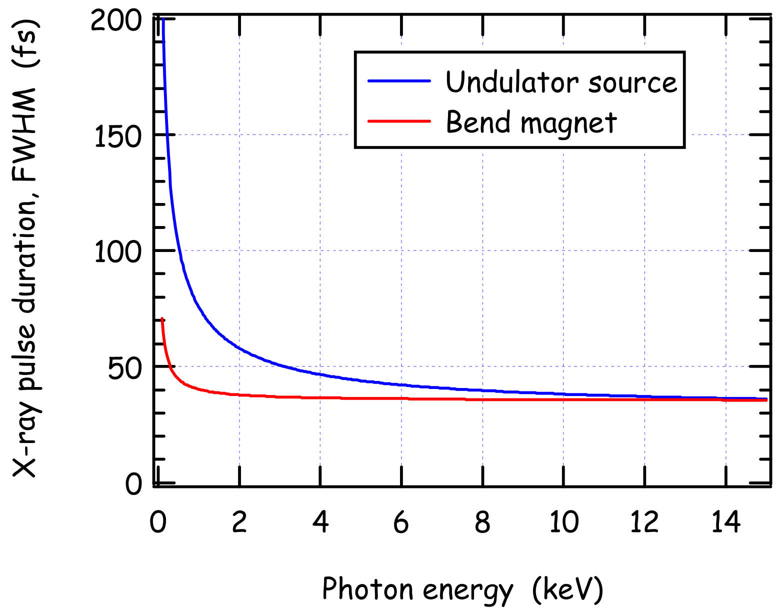




RF Magnetic field



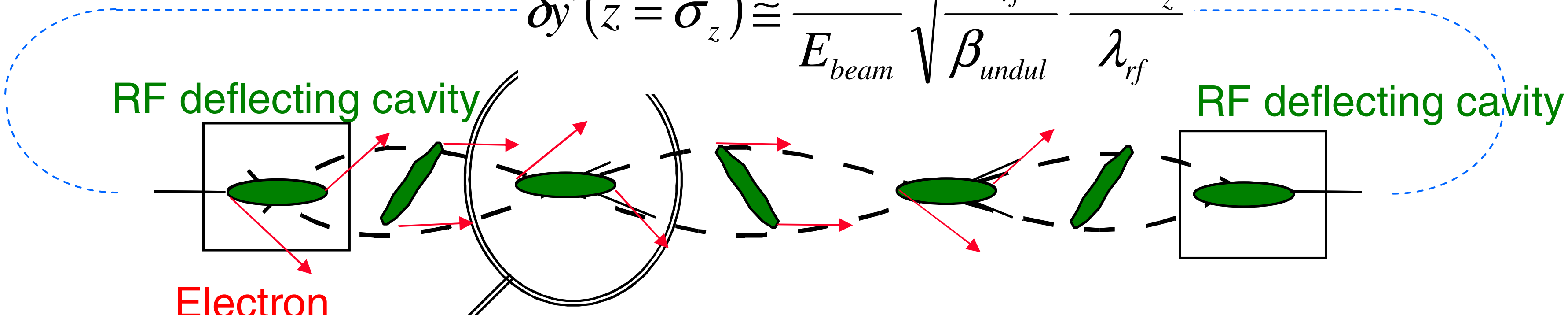
asymmetric-cut Bragg crystal

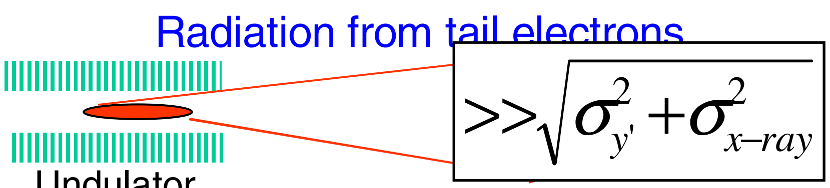


A. Zholents, P. Heimann, M. Zolotorev, and J. Byrd, *Nuc. Inst. Meth. A*, **425** (1999)

Obtaining short x-ray pulse from a “long” electron bunch

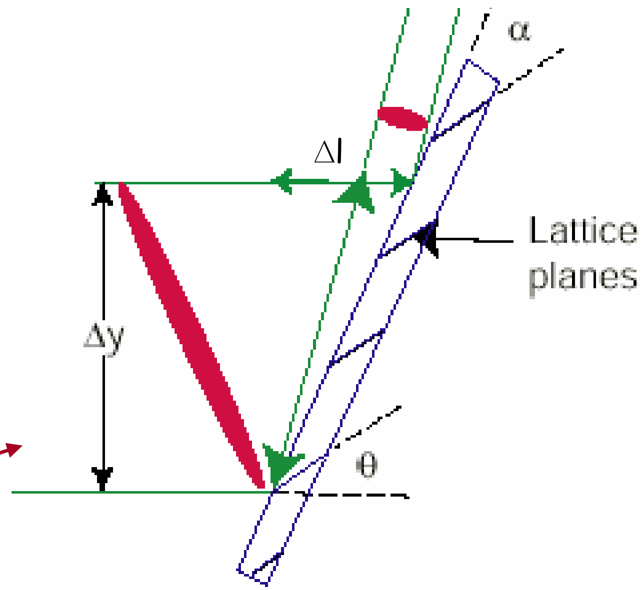
$$\delta y'(z=\sigma_z) \cong \frac{eU}{E_{beam}}\sqrt{\frac{\beta_{rf}}{\beta_{undul}}}\frac{2\pi\sigma_z}{\lambda_{rf}}$$





Collimating mirror

X-ray compression in asymmetric-cut crystals



Input x-ray pulse >> diffraction limited size and natural beamsize

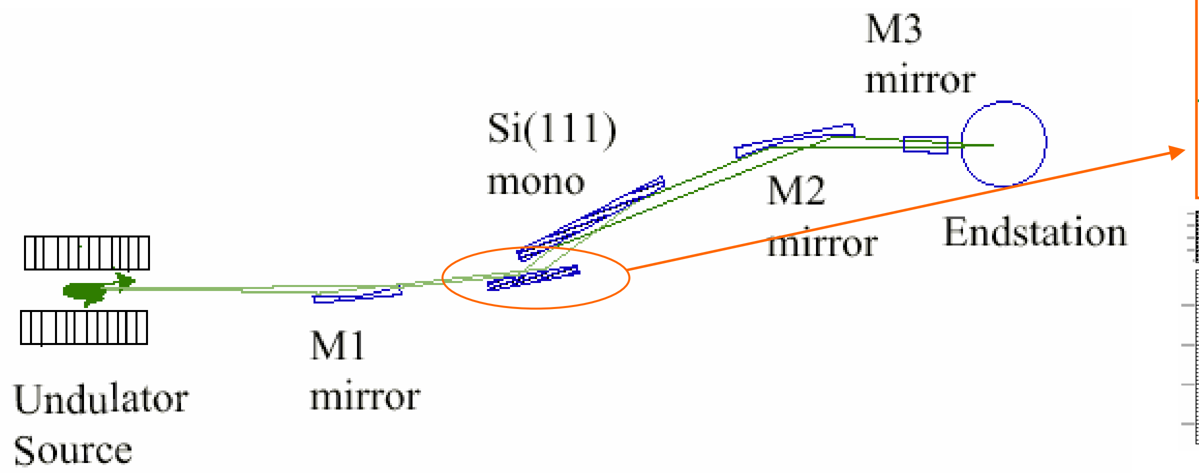
X-ray pulse compression (P. Heimann)

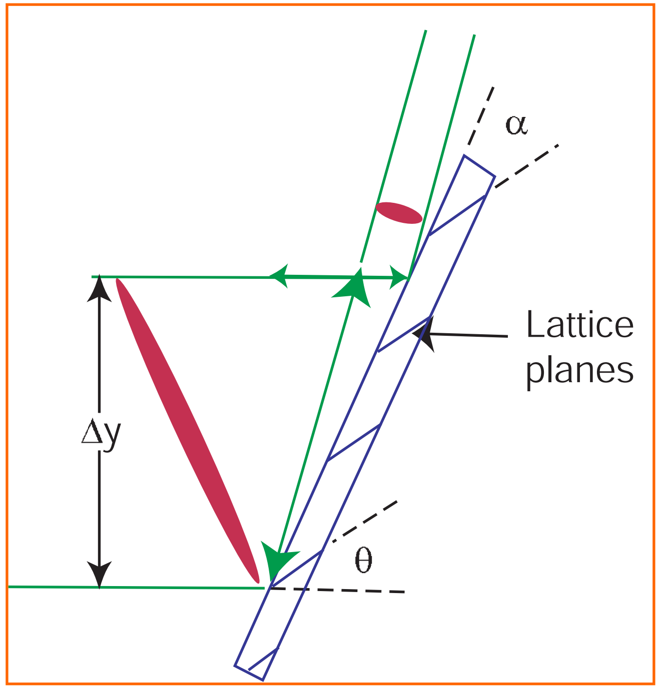
- **Optical path length Δl varies linearly with position Δy on crystal**

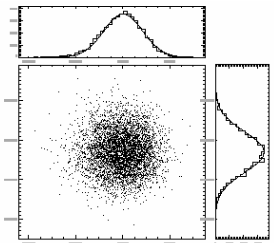
$$\Delta l = 2\,\Delta y \frac{\sin\theta\ \sin\alpha}{\sin(\theta+\alpha)}$$

Crystals	λ	Δy	θ	α	Δ*l*
Si(111)	1.5 A	3.8 mm	14.309°	-3.5°	0.6 mm (2 ps)

- **We propose to use a pair of asymmetrically cut silicon crystals following collection optics**

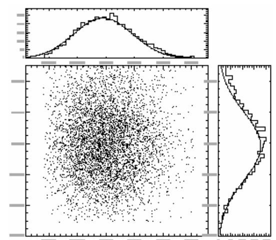






Focus dimensions

20μm (h) x 12 μm (v)

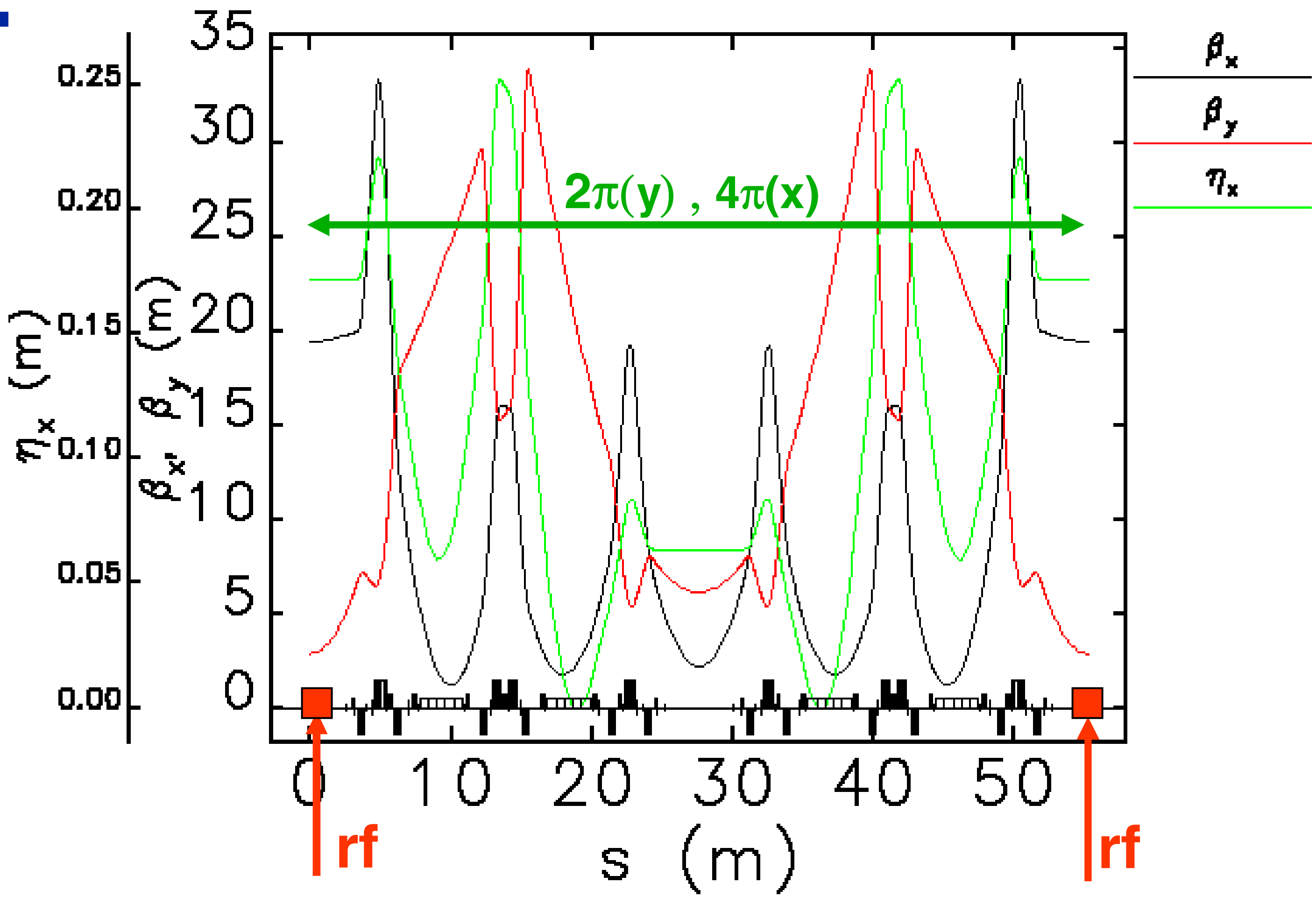


Focus divergence

1.2 mrad (h) x 500 μrad (v)

An example for the APS

V. Sajaev

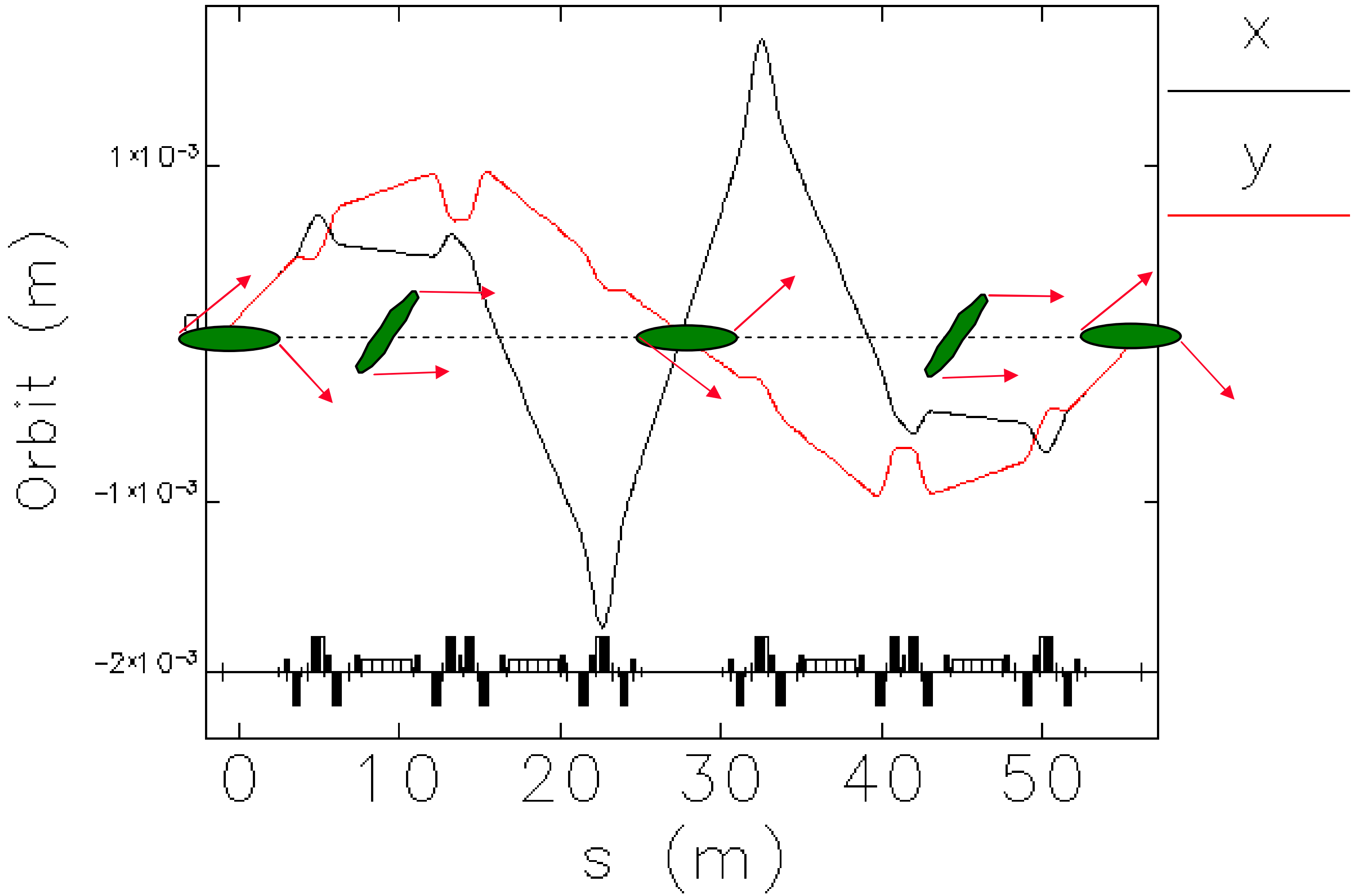


Twiss parameters for aps3

Two APS sectors are used.

Potentially: three undulator and four bend magnet beamlines.

Trajectory for an electron with $z=\sigma_z$ and 200 μrad kick



Parameters used:

Beam energy = 7 GeV

Vertical beam emittance = 2.5×10^{-11} m

Bunch length, σ = 40 ps

Undulator: 3.3 cm period, 73 periods

Bend magnet field = 0.6 T

Main RF frequency = 352 MHz

Deflection RF frequency= 4x352=1.408 GHz

Results obtained for undulator beamline:

Beam divergence, $\sigma_{y'}$= 2 μrad

X-ray divergence at 1Å, $\sigma_{r'}$= 3.7 μrad

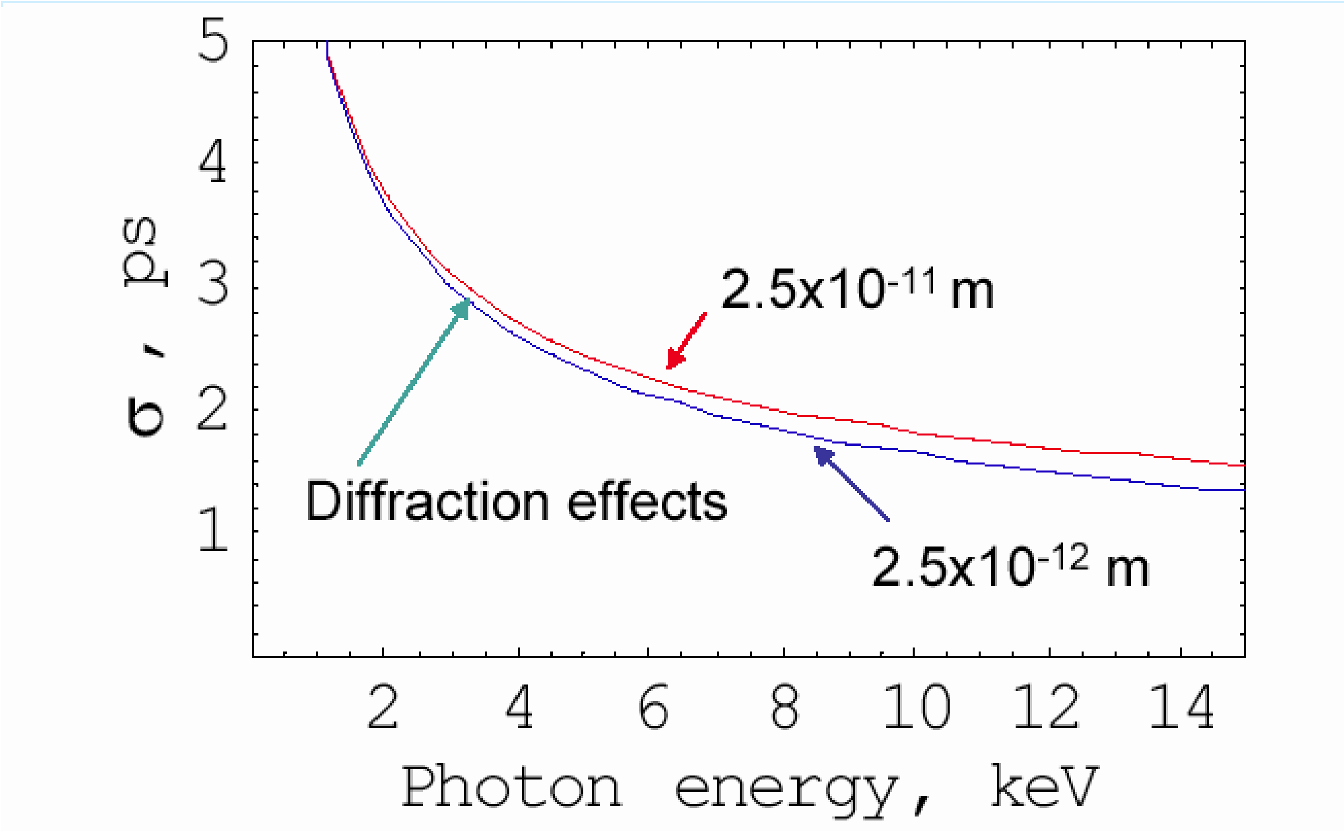
Total divergence = 4.2 μrad

Total transverse rf voltage = 2 MV

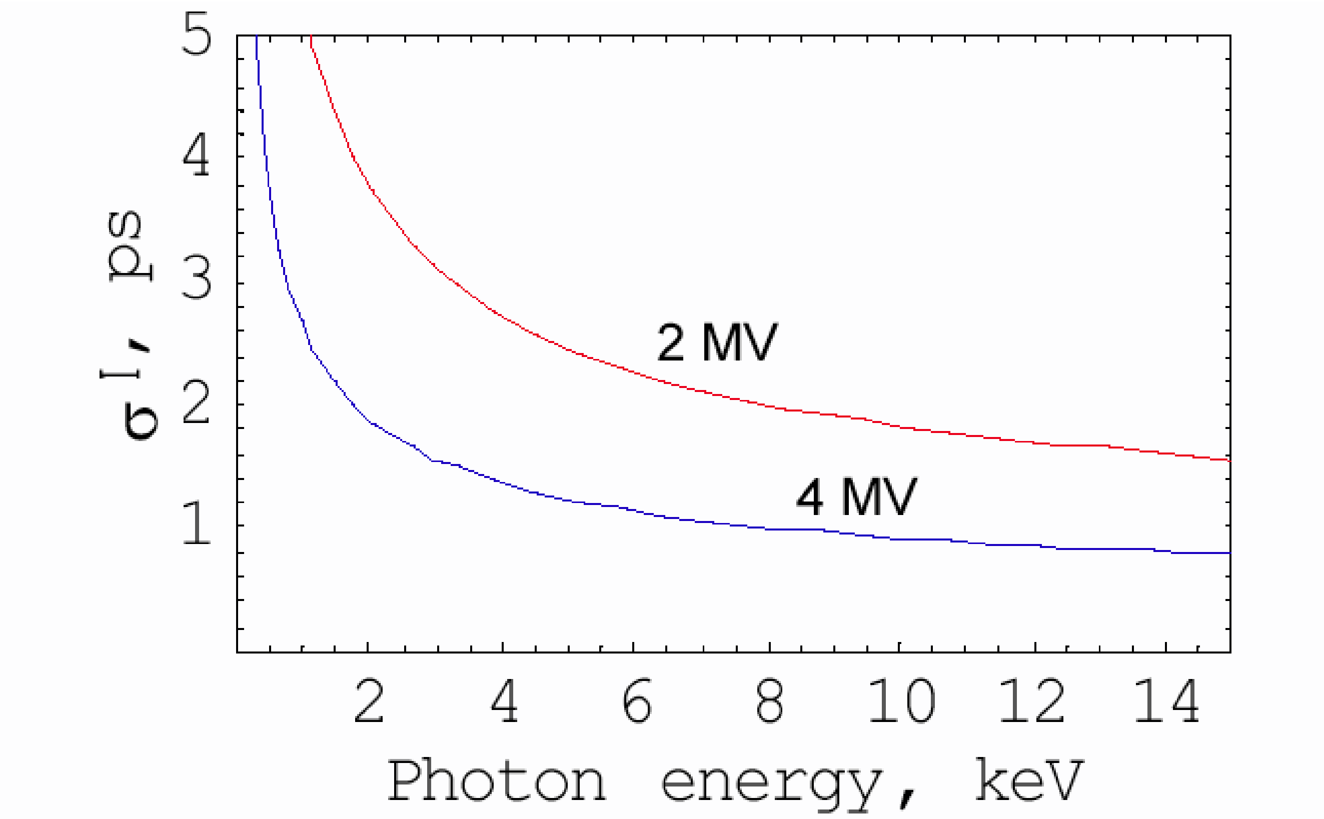
X-ray pulse duration (FWHM) = 2 ps

(compression factor ~ 50)

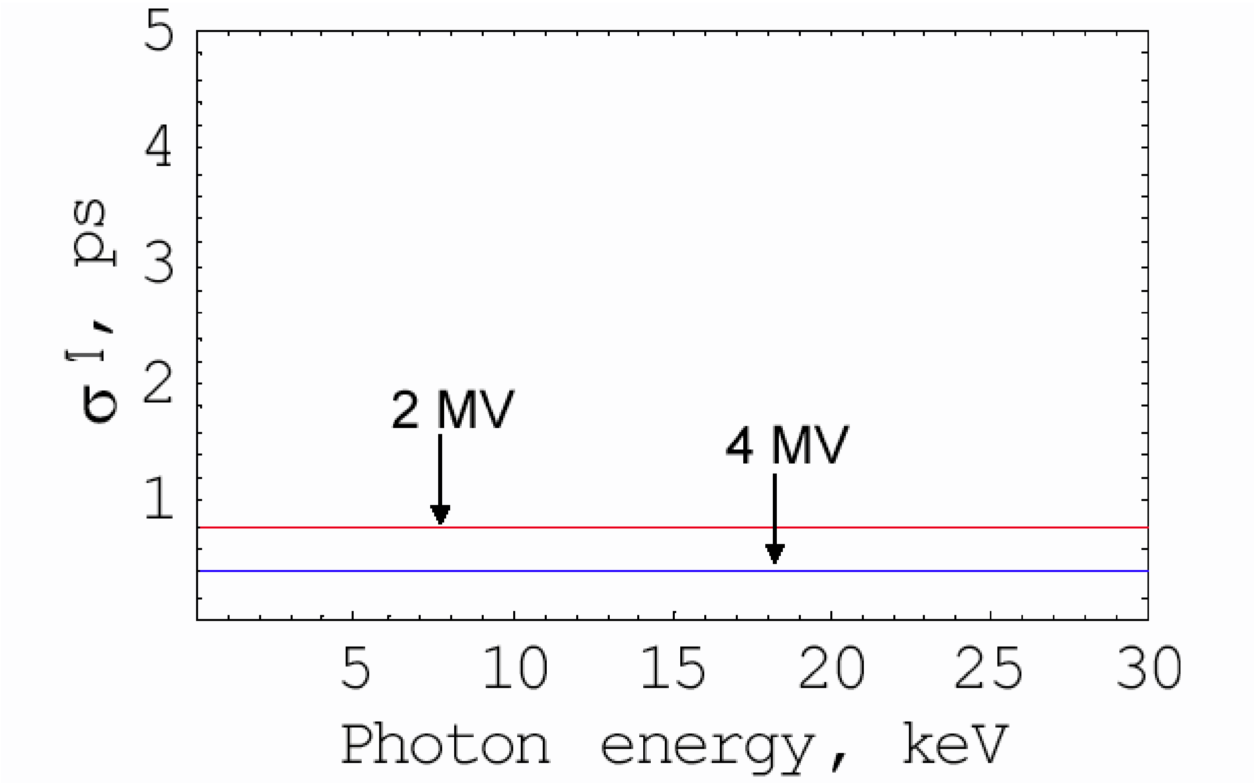
X-ray pulse duration at various photon energies and beam emittance for undulator beamline



Increasing rf deflection voltage



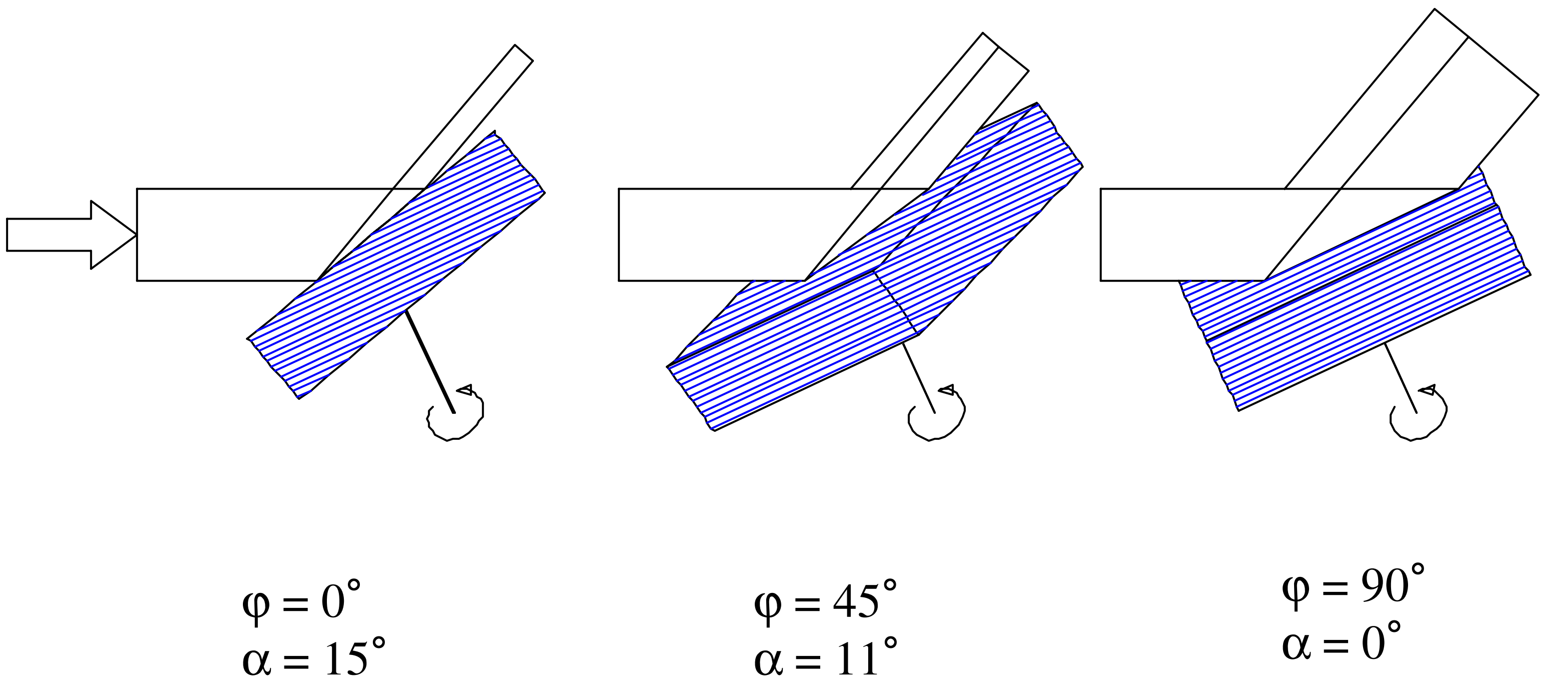
X-ray pulse duration at various photon energies and beam emittance for bend magnet beamline



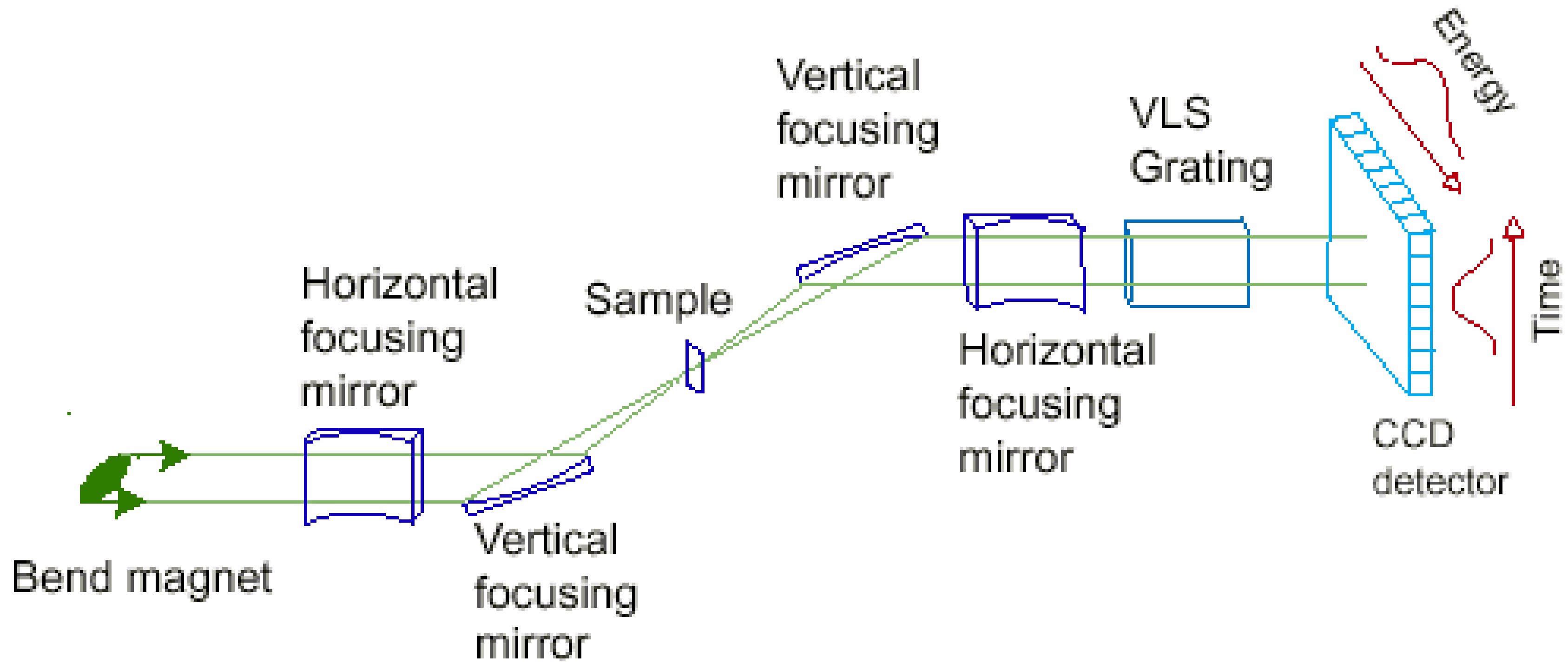
Tunability of x-ray pulse compression with photon energy (P. Heimann and H. Padmore)

- **Add rotation about axis normal to Bragg planes φ to rotation of Bragg angle θ**

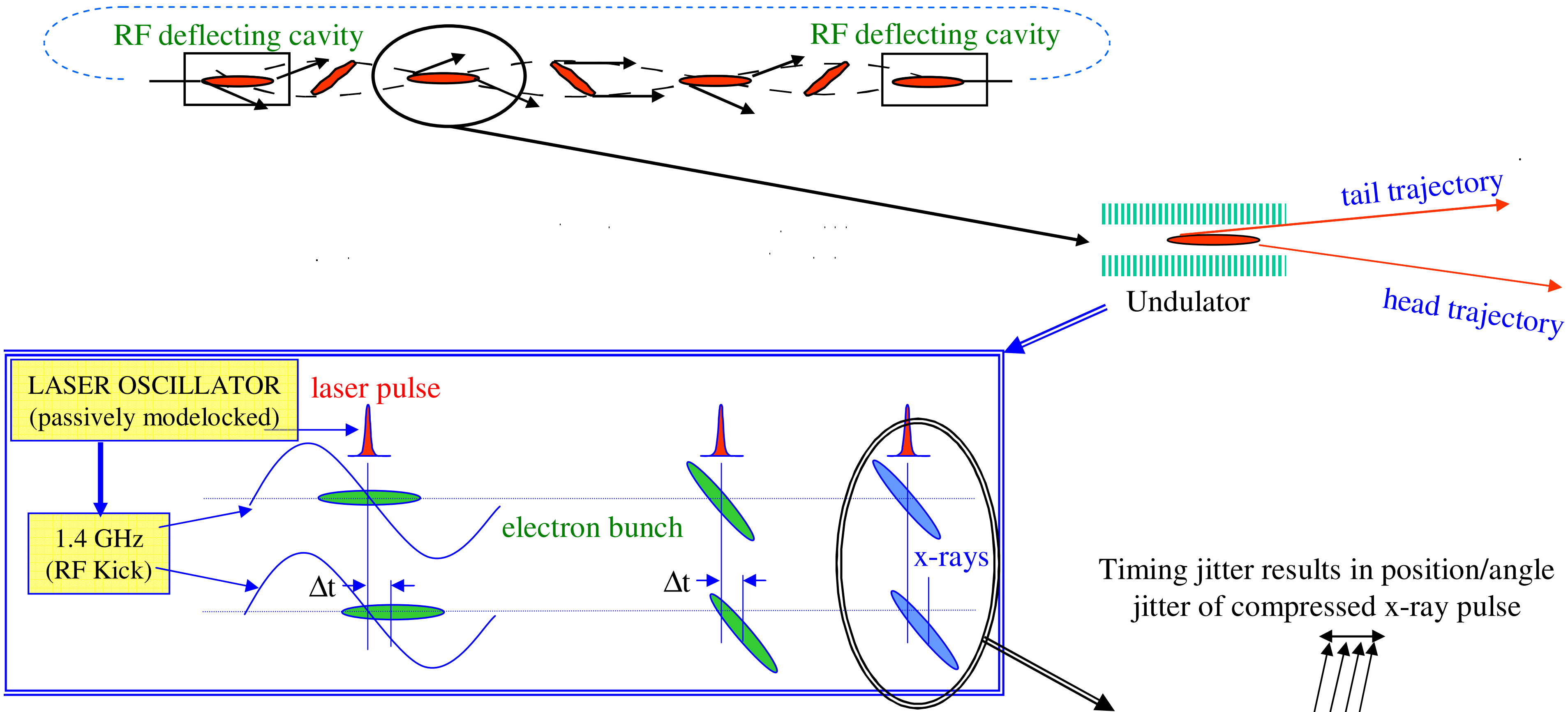
  ⇒ **Variation of crystal asymmetry α keeping pulse compression fixed**

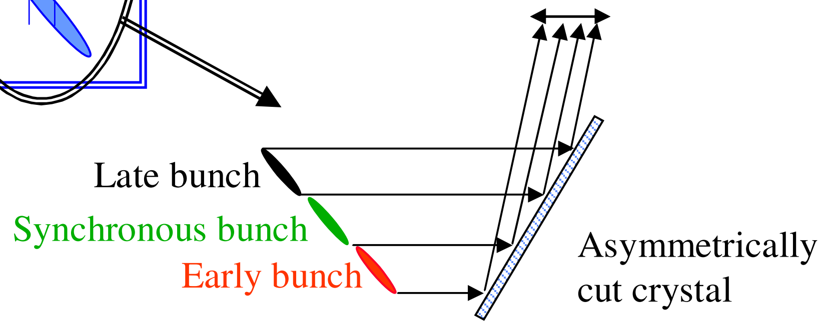


Simultaneous collection of different times and photon energies (appropriate for x-ray absorption, not photoemission)



Synchronization to the pump laser pulse

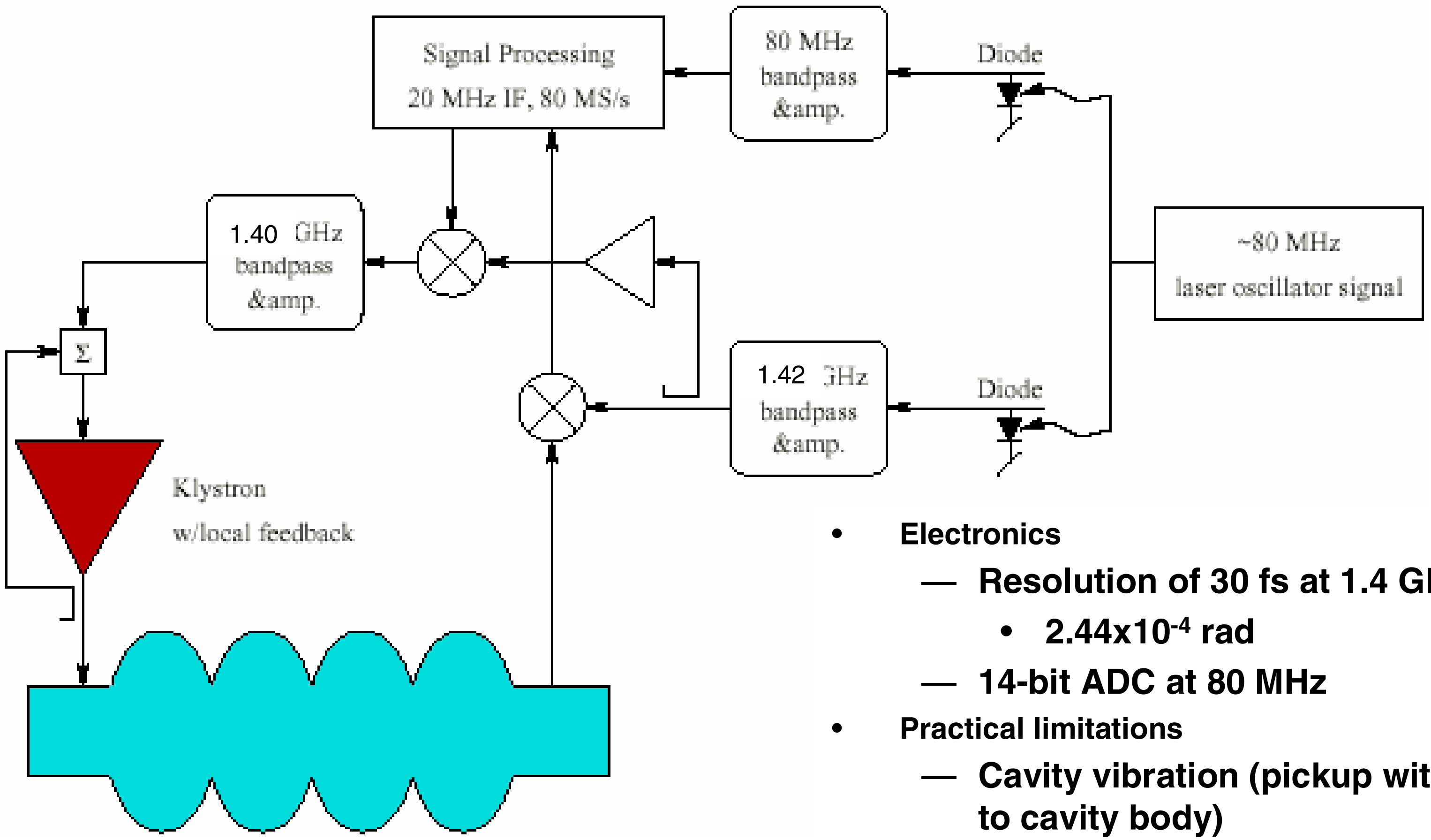




- **Electron bunch timing jitter ~ 500 fs**
- **Deflecting cavity phase stability ~ 0.01°**
  - **50 fs synchronization**

Deflecting cavity RF control (L. Doolittle)

- **Control cavity phase and amplitude to minimize timing jitter**
  - **— "Fast" feedback**



- **Electronics**
  - **— Resolution of 30 fs at 1.4 GHz**
    - **2.44×10^{-4} rad**
  - **— 14-bit ADC at 80 MHz**
- **Practical limitations**
  - **— Cavity vibration (pickup with respect to cavity body)**

- The rf orbit deflection does not affect the x-ray flux.
- Brightness is reduced by a compression factor.
- Laser repetition rate or sample “relaxation time” define the “useful” x-ray flux in the pump-probe experiments.

•Slicing technique is very challenging at the top energy.

•It might be possible at a lower energy (< 5 GeV)

•X-ray pulses less than 1 ps can be produce using RF orbit deflection technique

•Minimal modification to the APS lattice is required.

•Necessary to build a pair of SR RF deflection cavities.