Ultrafast X-Ray Experiments

New science applications at electron synchrotrons, linacs, and elsewhere

Philip H. Bucksbaum

FOCUS Center, University of Michigan

Ultrafast X-ray Science

- What's Ultrafast?
- X-ray Science Contributions to Ultrafast Science
- Challenges and Opportunities
- Ultrafast Science at XFEL's

Ultrafast Sources and Science:

X-ray sources:

Synchrotrons

Laser plasmas

XFEL's

Current lasers:

Ultrafast lasers

Science:

Acoustic phonons

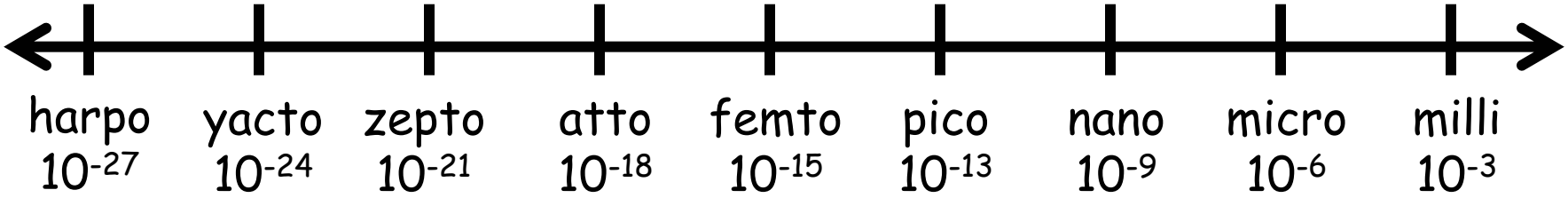
Vibrations (Optical phonons)

Chemistry and Biochem

Strings, Cosmology

Particle Collisions

Electron dynamics



Pulse duration (seconds)

Virtual Journal of

Ultrafast Science

a multijournal compilation of the latest research on ultrafast phenomena in physics, optics, chemistry, and biology

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News 15-Aug-03

- A Water Molecule's Chemical Formula is Really Not H2O
- Picosecond X-ray Crystallography of a Protein

Announcements 28-Jul-03

- Super Intense Laser Atom Physics (16-19 Nov 2003; Dallas, TX, USA)
- OSA Topical Meeting on High Fields and Short Wavelengths (12-15 Oct 2003; Biarritz, France)
- Intl. Conf. on Ultrashort High-Energy Radiation and Matter (7-10 Oct 2003; Varenna, Italy)
- Frontiers in Optics/Laser Science (5-9 Oct 2003; Tucson, AZ, USA)

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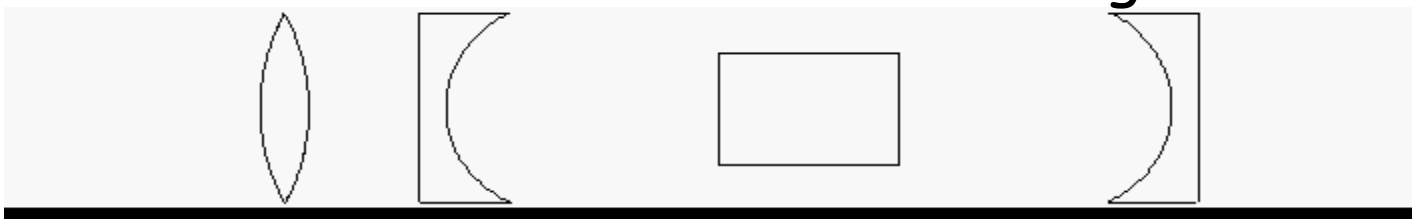
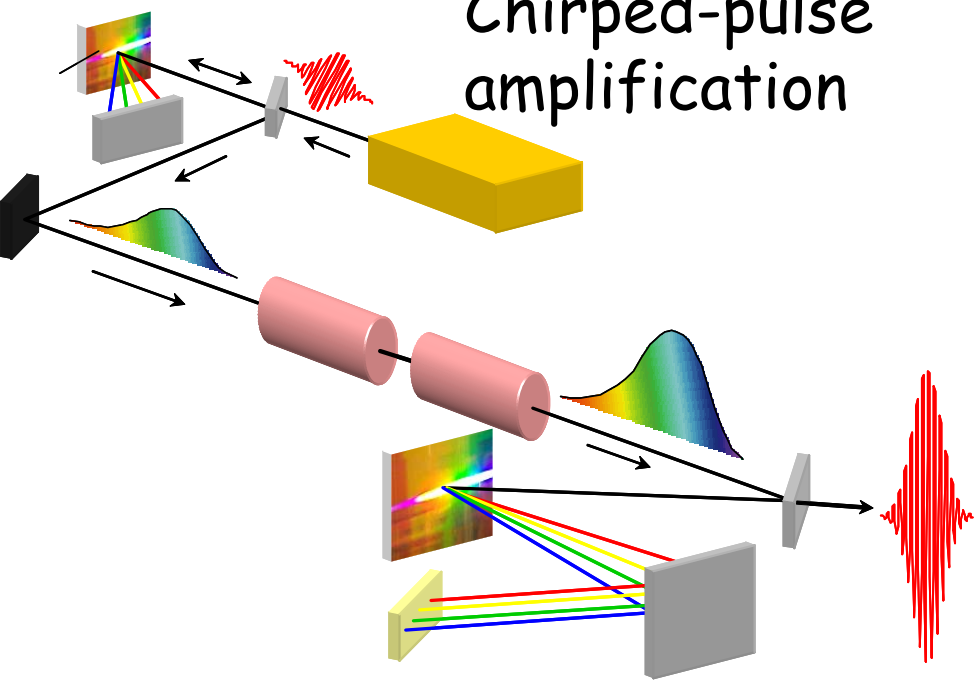
Tools:

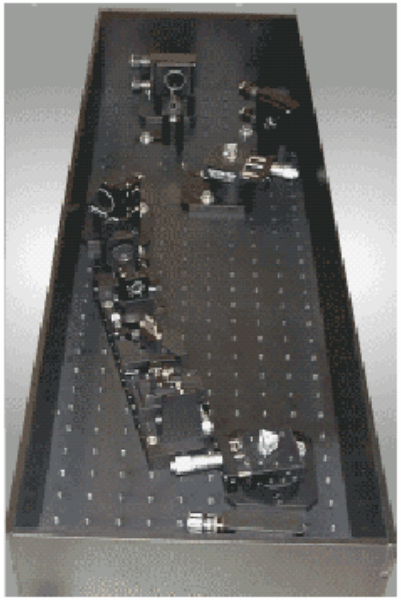
Kerr-lens modelocking



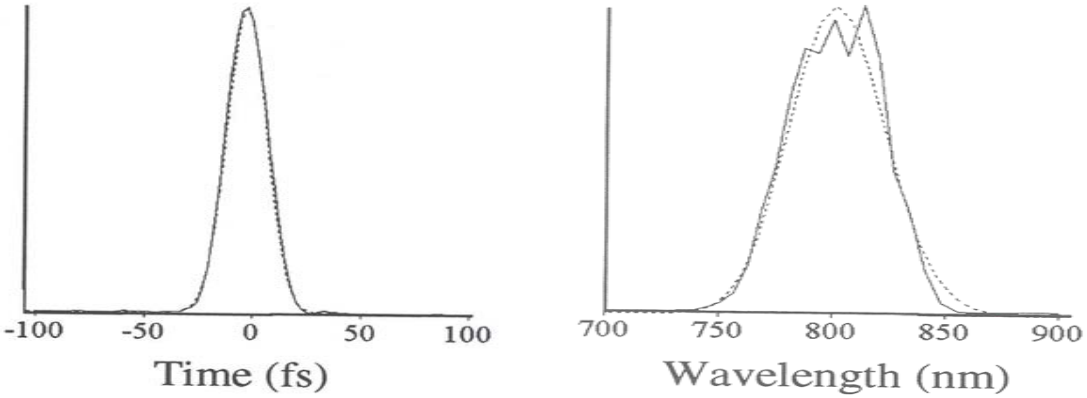
Chirped-pulse amplification



Ultrafast Ti:Saph Laser Pulses



Ti:Saph Oscillator
(KMLabs)



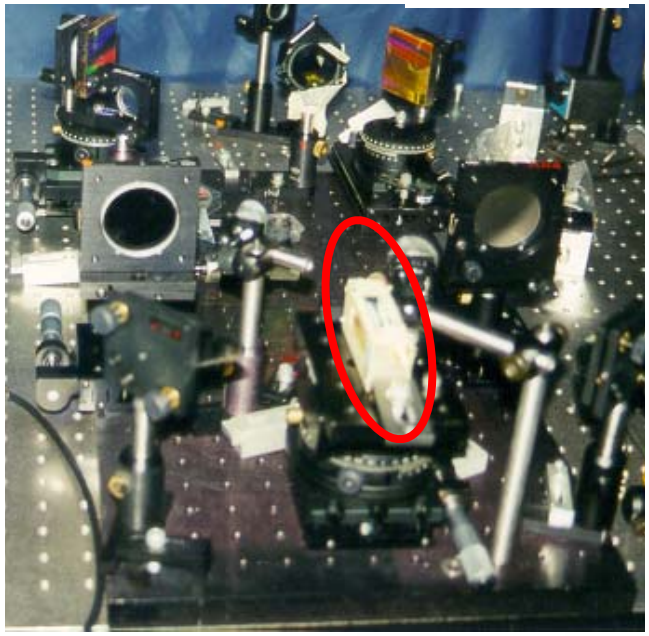
Backus, S., et.al., Review of Scientific Instruments, 69, 1207 (1998).

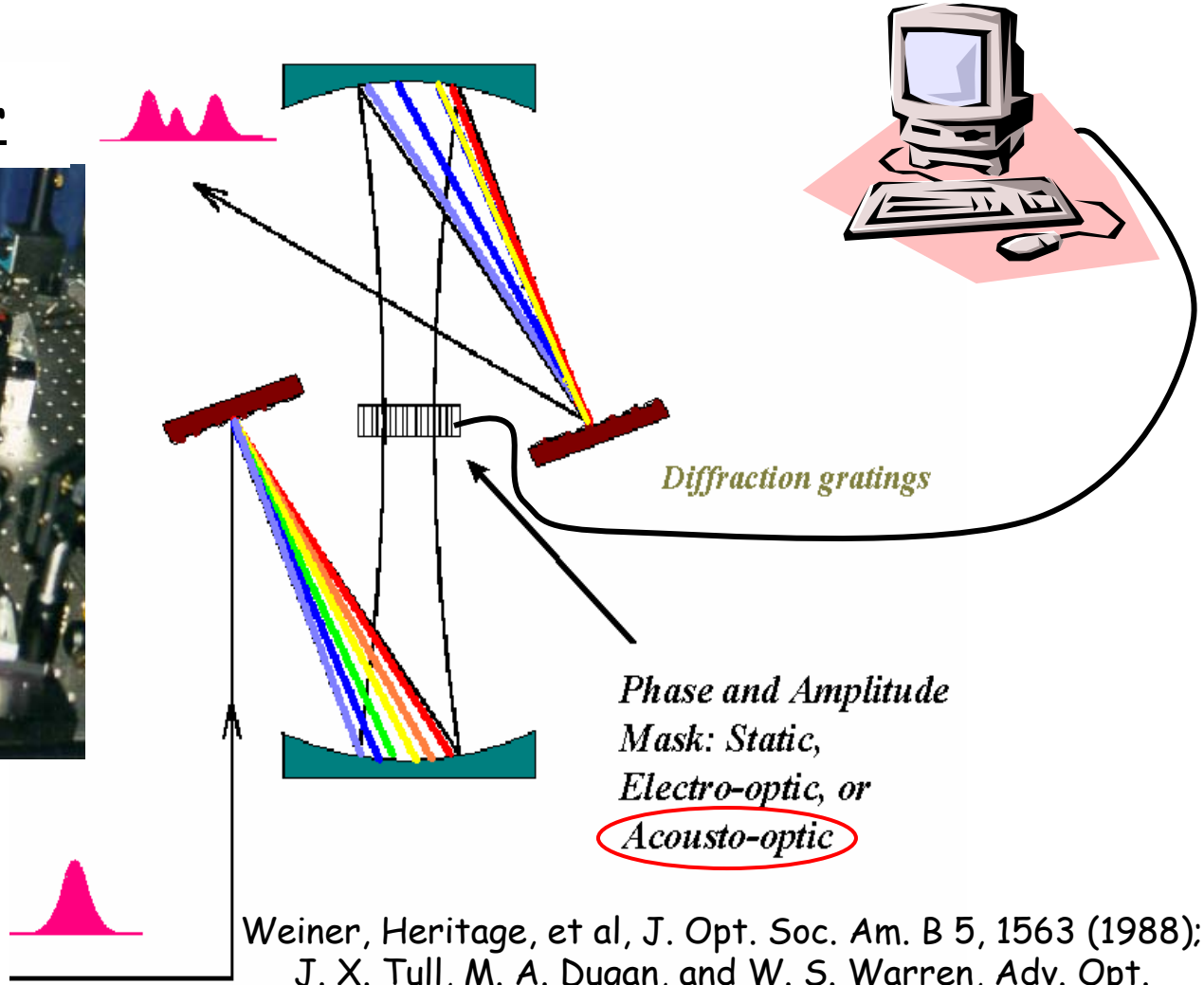
Advantages of Short Pulses:

1. fs time scale – observe/control fast dynamics
2. High intensity – strong field, multiphoton processes
3. Wide spectrum – ability to shape the pulse

2a. Optical Pulse Shaping

AOM Pulse Shaper

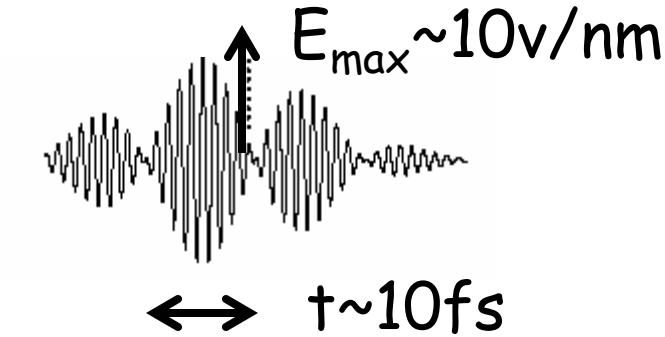




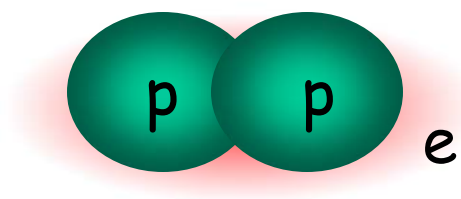
Weiner, Heritage, et al, J. Opt. Soc. Am. B 5, 1563 (1988); J. X. Tull, M. A. Dugan, and W. S. Warren, Adv. Opt. Mag. Resonance **20**, 1 (1997).

The Things We Can Do…

Total control of the laser field…



…can control quantum dynamics in molecules…



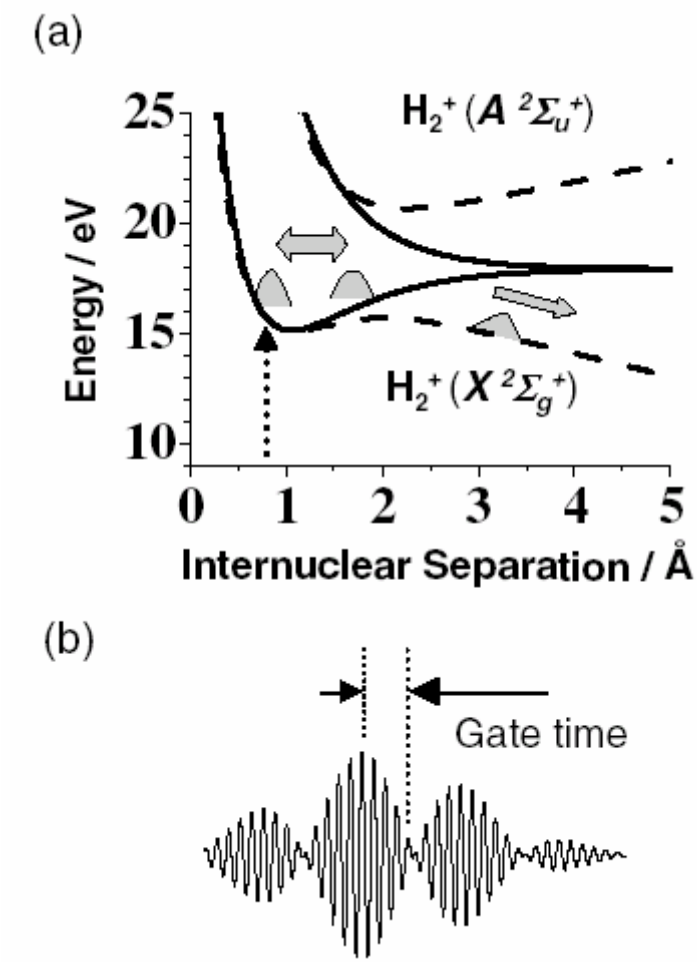
The Things You Can Do…

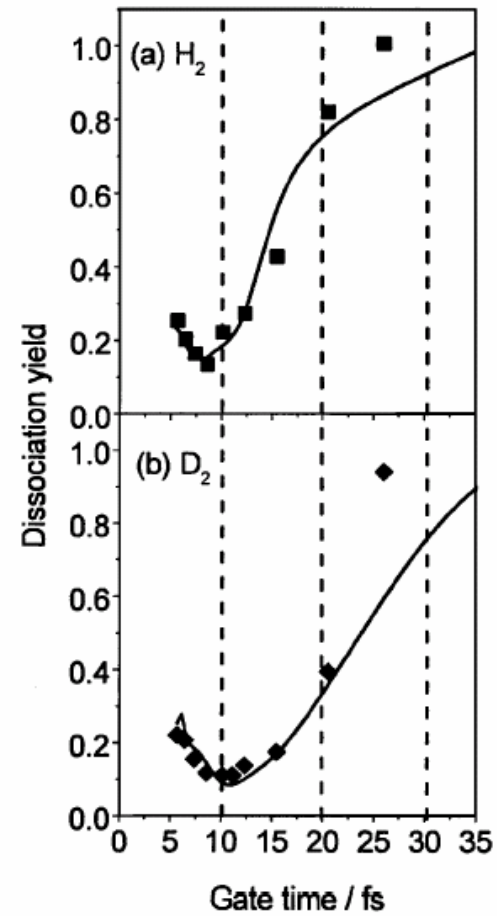
Controlling Vibrational Wave Packet Motion with Intense Modulated Laser Fields

Hiromichi Niikura, P. B. Corkum, and D. M. Villeneuve

National Research Council of Canada, 100 Sussex Drive, Ottawa, Ontario, Canada K1A 0R6

(Received 23 January 2003; published 21 May 2003)

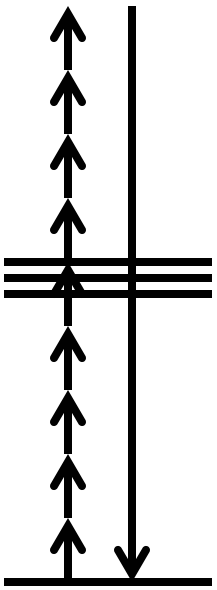


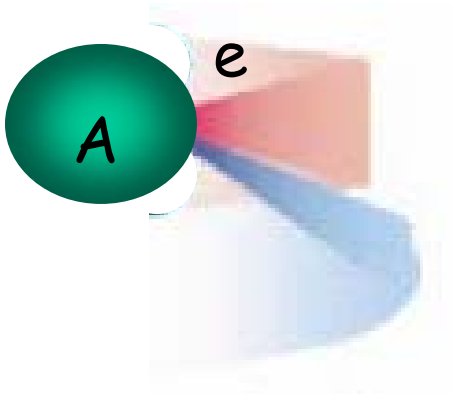


Niikura et al, PRL 90, 203601 (03)

High Harmonics: Bremmstrahlung from atomic electrons driven coherently by the oscillating laser field

$E_{max}=3.17U_p$





U_p=wiggle energy

Ultrafast soft x-rays from high harmonics: The ultrafastest light pulses

VOLUME 79, NUMBER 16 PHYSICAL REVIEW LETTERS 20 OCTOBER 1997

Generation of Coherent Soft X Rays at 2.7 nm Using High Harmonics

Zenghu Chang, Andy Rundquist, Haiwen Wang, Margaret M. Murnane, and Henry C. Kapteyn
Center for Ultrafast Optical Science, University of Michigan, Ann Arbor, Michigan 48109-2099
(Received 3 March 1997)

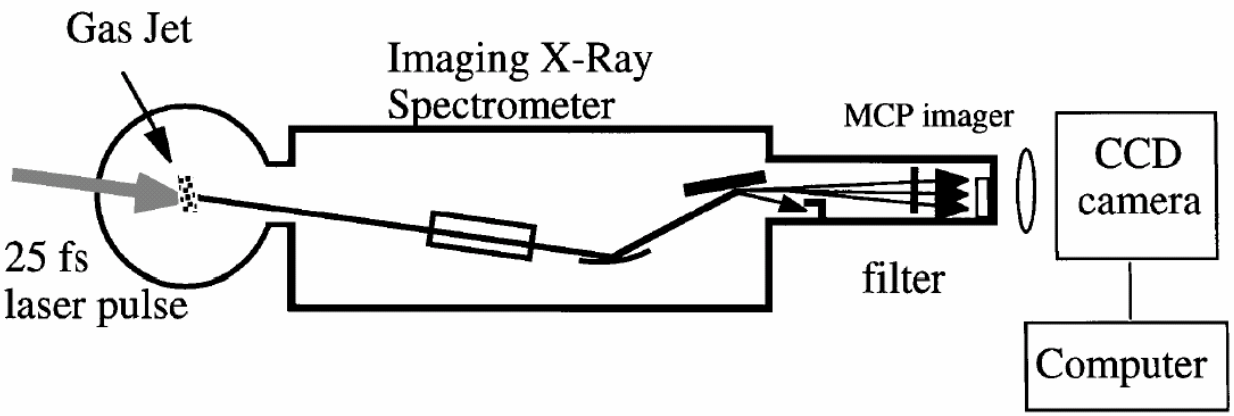


FIG. 1. Experimental setup for high-harmonic generation.

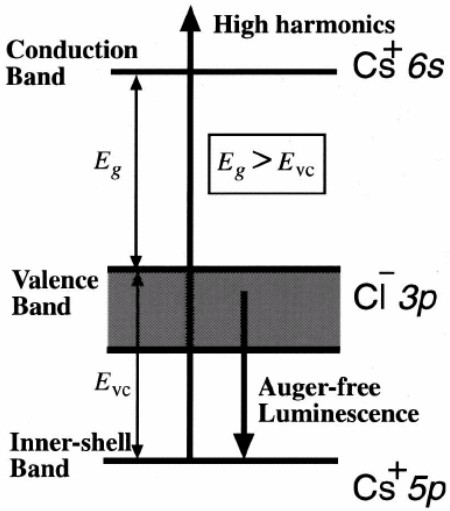
High harmonics are fast (<1fs!) but soft (<0.5keV)

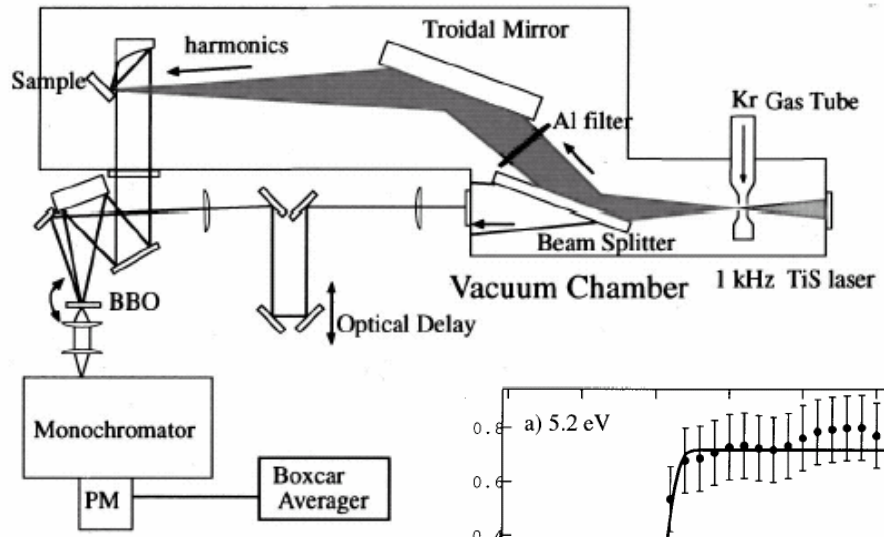
Femtosecond lattice relaxation induced by inner-shell excitation

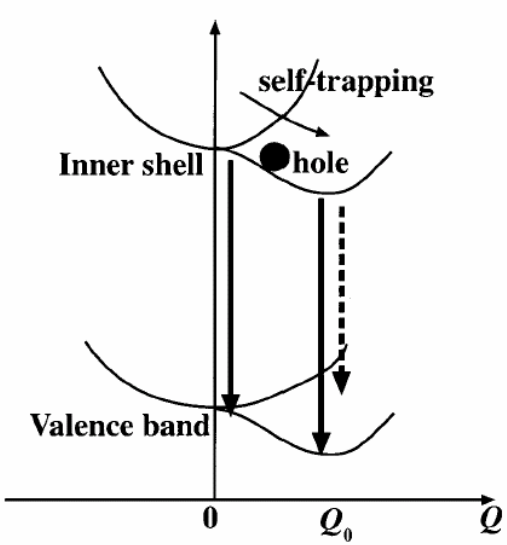
Ultrafast crystal dynamics viewed with High Harmonics

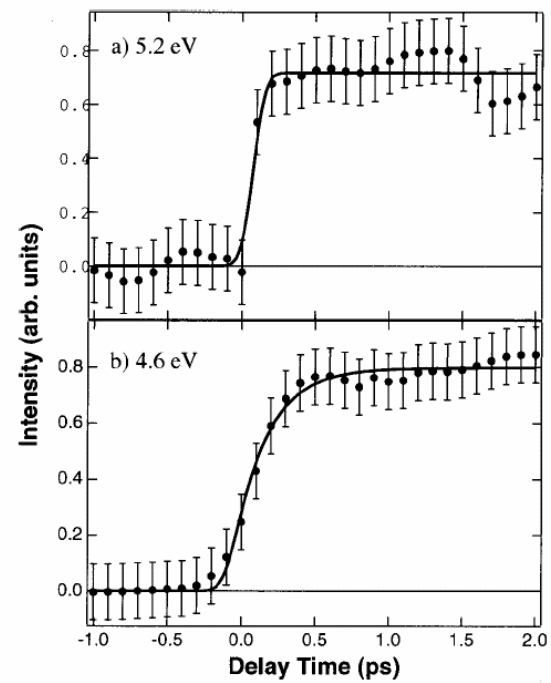
Taro Sekikawa, Tomohiro Yamazaki, Yasuo Nabekawa, and Shuntaro Watanabe

Institute for Solid State Physics, University of Tokyo, 5-1-5 Kashiwanoha, Kashiwa 277-8581, Japan

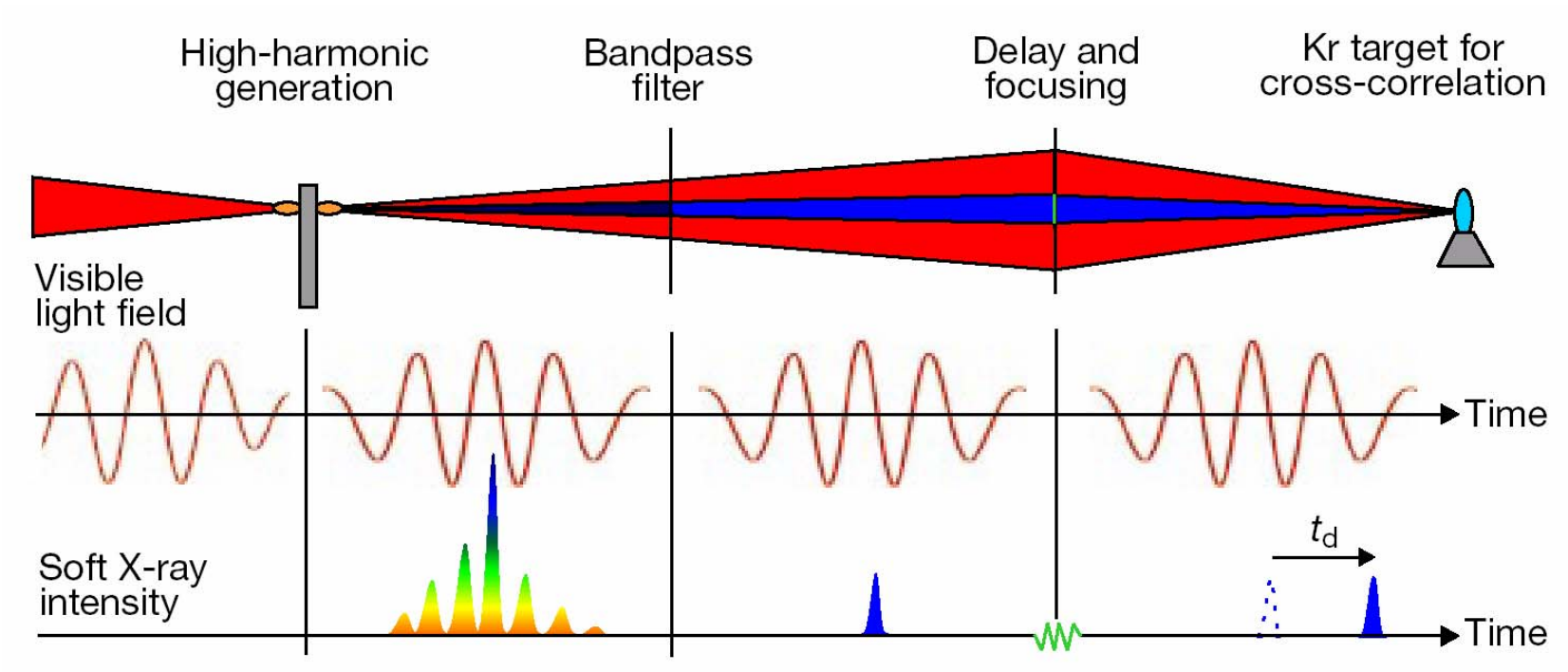








Sub-femtosecond pulses



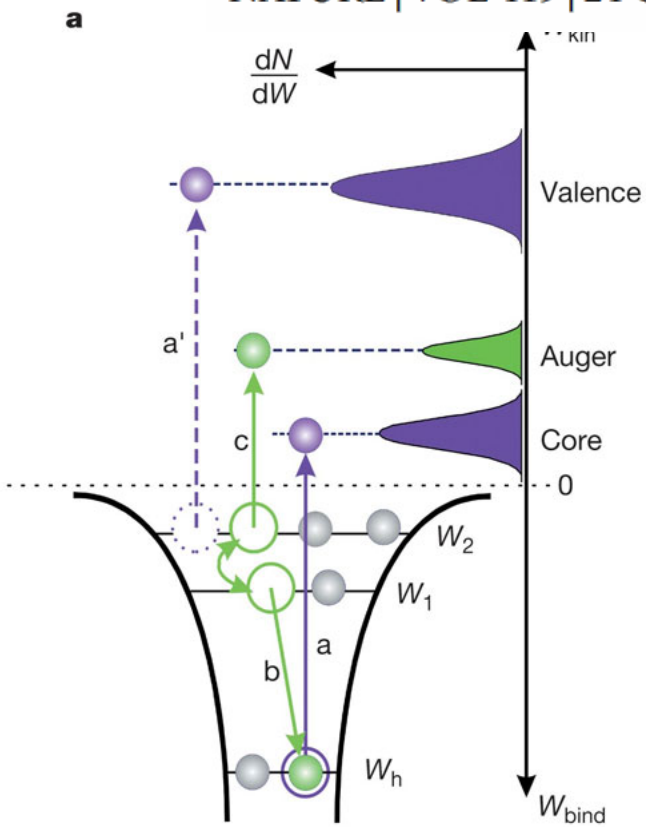
M. Hentschel, Nature **414**, 509 (01)

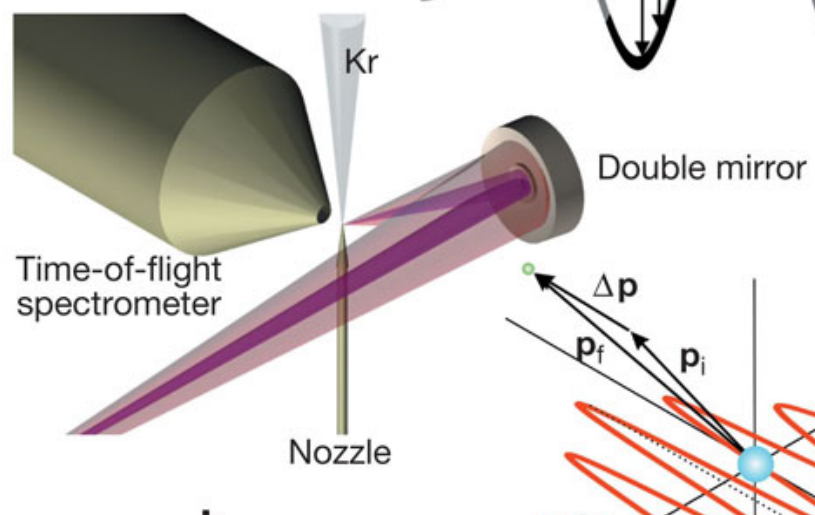
Attosecond high harmonics:

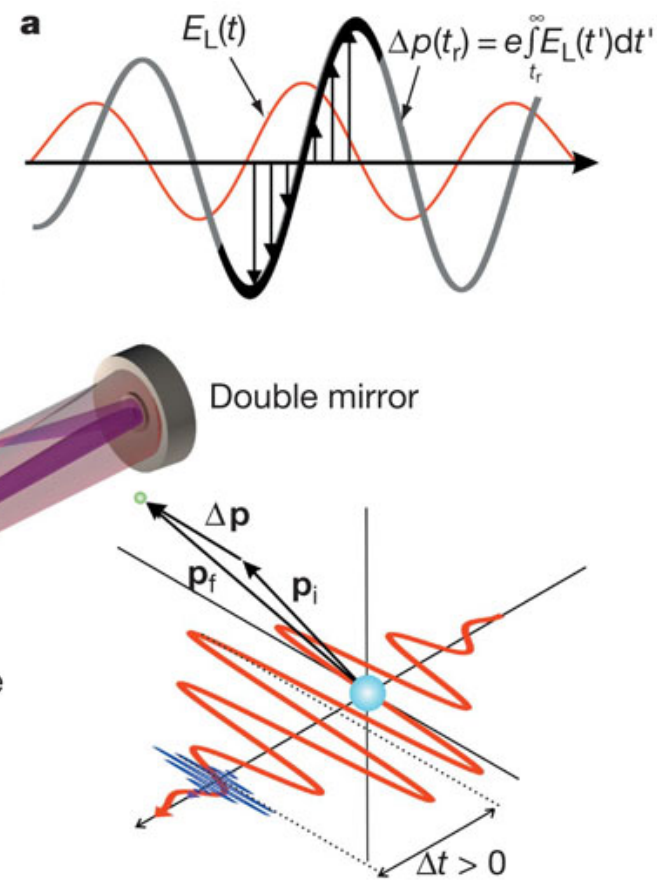
Time-resolved atomic inner-shell spectroscopy

M. Drescher*†, M. Hentschel*, R. Kienberger*, M. Uiberacker*, V. Yakoviev*, A. Scrinzi*, Th. Westerwalbesloh†, U. Kleineberg†, U. Heinzmann† & F. Krausz*

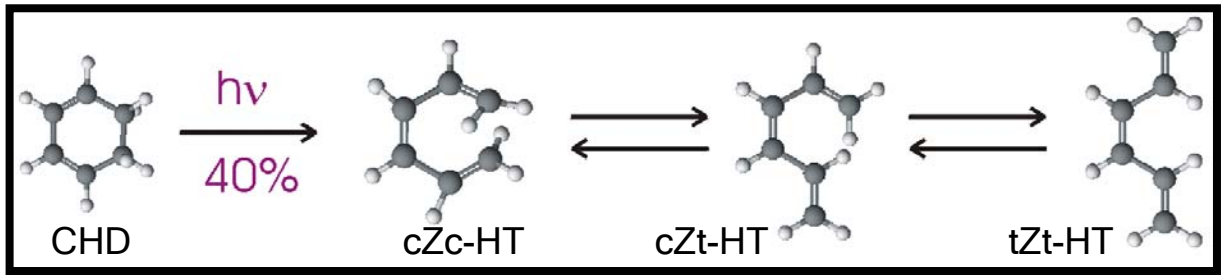
NATURE | VOL 419 | 24 OCTOBER 2002 |

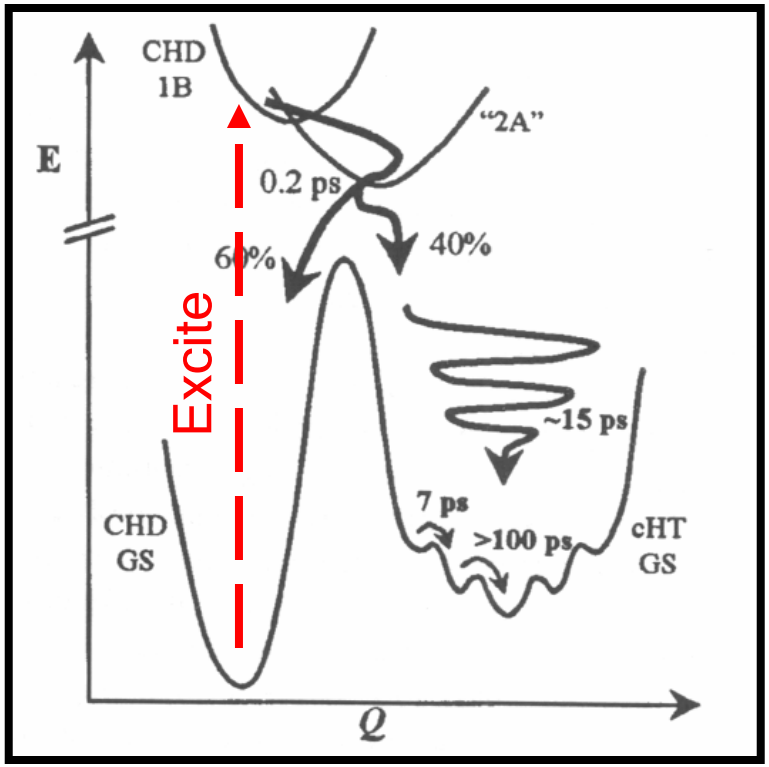


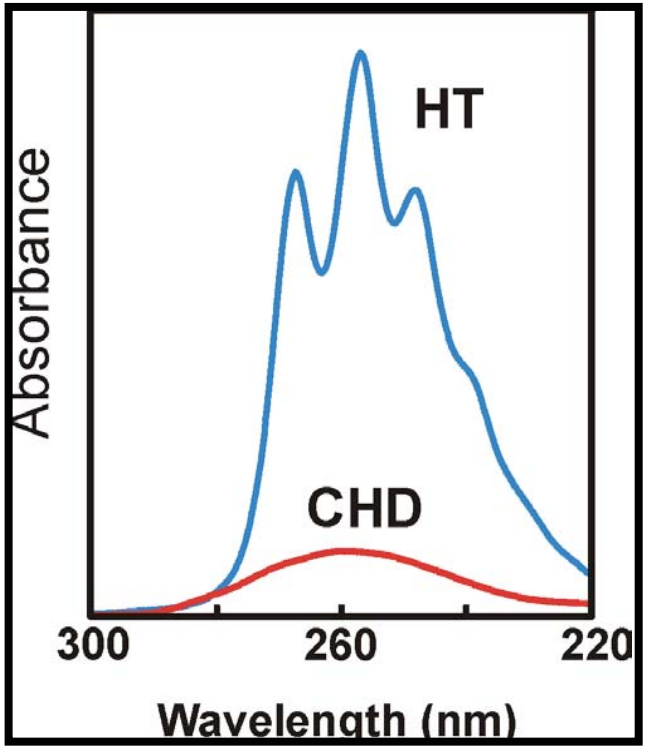




Optical measurements don't resolve molecular structure

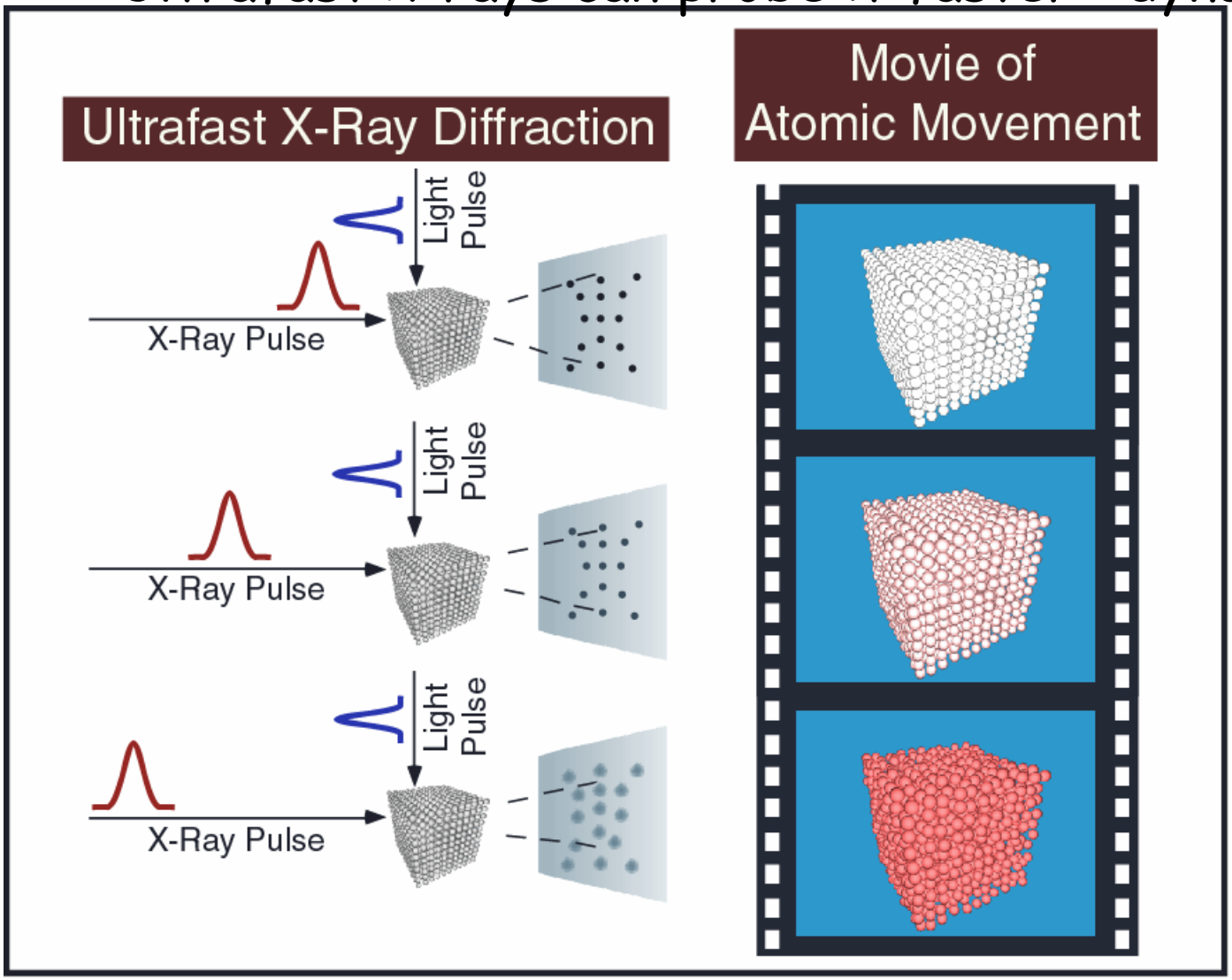






From *J. Chem. Phys.* **108**, 556 (1998)

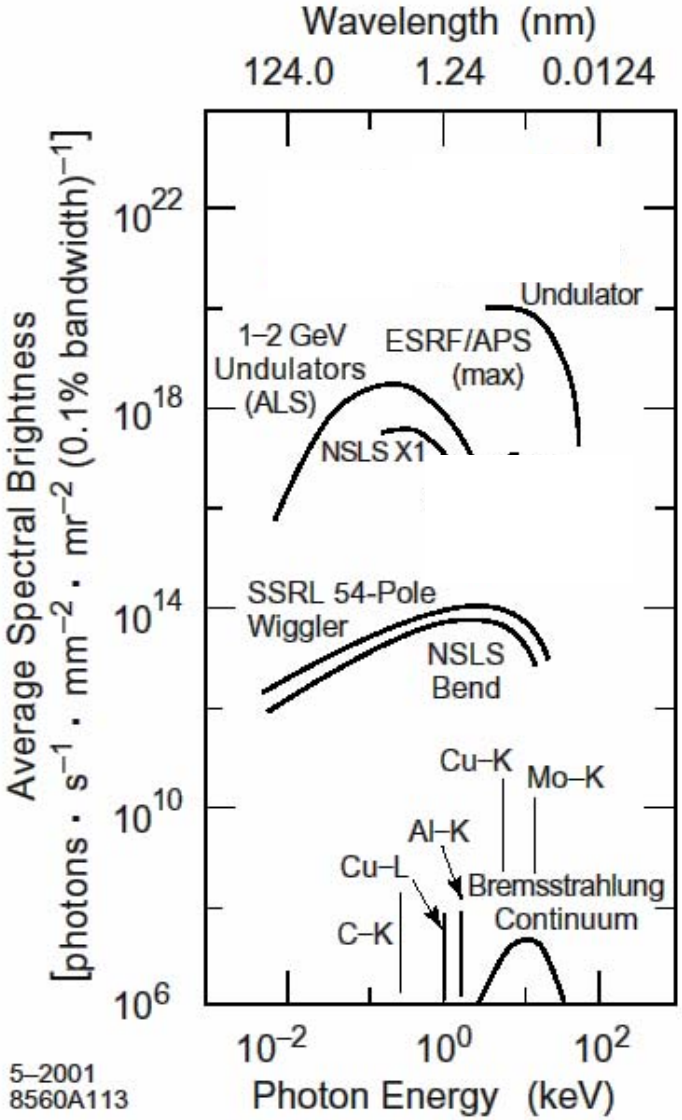
Hard X-rays can probe structure
Ultrafast x-rays can probe it faster: dynamics

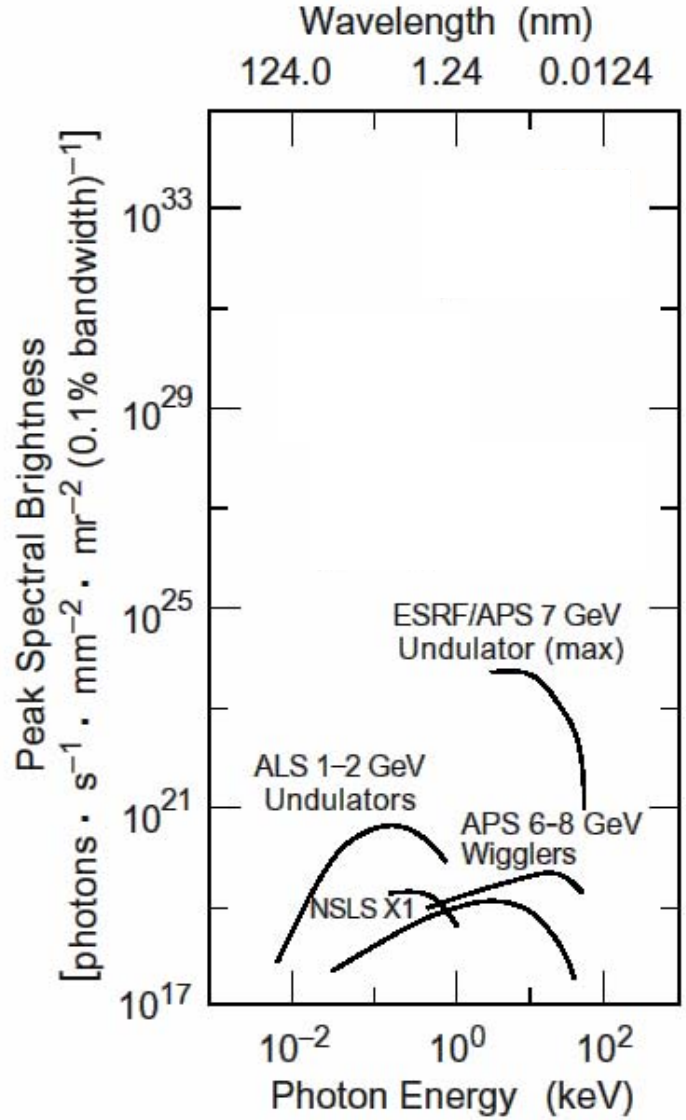


Stuff moves in 0.01-10 ps

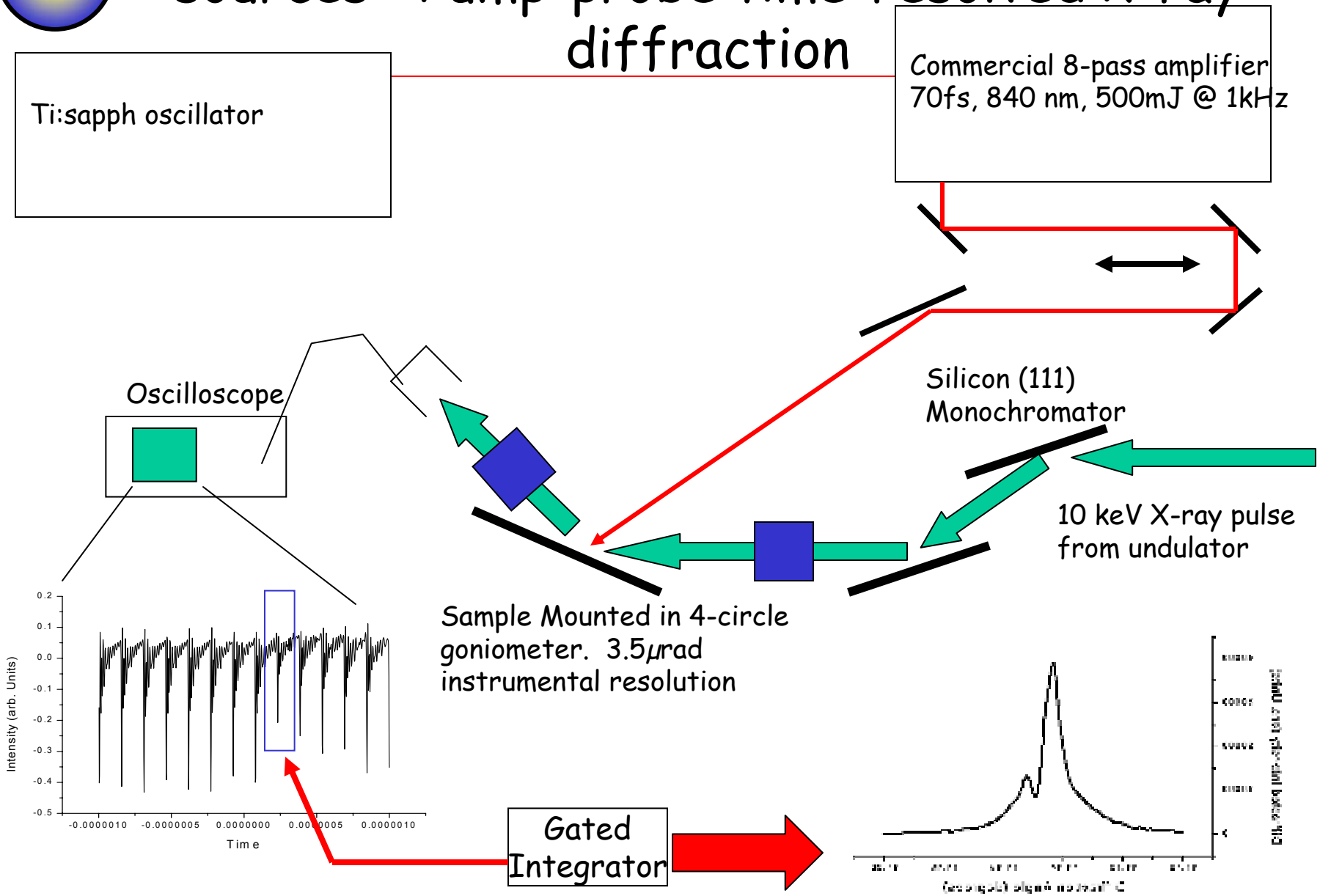
Source: LCLS – The First Experiments

3d generation synchrotrons have unprecedented brightness

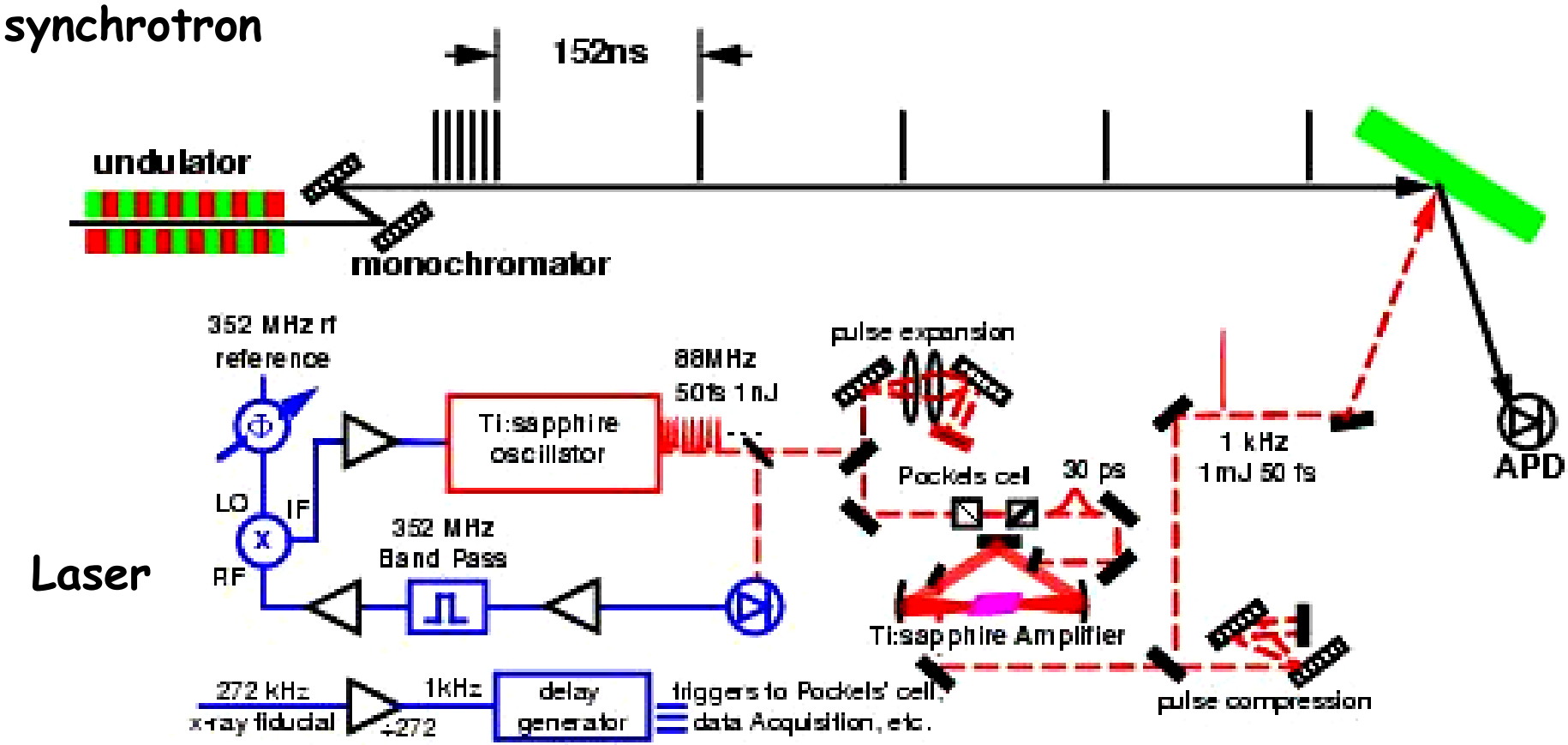




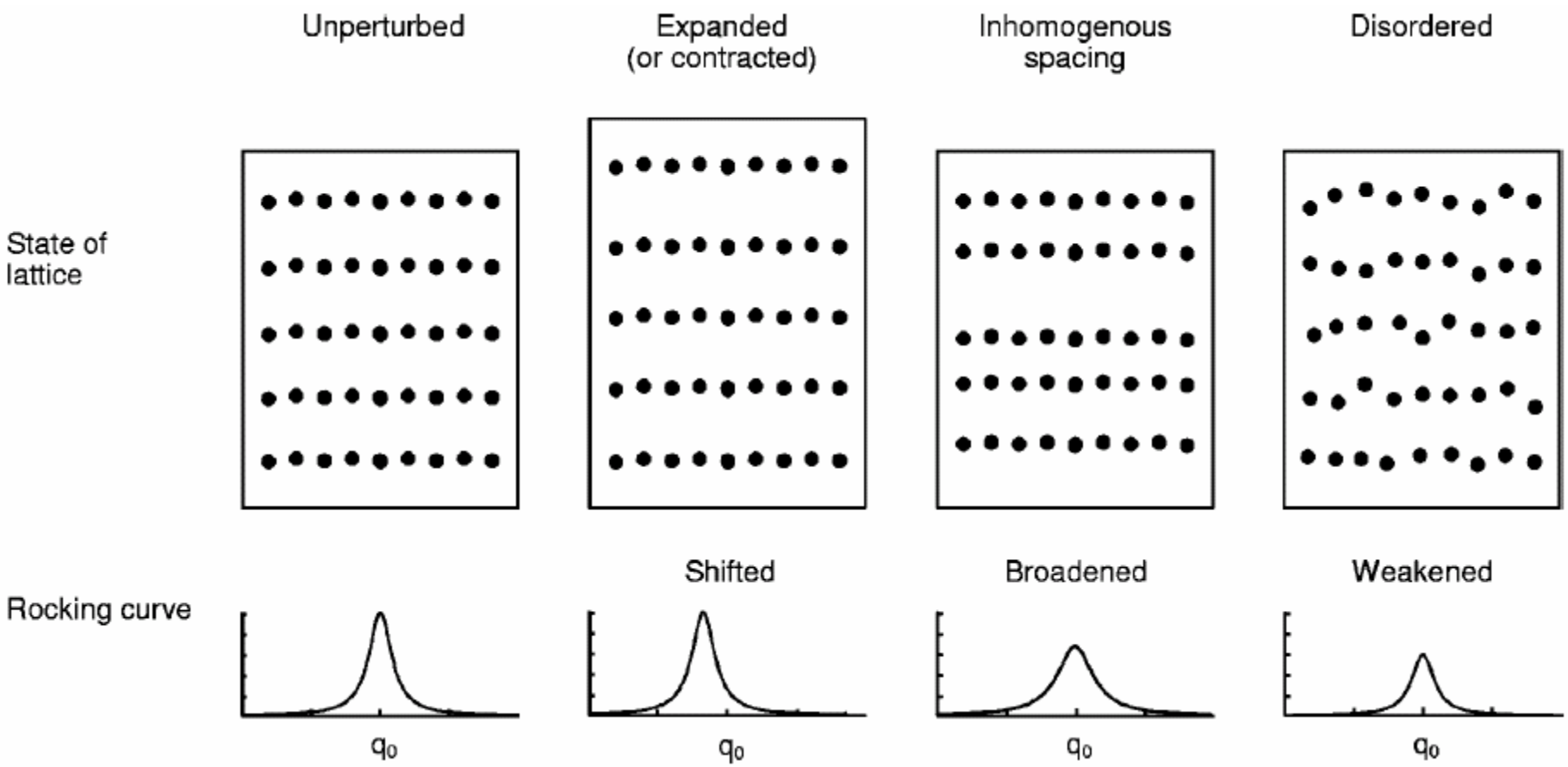
Ultrafast x-ray physics with synchrotron sources: Pump-probe time-resolved x-ray diffraction



Synchronizing the x-rays and the laser:



Probing dynamics with Bragg diffraction



Rousse et al., RMP **73**, 17 (2001)

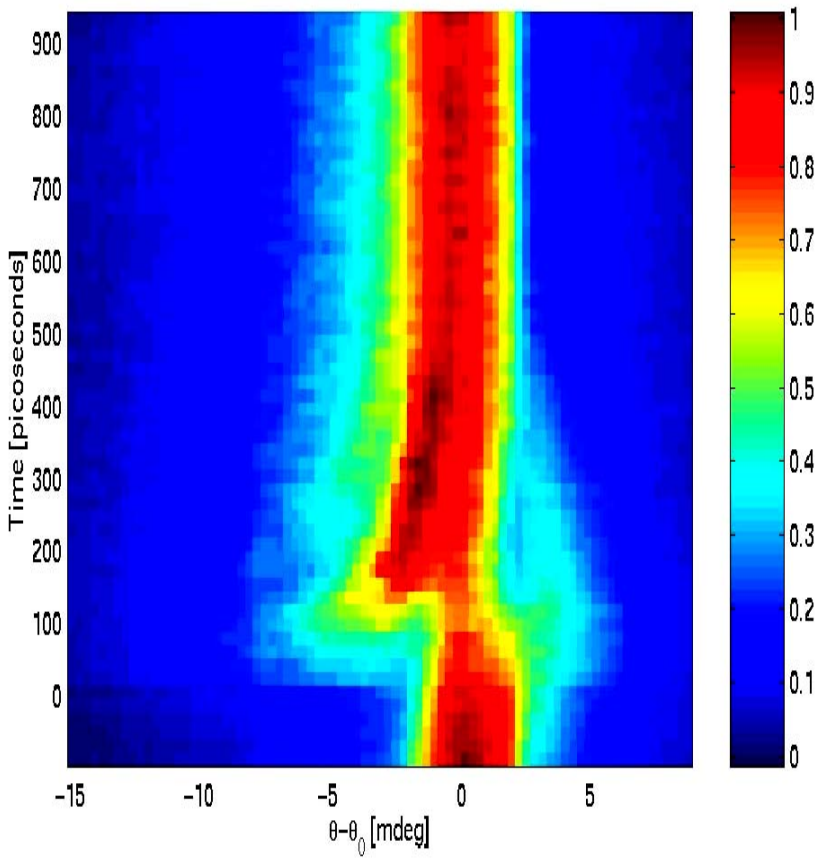
Time-resolved Bragg Diffraction: Coherent Acoustic Phonons

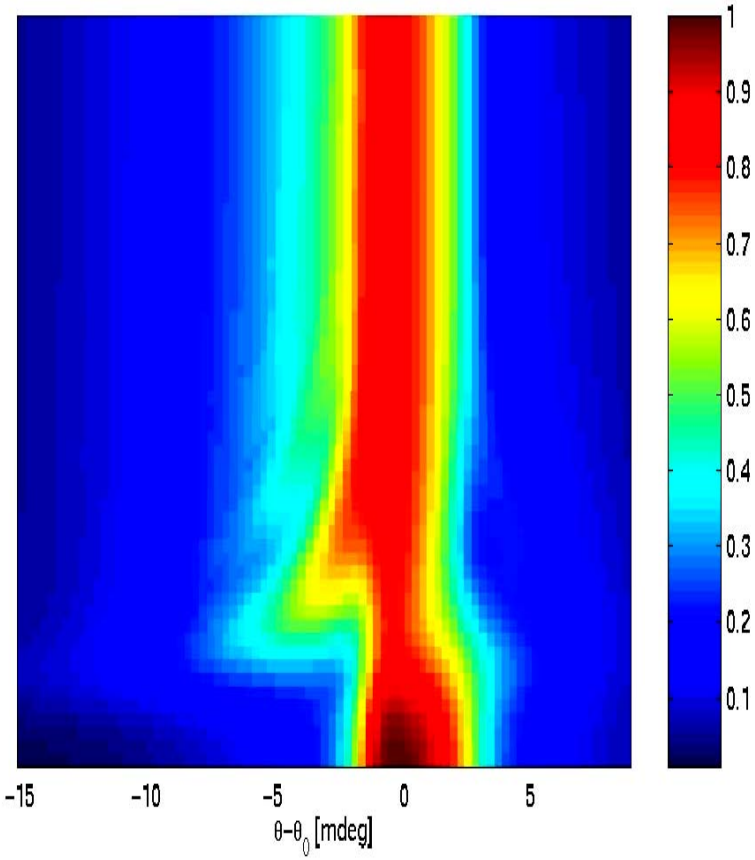
Laser Pump/X-ray Probe

Reis *et al.* Phys Rev. Lett.(86) 2001

Impulsive Strain Generation

(Thomsen *et al.* Phys Rev. B (24) 1986.)

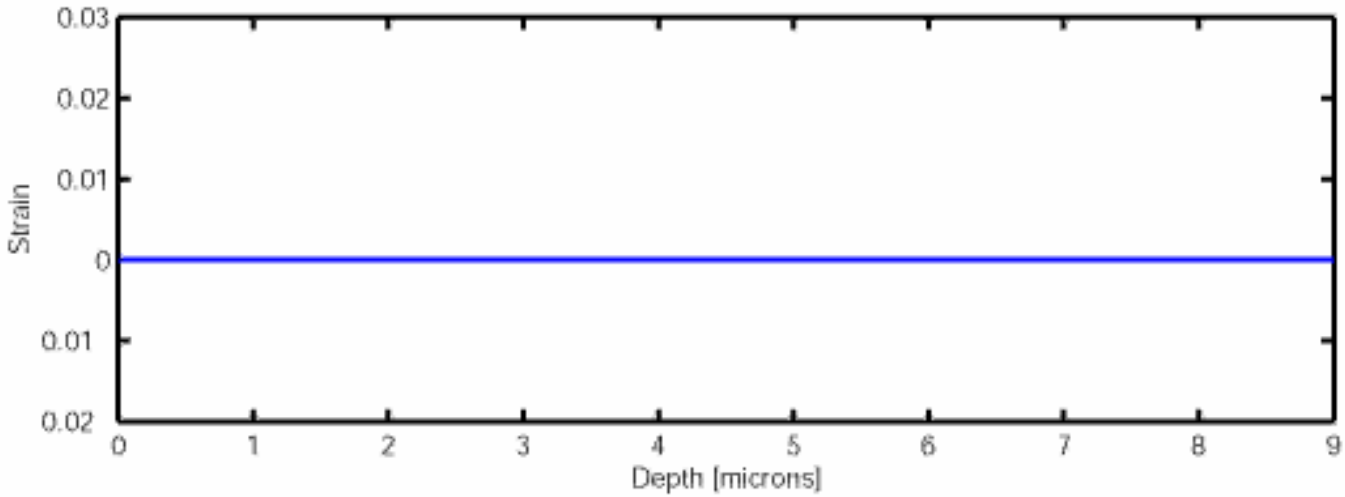


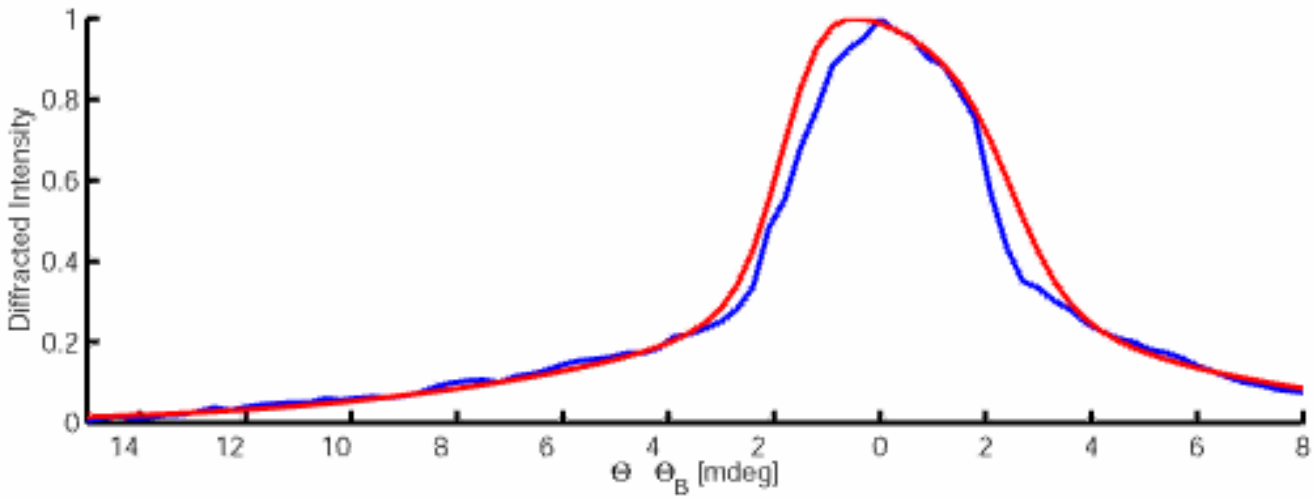


experiment: InSb 111, 10mJ/cm^2

simulation: 100ps & 1.25mdeg conv.

Even on the 100 ps time scale, we still learn things: Partition of energy in the crystal

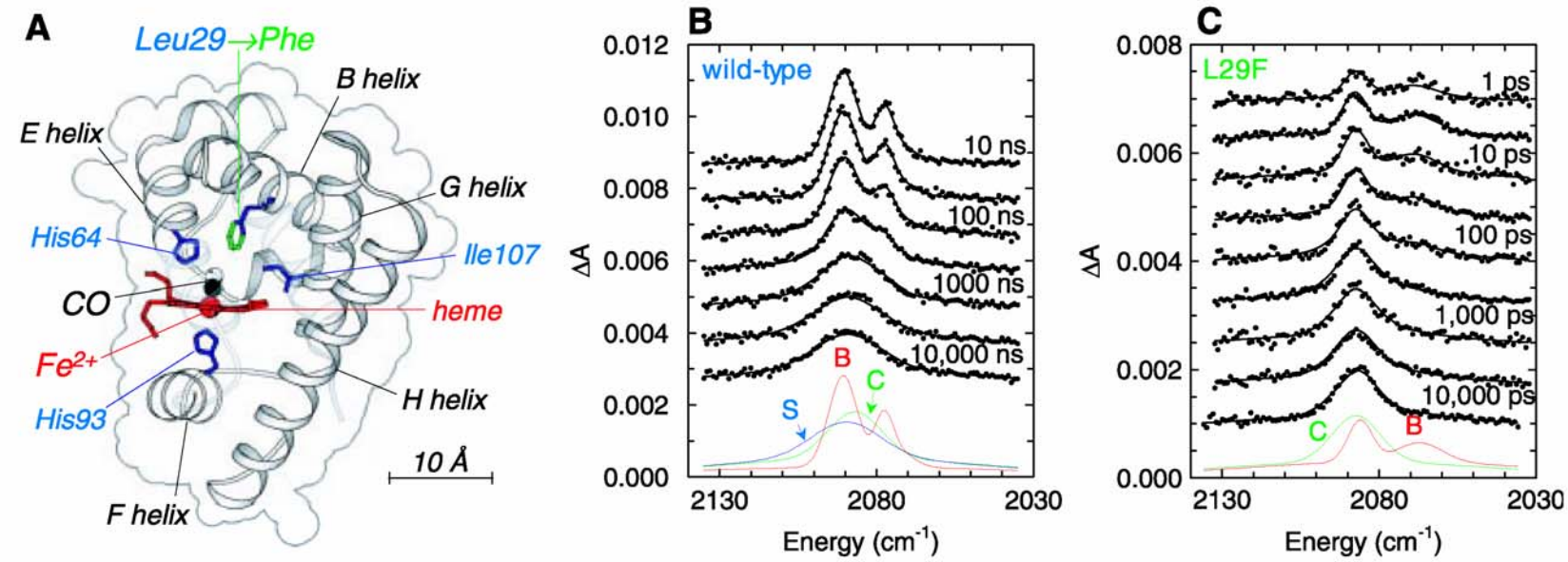




Current state of the art at 100 ps:

Watching a Protein as it Functions with 150-ps Time-Resolved X-ray Crystallography

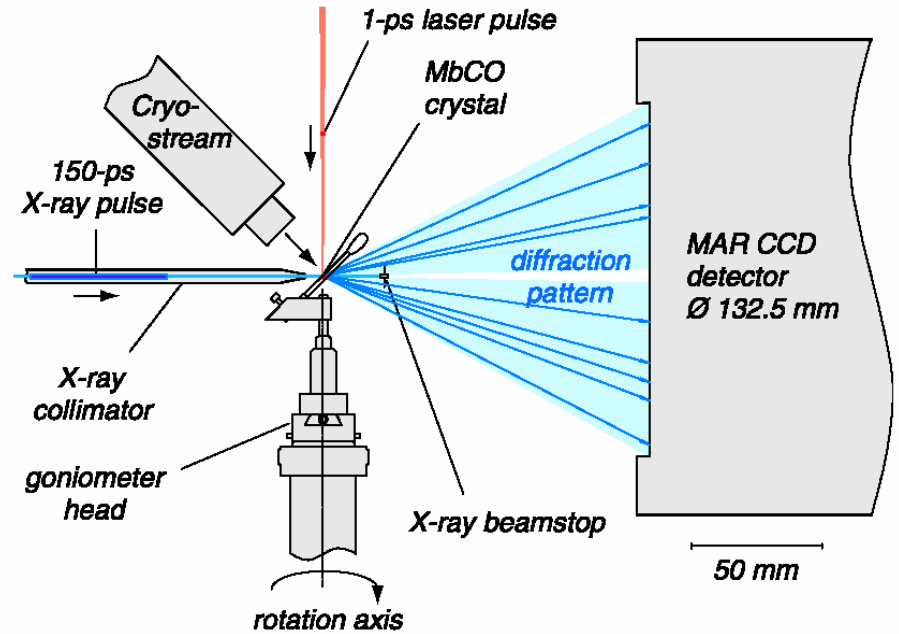
Friedrich Schotte,[1] Manho Lim,[2] Timothy A. Jackson,[3] Aleksandr V. Smirnov,[1] Jayashree Soman,[4] John S. Olson,[4] George N. Phillips Jr.,[5] Michael Wulff, [6] Philip A. Anfinrud[1]

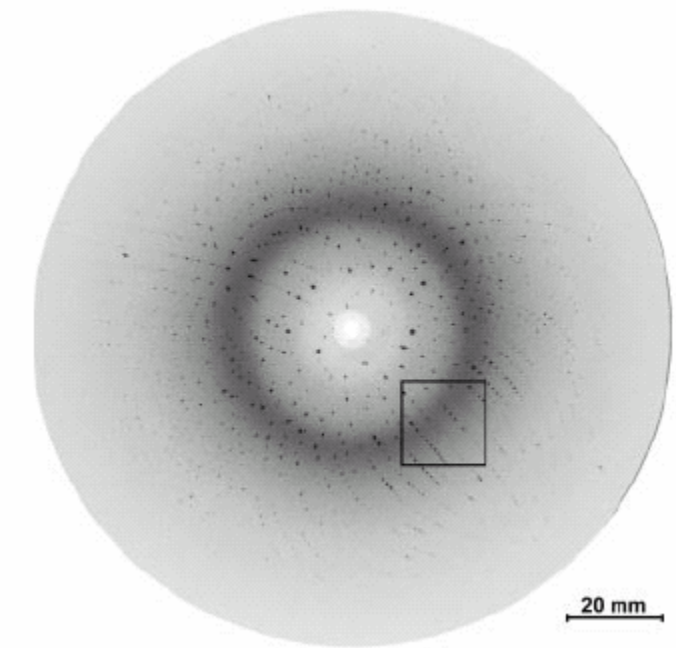


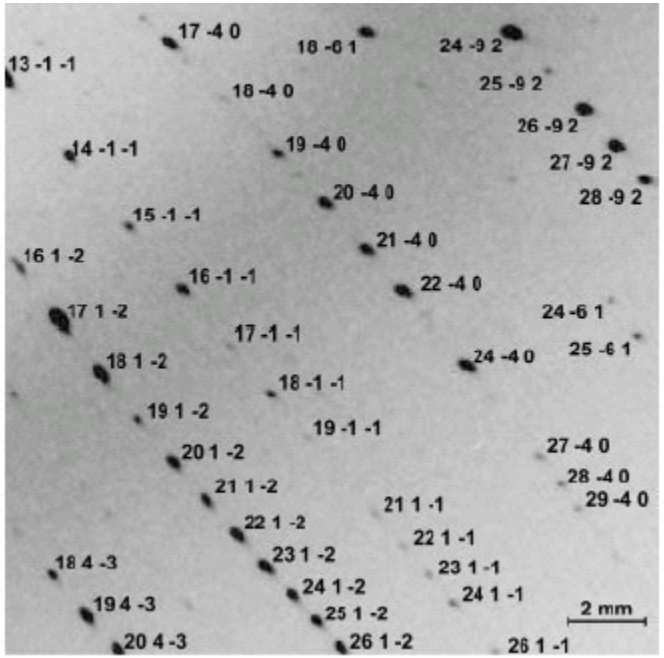
Mid-IR absorption spectra (CO stretch band)

Schotte et al., Science 300, 1944 (03)

X-ray diffraction version of this

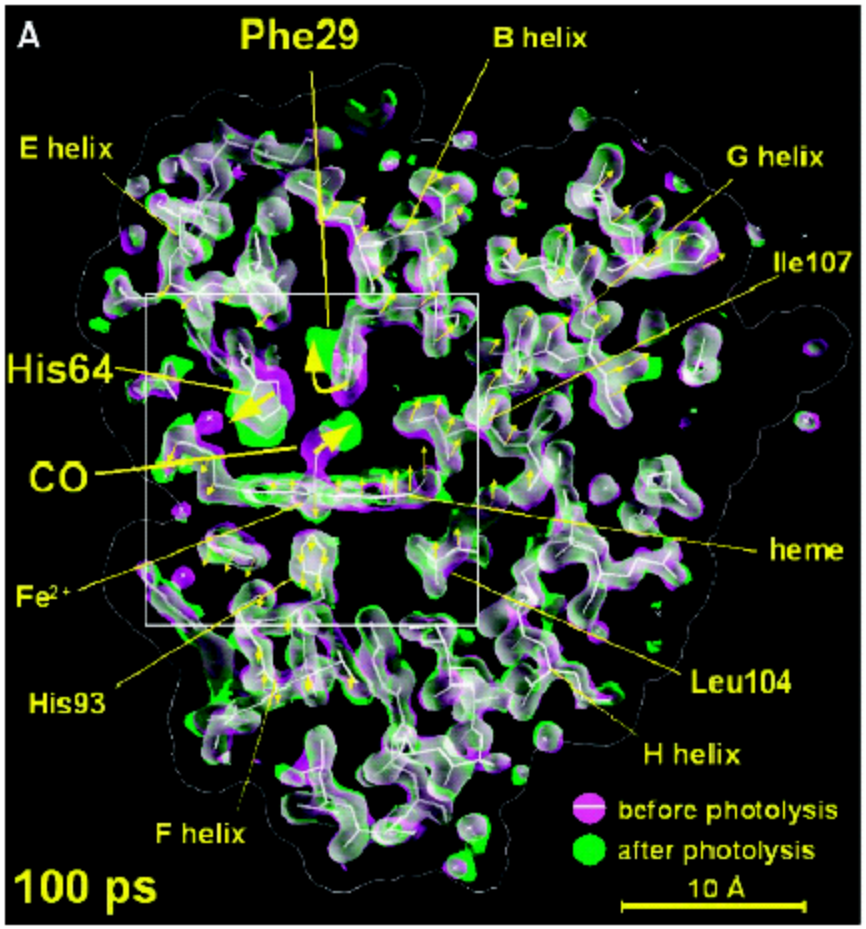


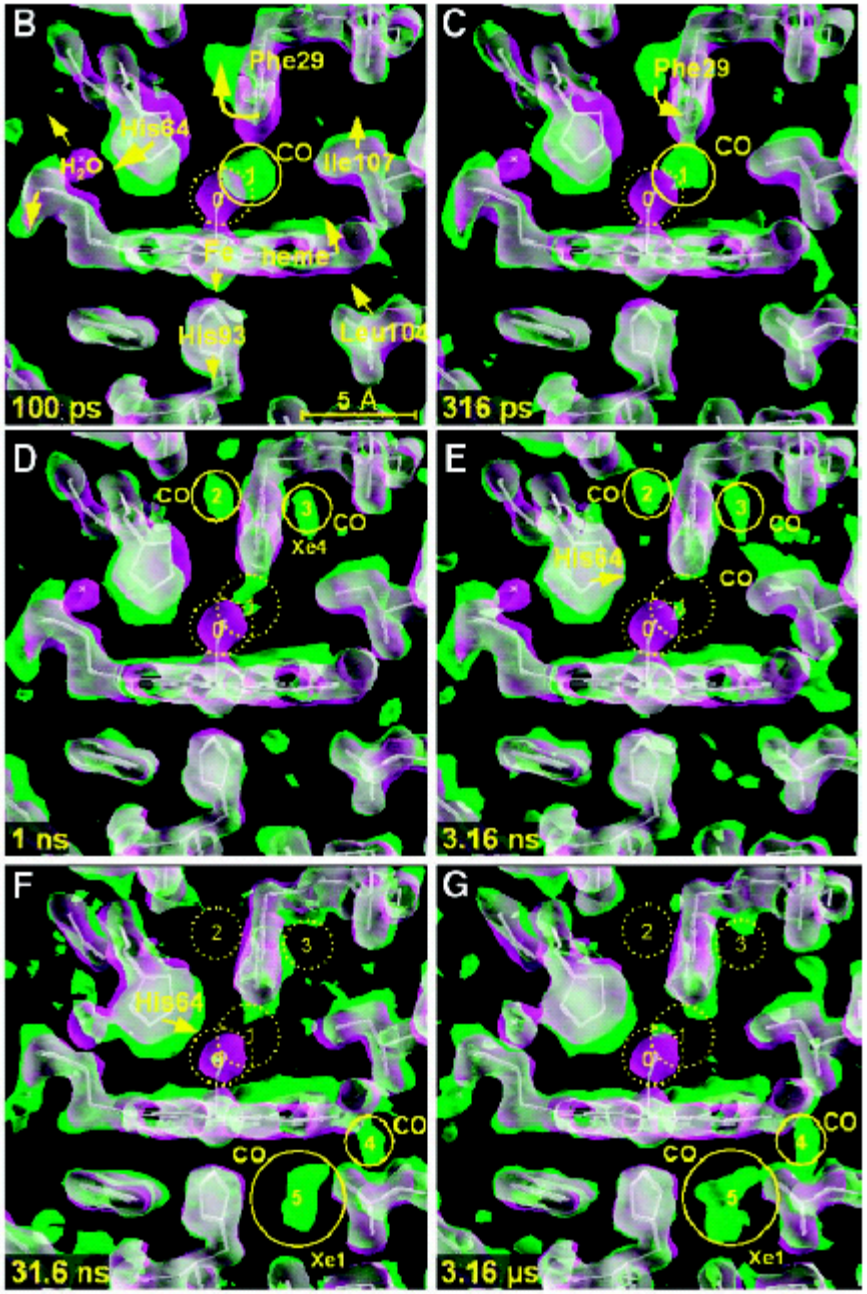




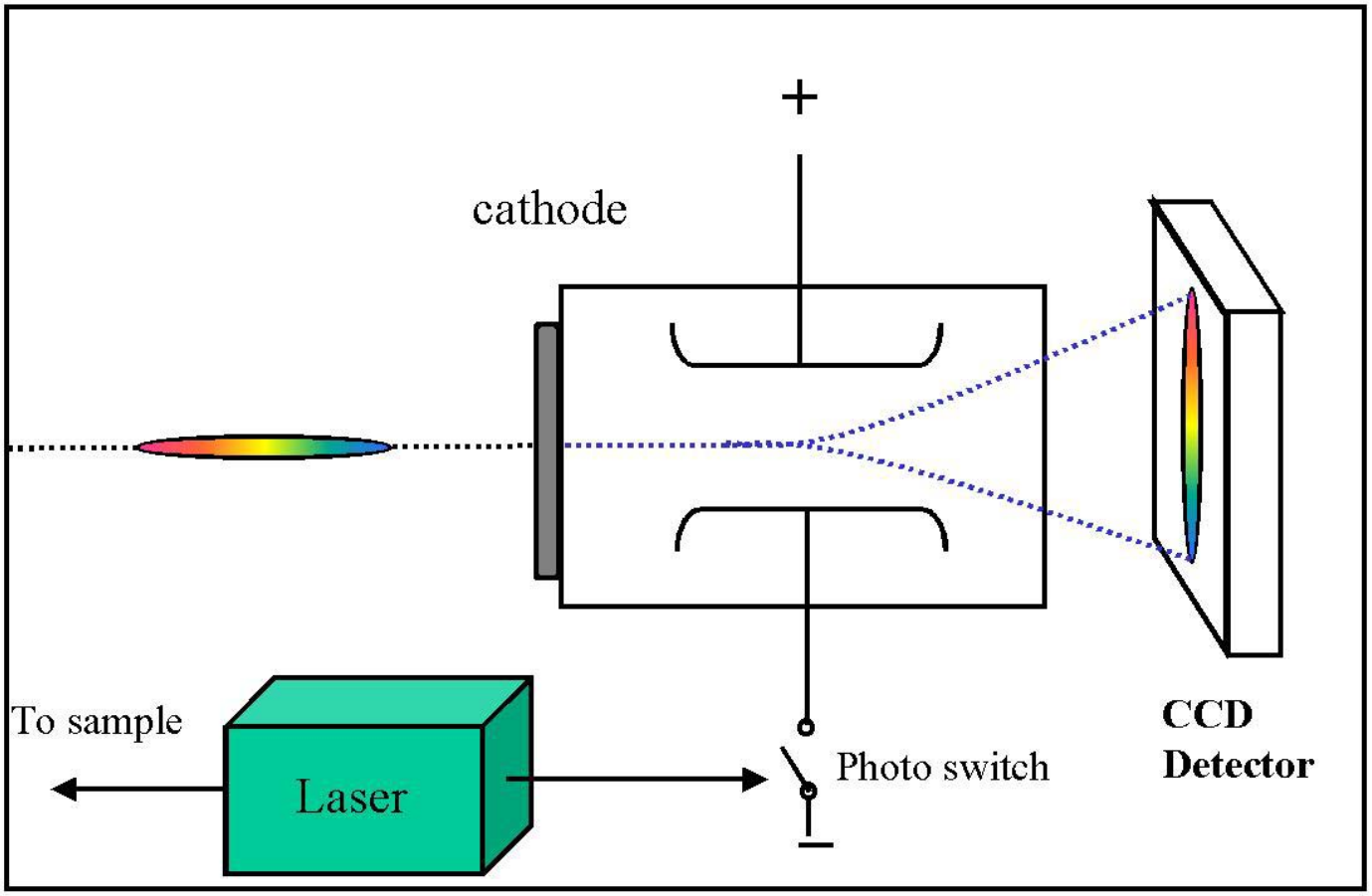
Schotte et al., Science 300, 1944 (03)

FOCUS

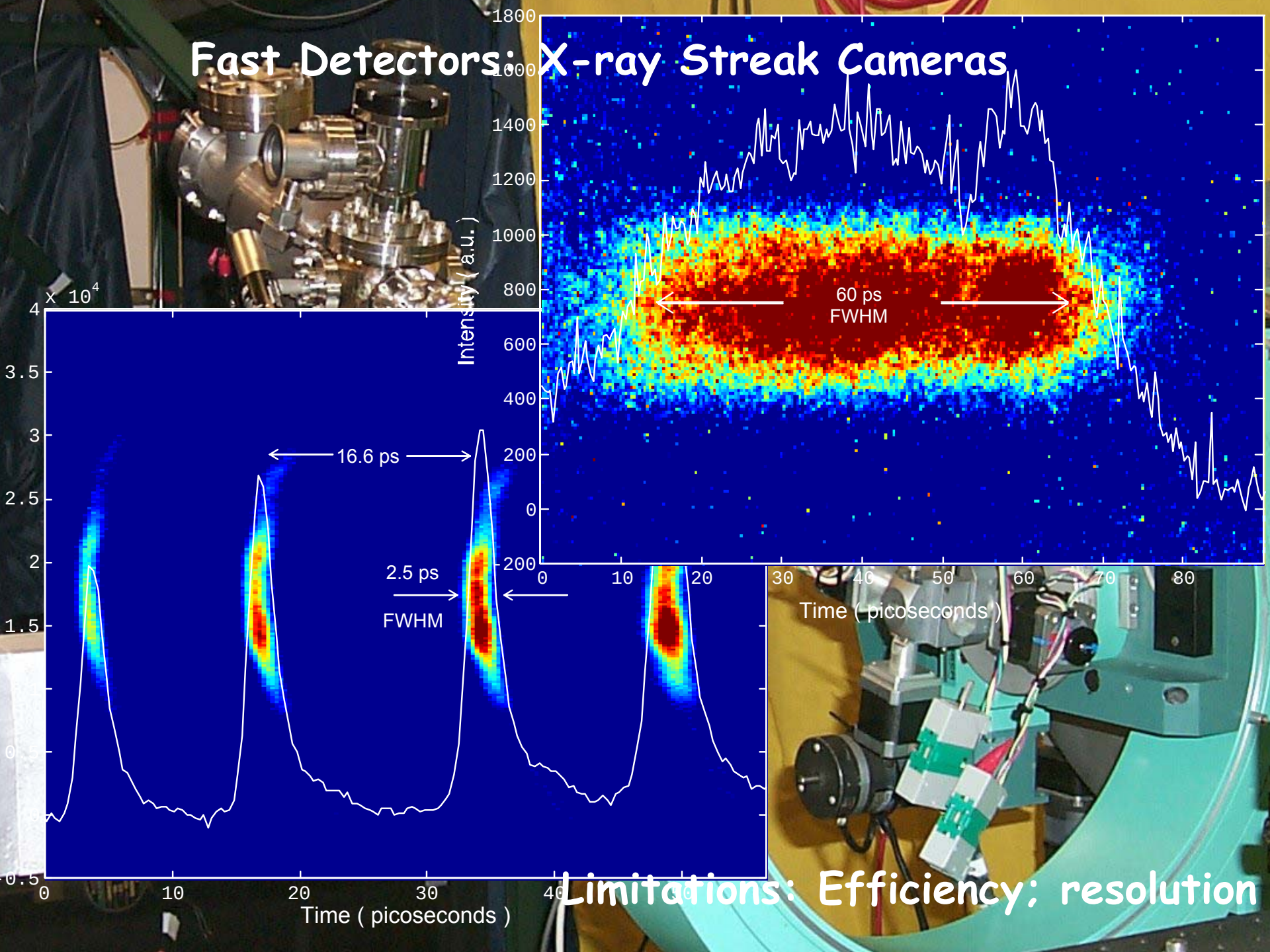

A
Phe29
B helix
E helix
G helix
Ile107
His64
CO
heme
Fe2+
His93
Leu104
H helix
F helix
before photolysis
after photolysis
10 Å
100 ps


B
Phe29
His64
H2O
CO
Ile107
Fe
heme
His93
Leu104
5 Å
100 ps
C
Phe29
CO
316 ps
D
CO
CO
Xe4
1 ns
E
CO
CO
His64
CO
3.16 ns
F
His64
CO
CO
Xe1
31.6 ns
G
CO
CO
Xe1
3.16 µs

Streak Camera: viewing dynamics that are faster than the x-ray pulse

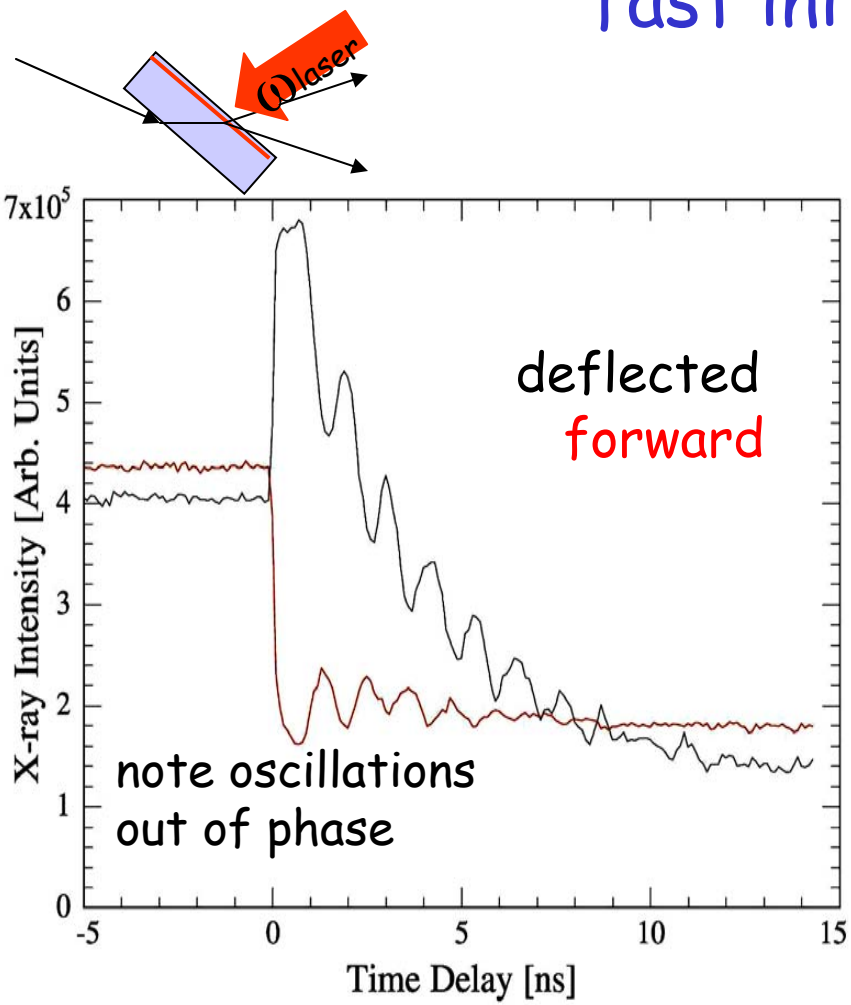


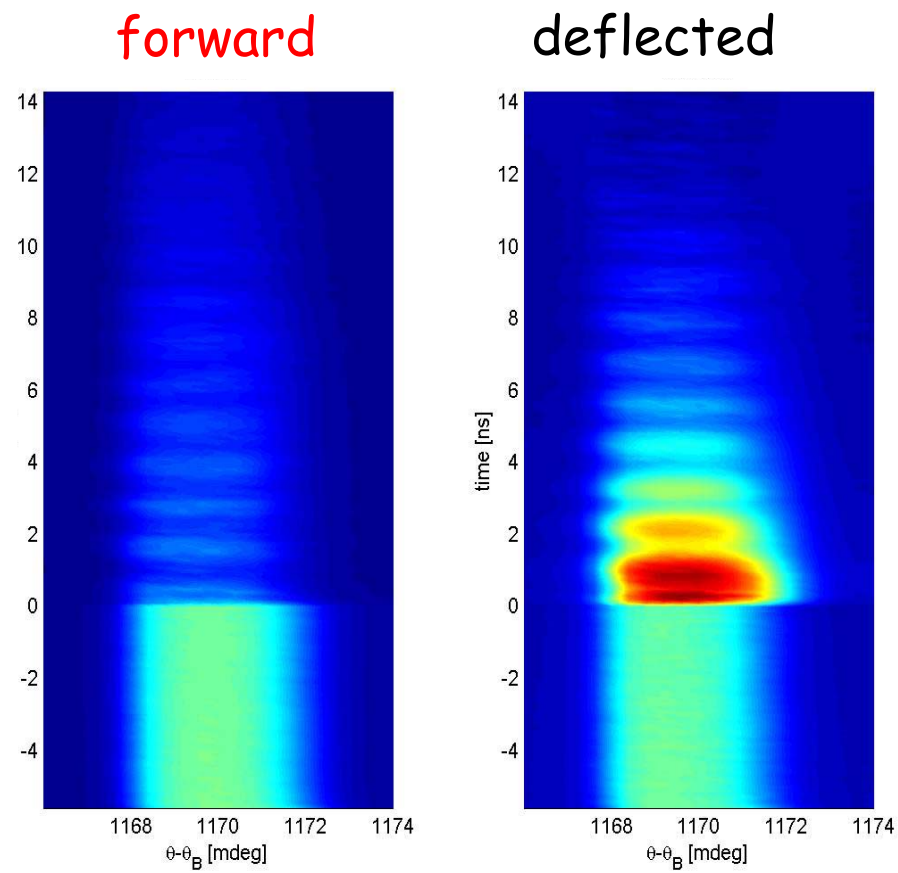
Fast Detectors: X-ray Streak Cameras



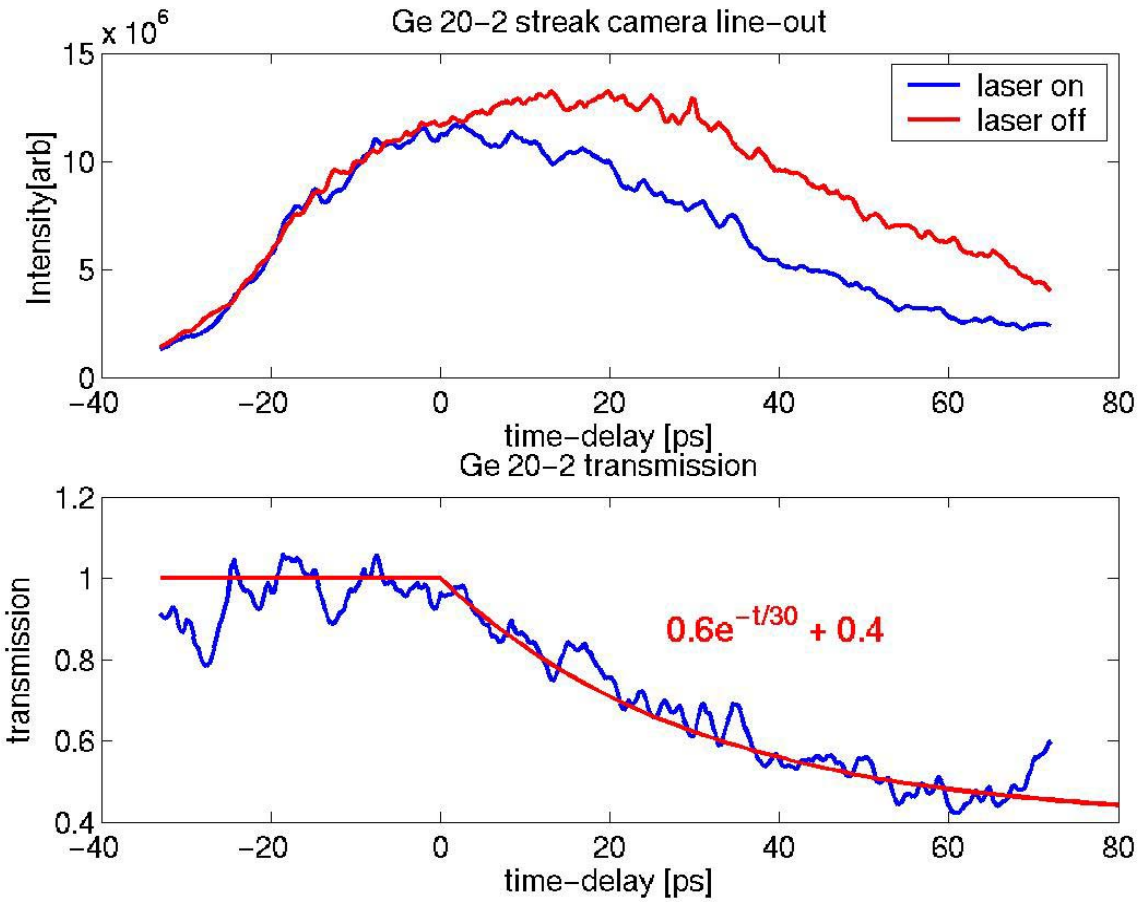
Limitations: Efficiency; resolution

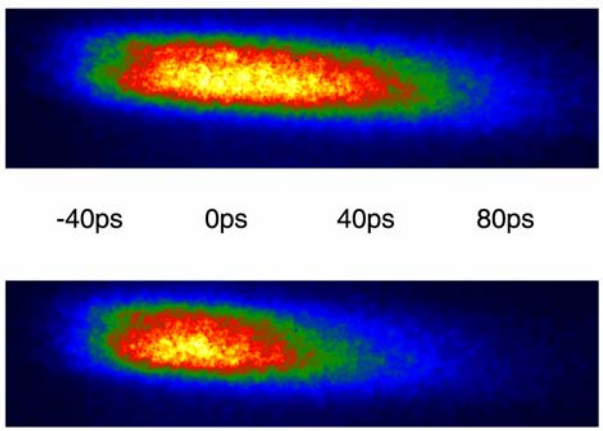
Laser incident x-ray exit face
fast initial switch



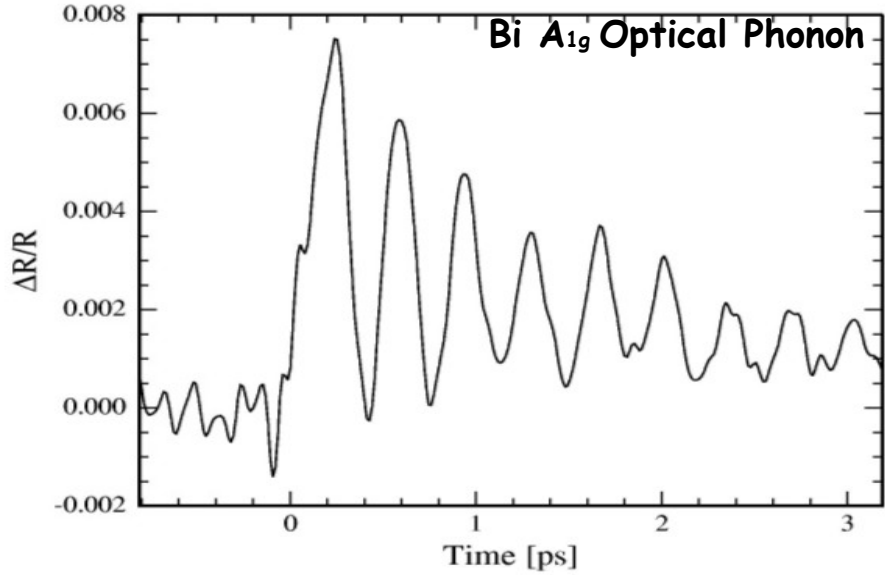


Initial switching ~30 ps
X-ray Streak Camera resolution is about 1 ps

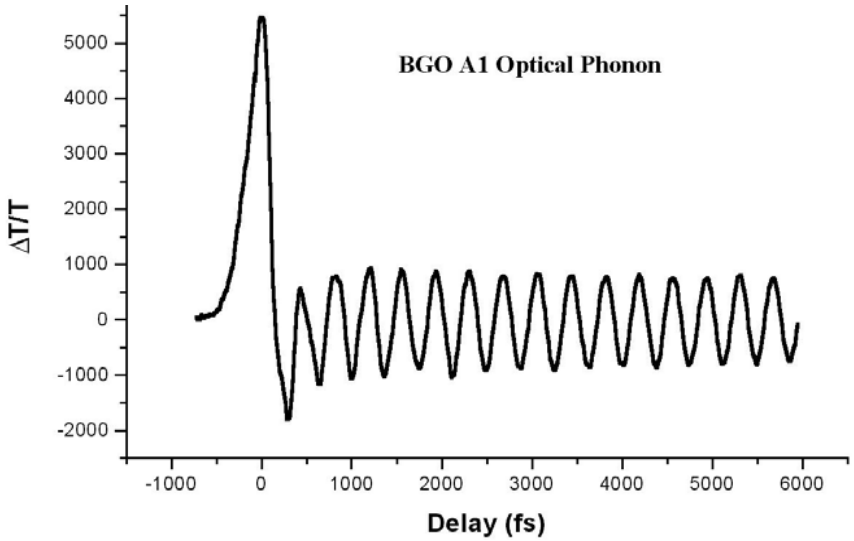


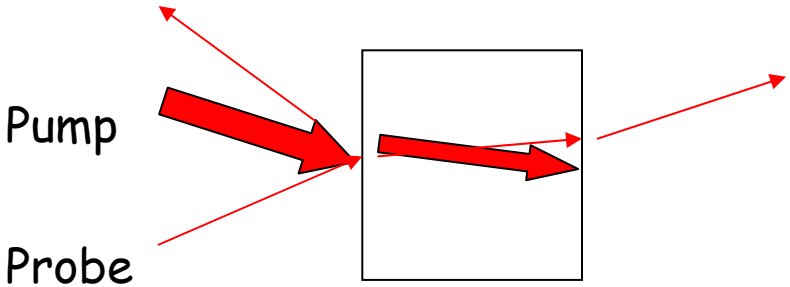


Ultrafast dynamics just beyond the streak camera's resolution



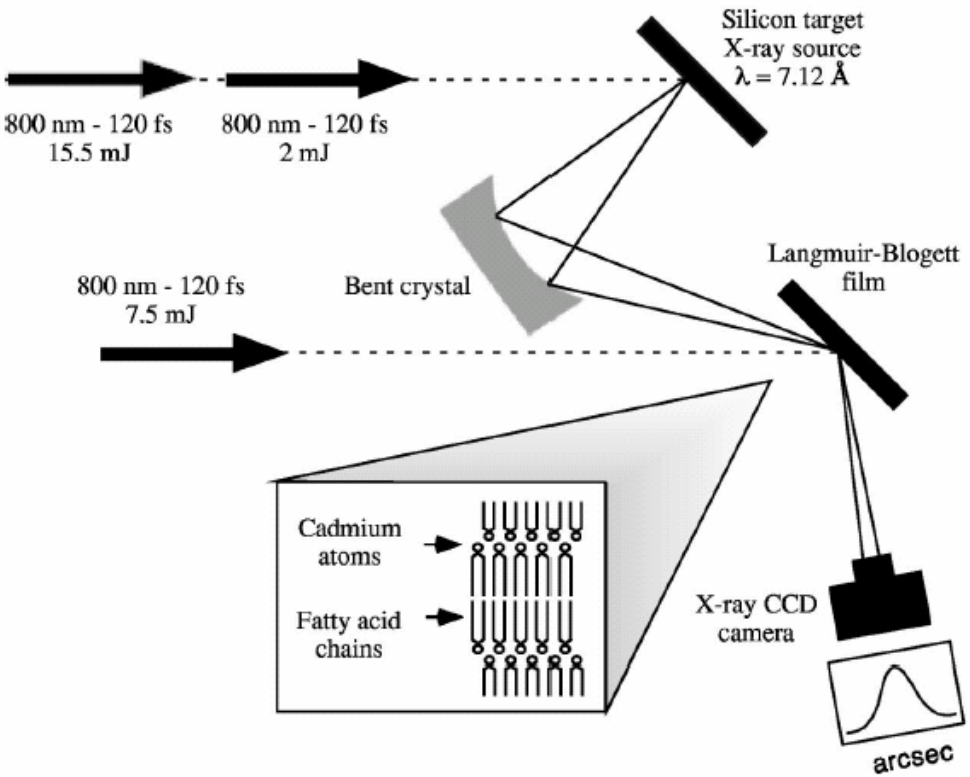
- Near single frequency: Can coherently switch on and off.
- Optical phonons typically in the 3–12 THz range: <50 fs switches possible.
- Create switch by inducing an optical phonon super-lattice with $k \sim 1/\lambda_{laser}$.
- Or, In "Forbidden" and "Weak"Reflections can use k~0 phonons.

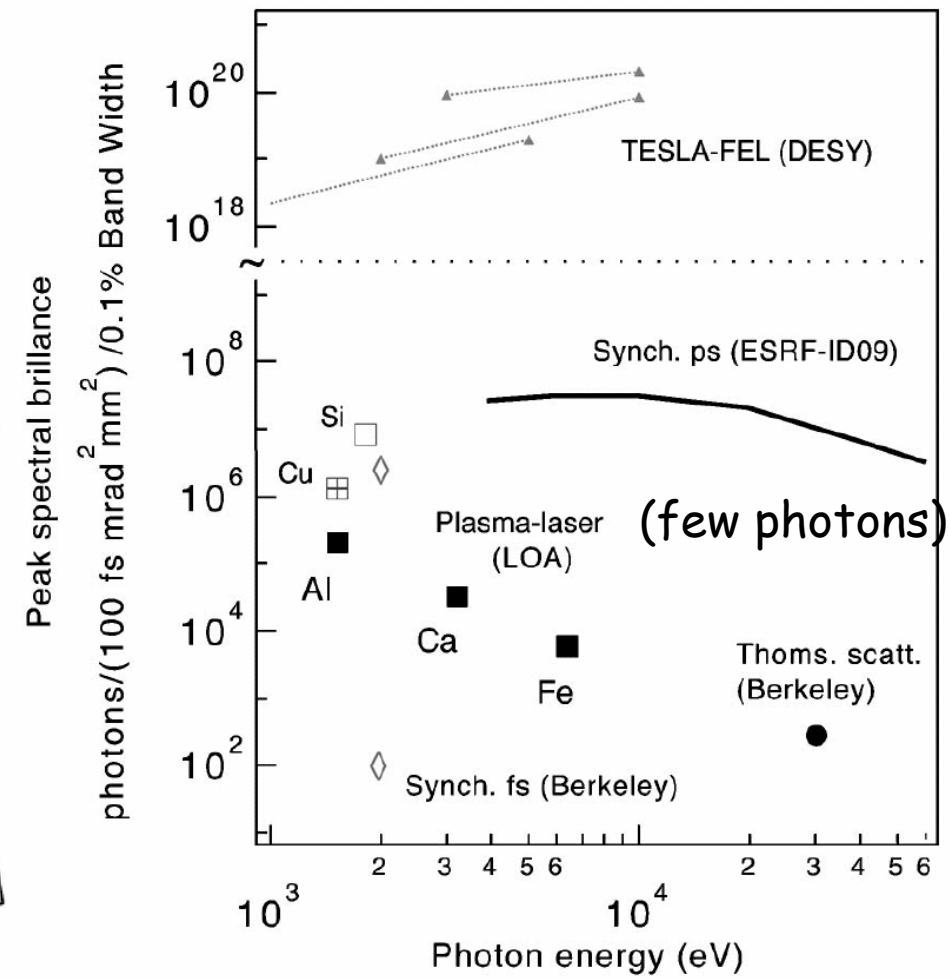




Experimental setup

Other ways to make hard x-rays: Laser plasmas





Source: Rousse et al., RMP 73, 17 (2001)

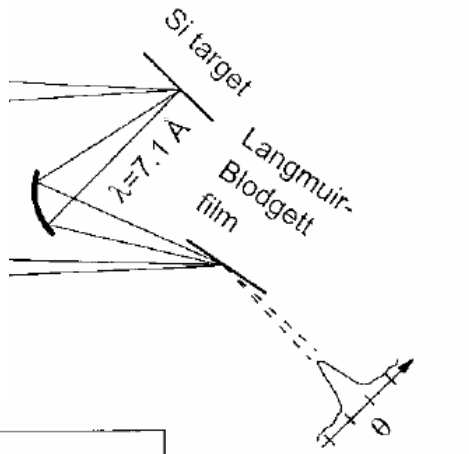
Femtosecond time-resolved X-ray diffraction from laser-heated organic films

Tradeoffs: Laser plasma source

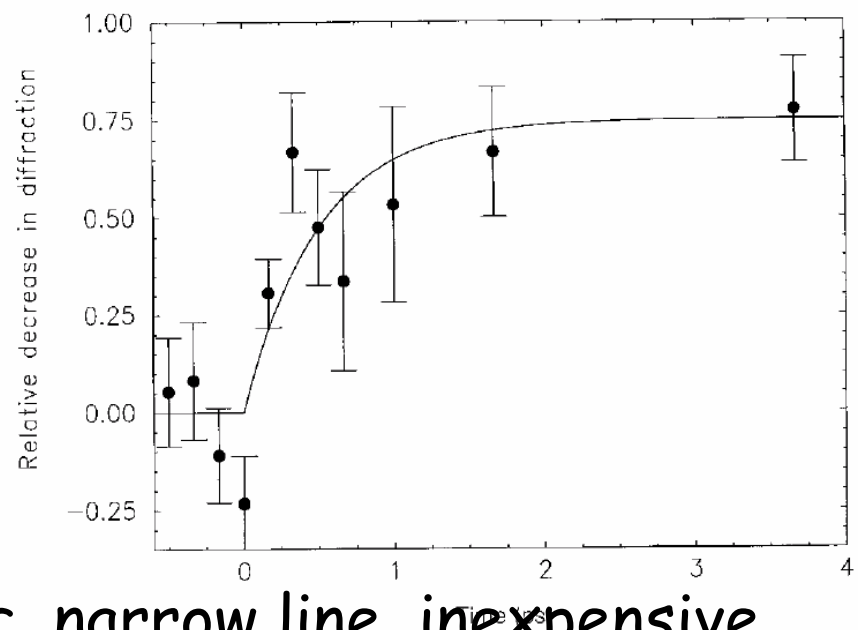
Rischel, Rousse, et al.

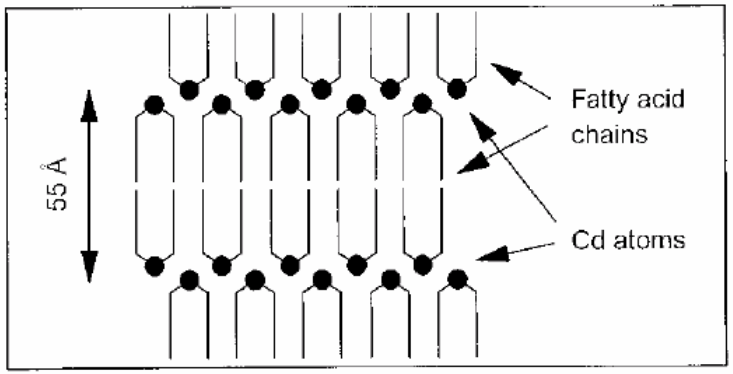
NATURE | VOL 390 | 4 DECEMBER 1997

15mJ, 150 fs
On a solid target



Diffraction detected by CCD camera





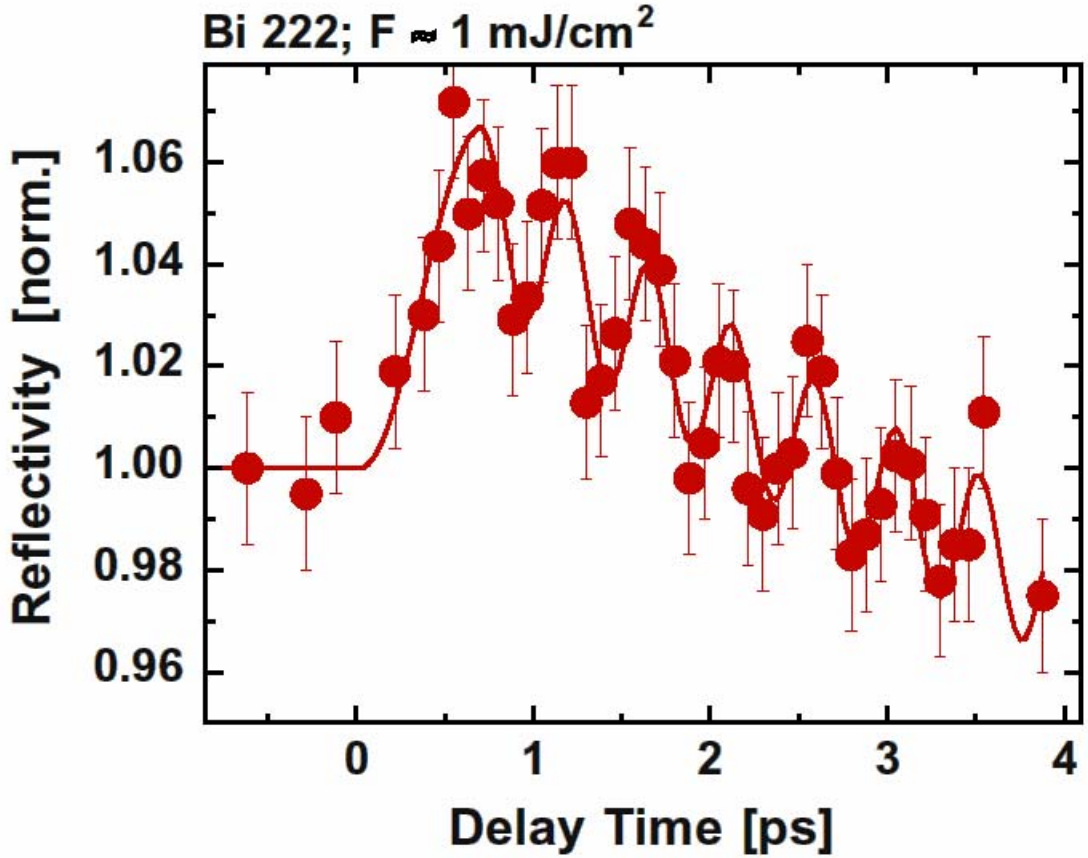
Structure of Langmuir-Blodgett film

<1psec, narrow line, inexpensive

Very few useful photons

Phonon oscillations viewed on a strong Bragg reflection, using x-rays from a laser plasma

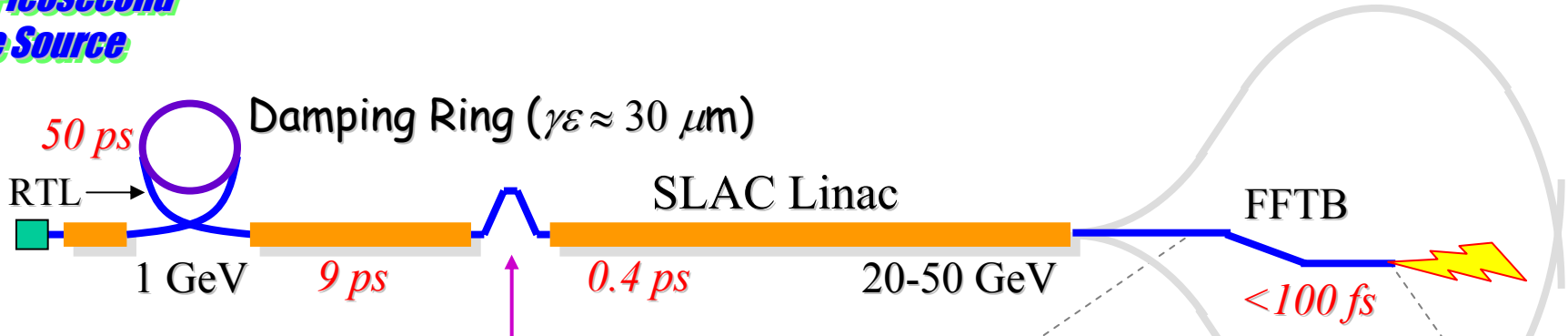
(Sokolowski-Tinten et al., Nature **422**, 287 (2003))



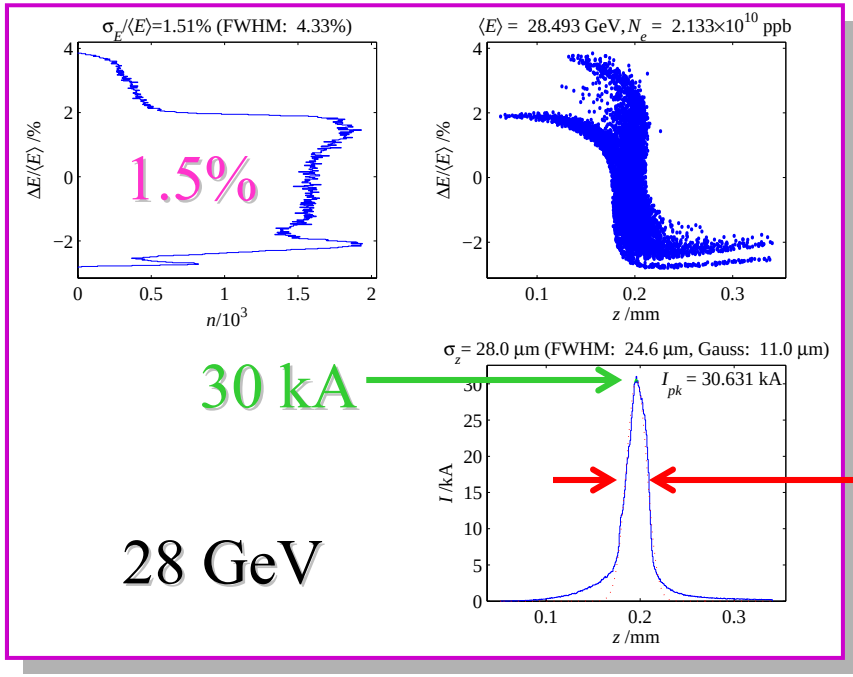
<1psec, narrow line, inexpensive, but very few useful potons

Sub-Picosecond Pulse Source

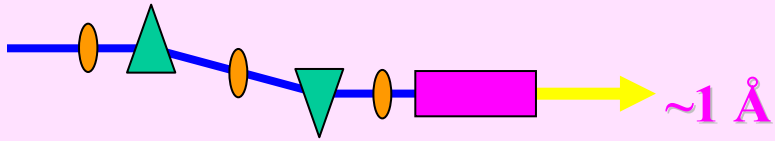
Short Bunch Generation in the SLAC Linac



Add 12-meter chicane compressor in linac at 1/3-point (9 GeV)

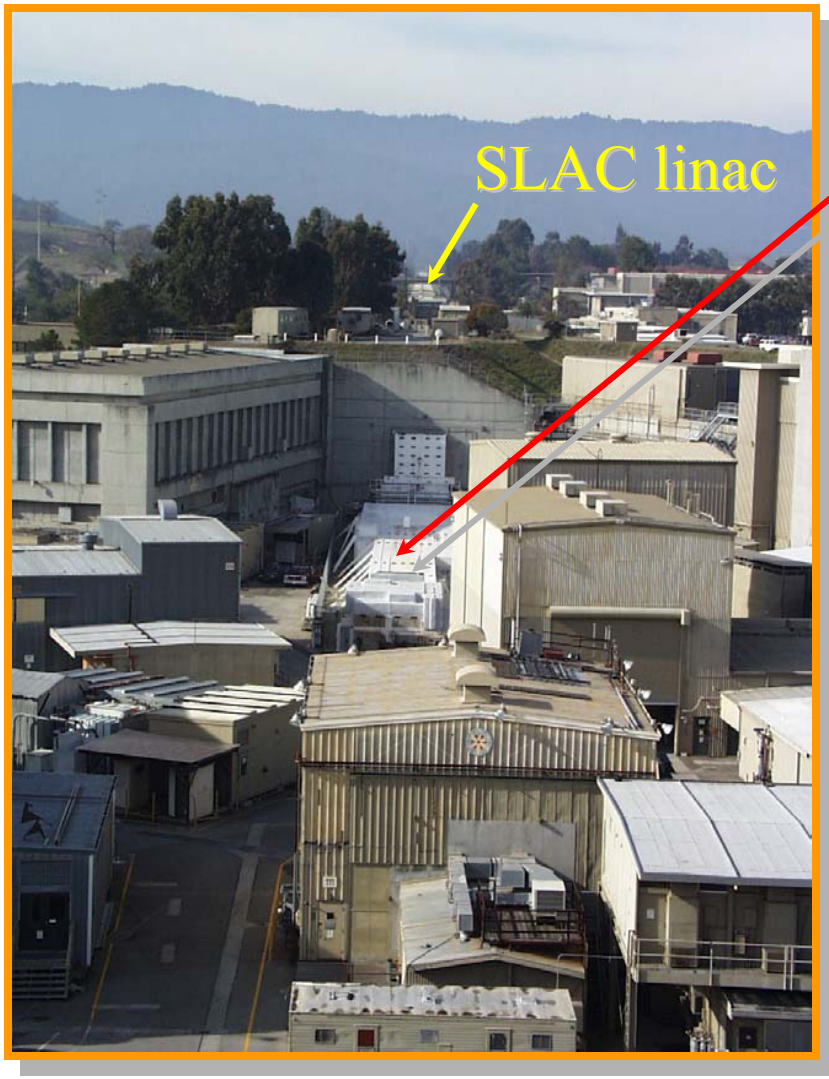


Existing bends compress to **<100 fsec**

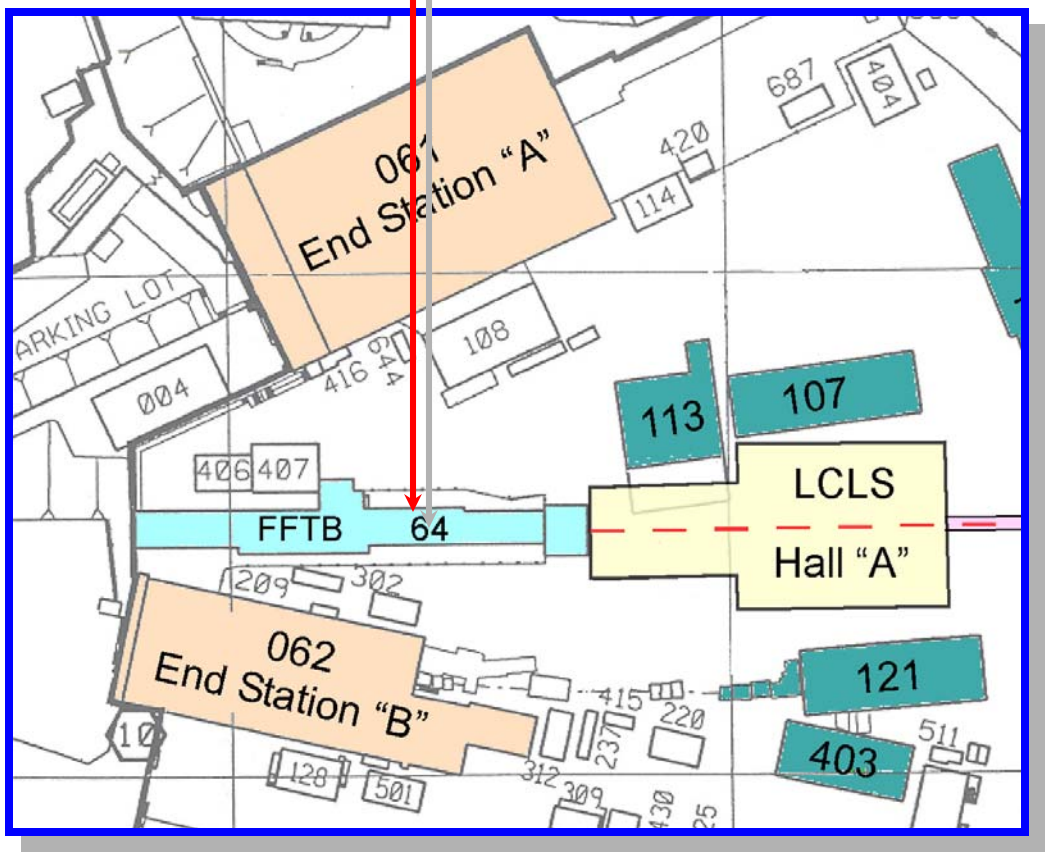


80 fsec FWHM

Where: Linac and FFTB Hall



add undulator to FFTB hall at end of linac



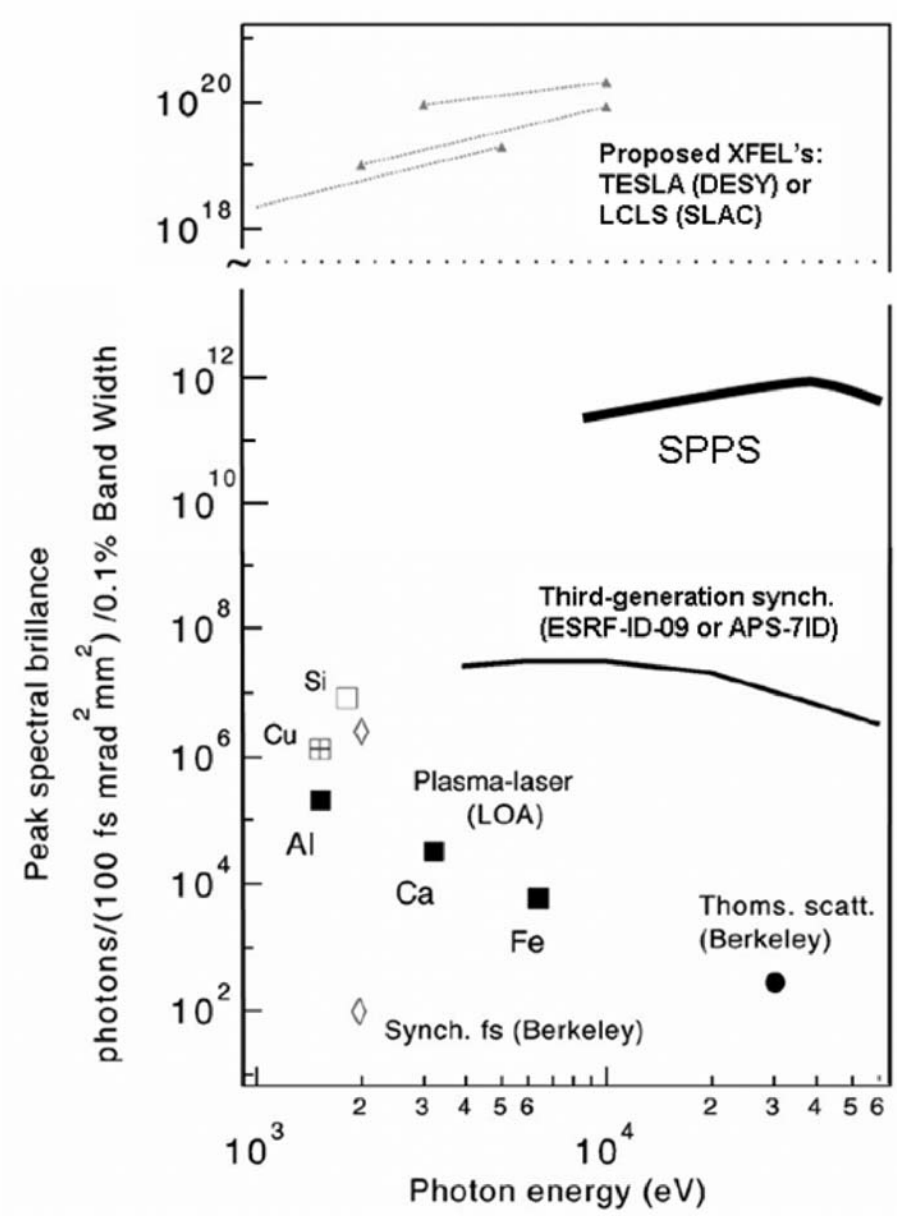
Sub-Picosecond Pulse Source

Why is SSRL/SLAC doing this?

- Accelerator R&D: bunch compression
- Development of methods to cross correlate the optical pump laser with the x-ray beam
- Develop electron and photon beam pulse length diagnostics for the future XFEL
- Ultra-fast x-ray scattering
- It's relatively cheap (if you don't count SLAC)

Sub-Picosecond Pulse Source

SPPS is presently the world's most brilliant source of hard x-rays

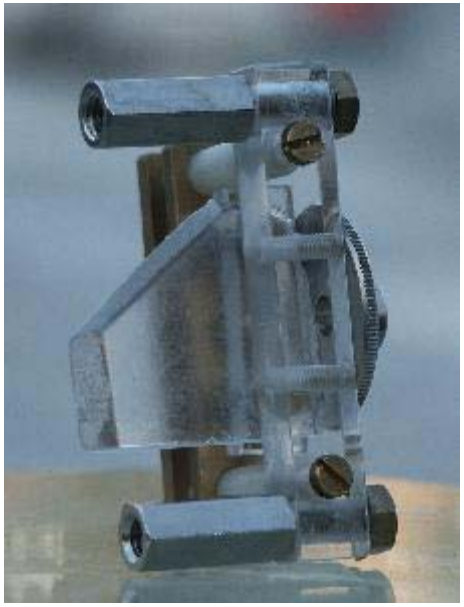


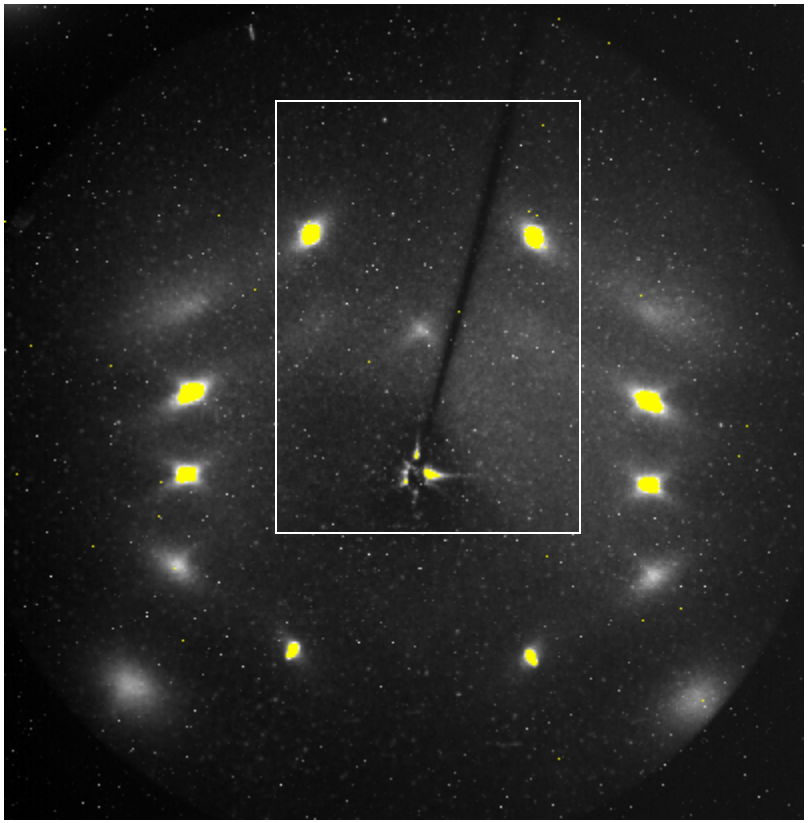
Sub-Picosecond Pulse Source

Diffraction from Salol

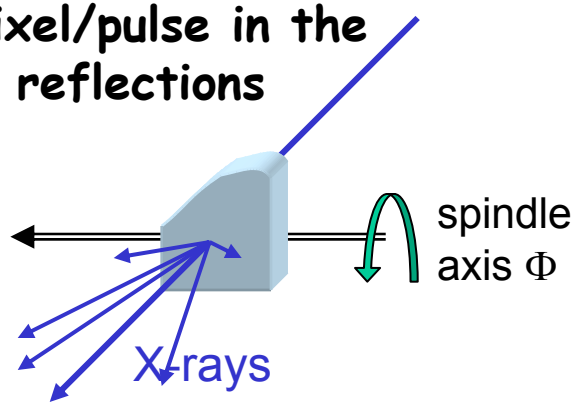
Salol
Phenylsalicylate

$C_{13}H_{10}O_3$
Orthorombic
Pbca
a=7.961 Å
b=11.258 Å
c=23.402 Å
$\alpha = \beta = \gamma = 90°$
$\Delta\theta$ (0 16 0)=0.3 mdeg





300 ph/pixel/pulse in the strongest reflections



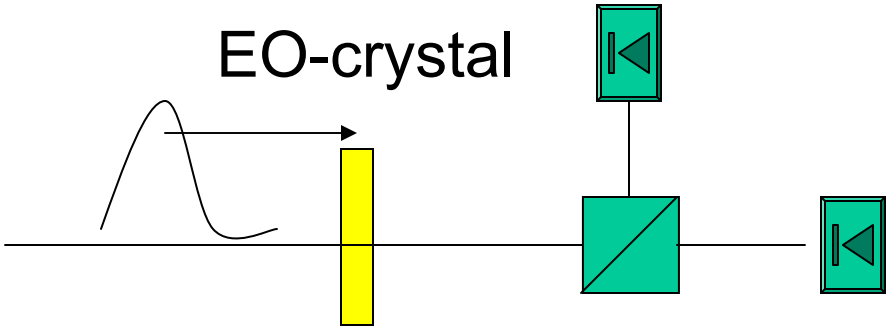
~10^6 photons/pulse
1 x 1 mm^2
1% bandwidth
300 s, 10 Hz

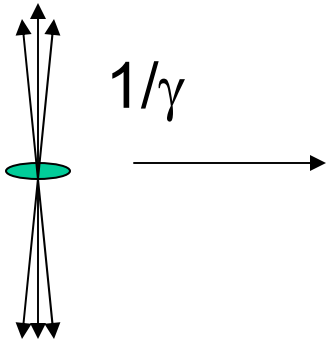
9.365 keV
image int. CCD
at 120 mm

Collaboration & samples E. Weckert, HASYLAB

Sub-Picosecond Pulse Source

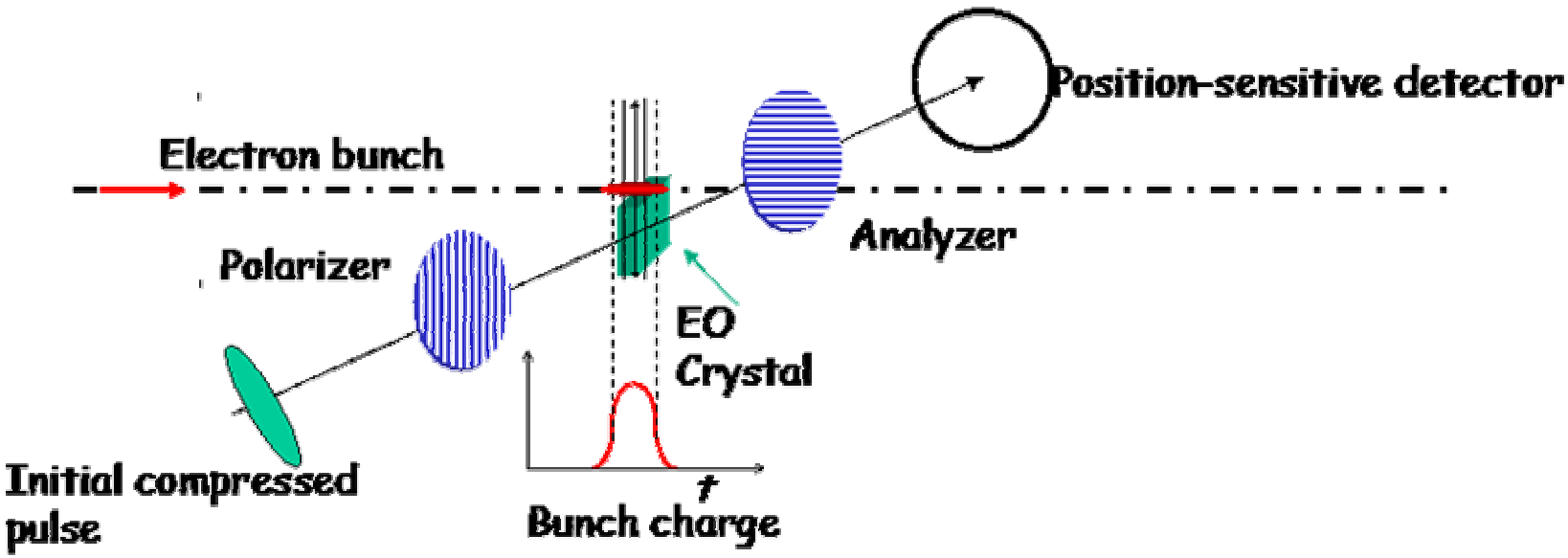
Laser-X-ray synchronization can use EO sampling of the relativistic e-beam





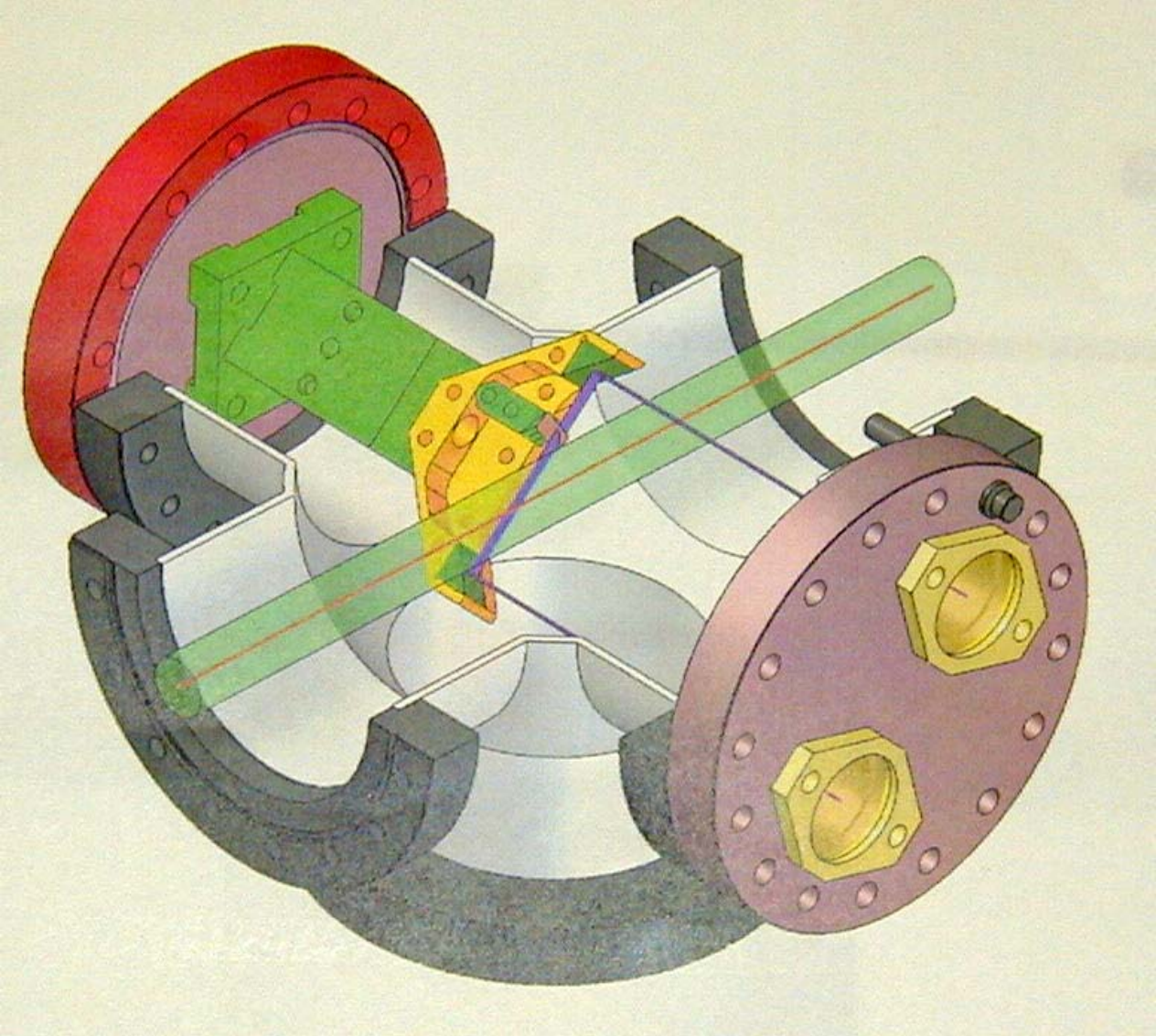
Sub-Picosecond Pulse Source

We can sweep the laser past the e-beam for single-shot cross-correlation



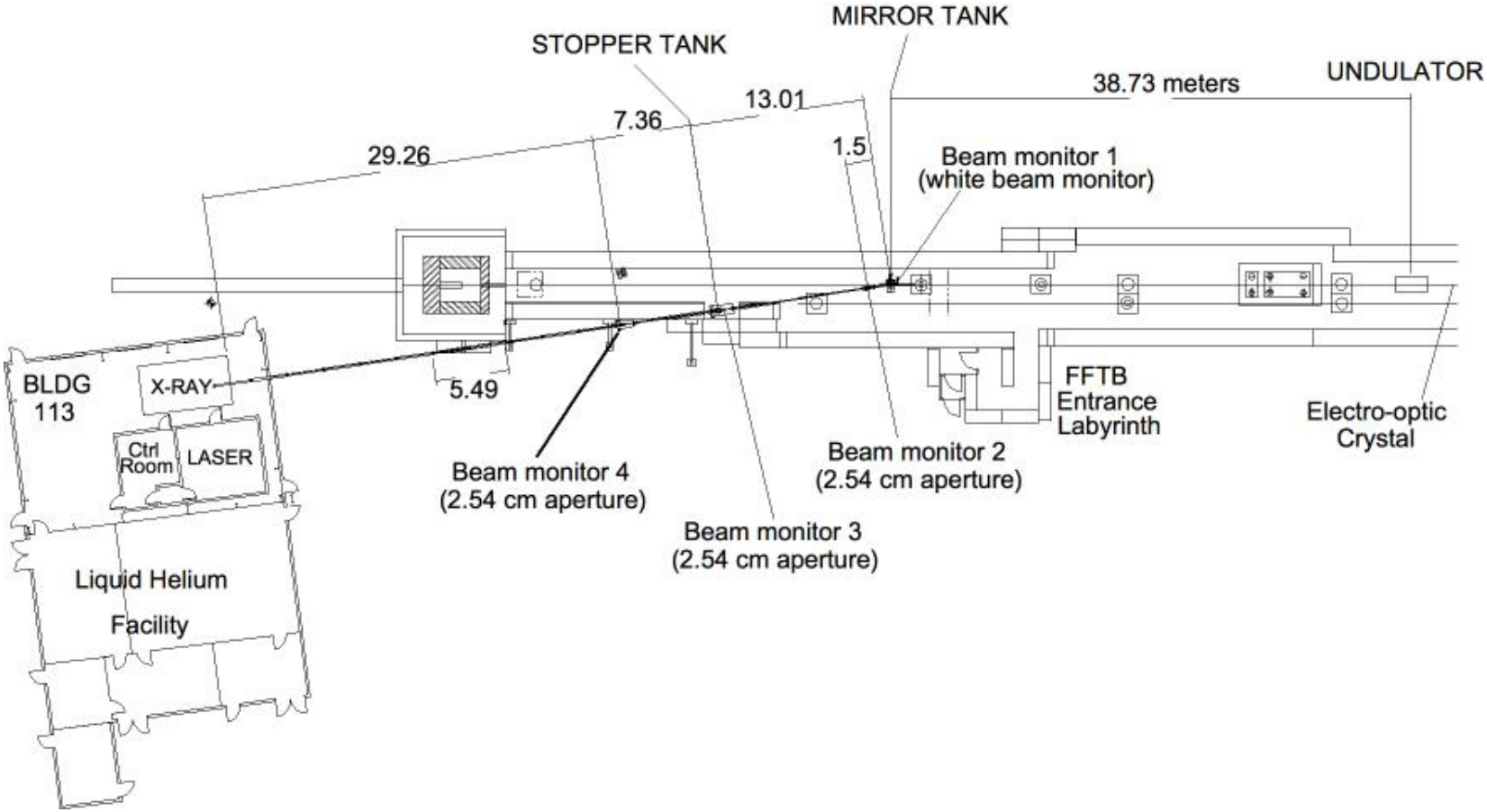
Sub-Picosecond Pulse Source

EO Chamber (Eric Bong)



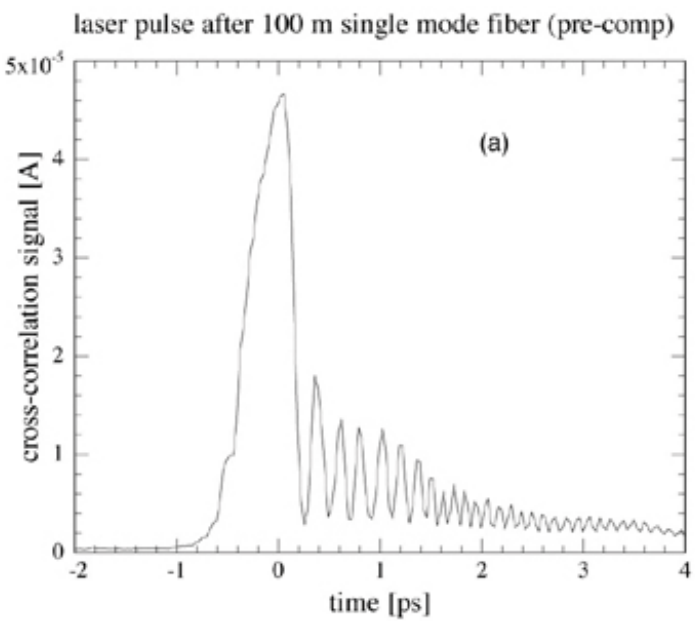
Undulator,
View upstream
Dave Fritz, Soo Lee, David Reis

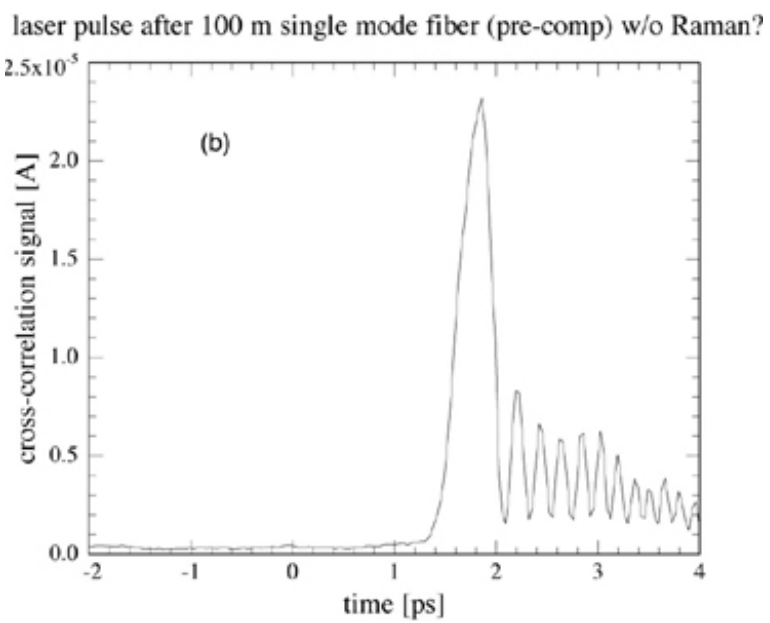
Sub-Picosecond Pulse Source



Sub-Picosecond Pulse Source

Precompensation for GVD.
Cross-correlation after 100m SM PM fiber

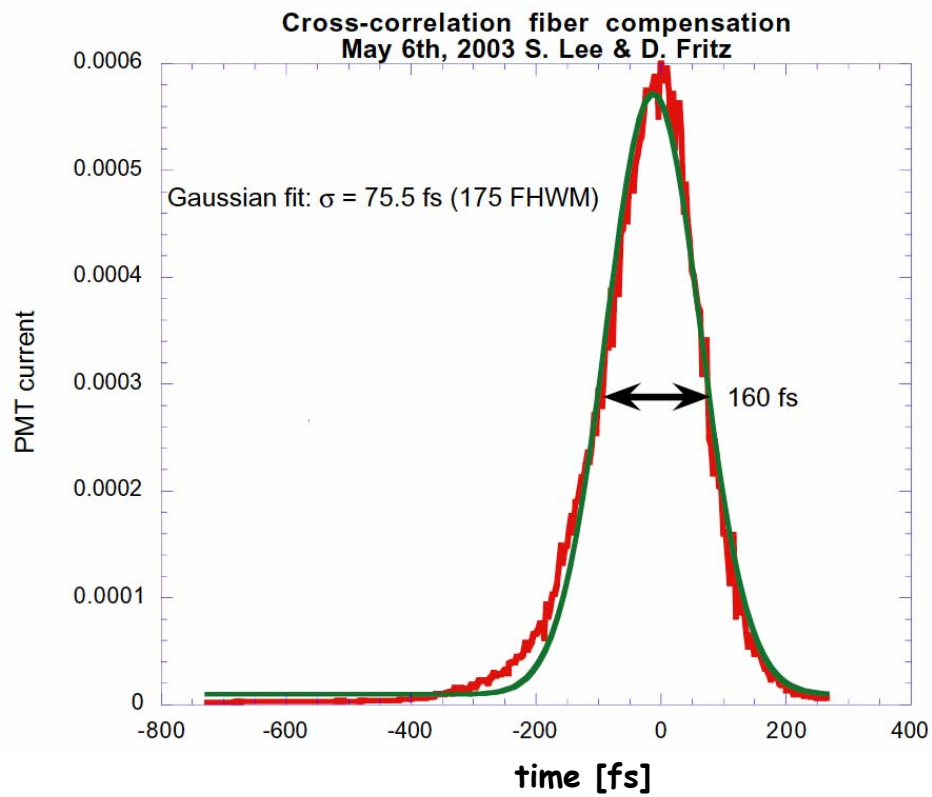


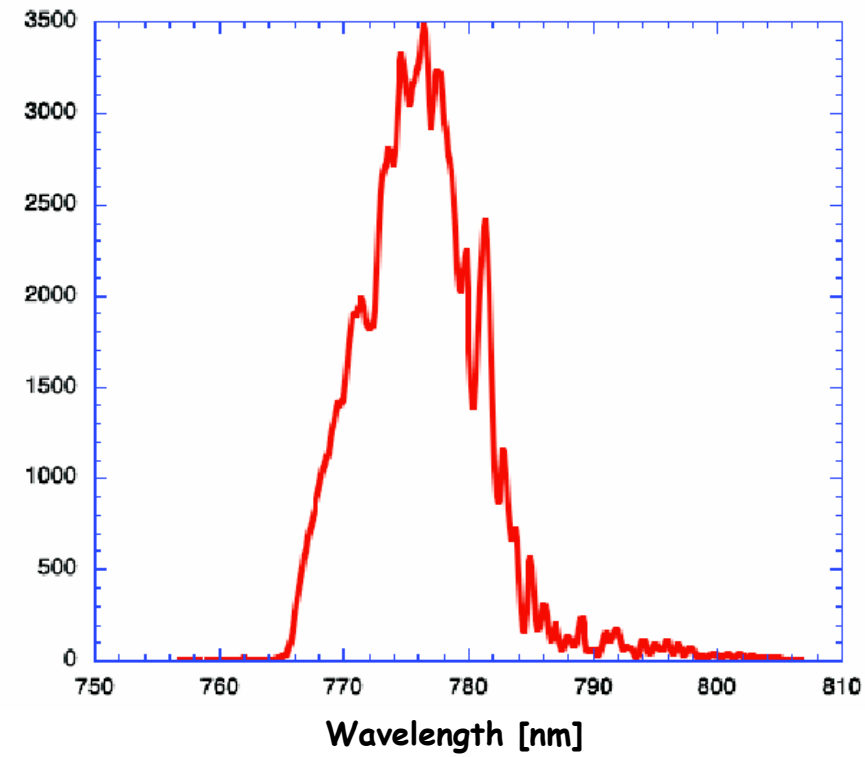


- Uncompensated and additional higher order phase.
- Nonlinearities beginning to kick in at exit of fiber
- Δt_0 due to "long" term drift of fiber

David Reis

Pulse Shaper + Learning Algorithm finds < 2X transform limited pulse

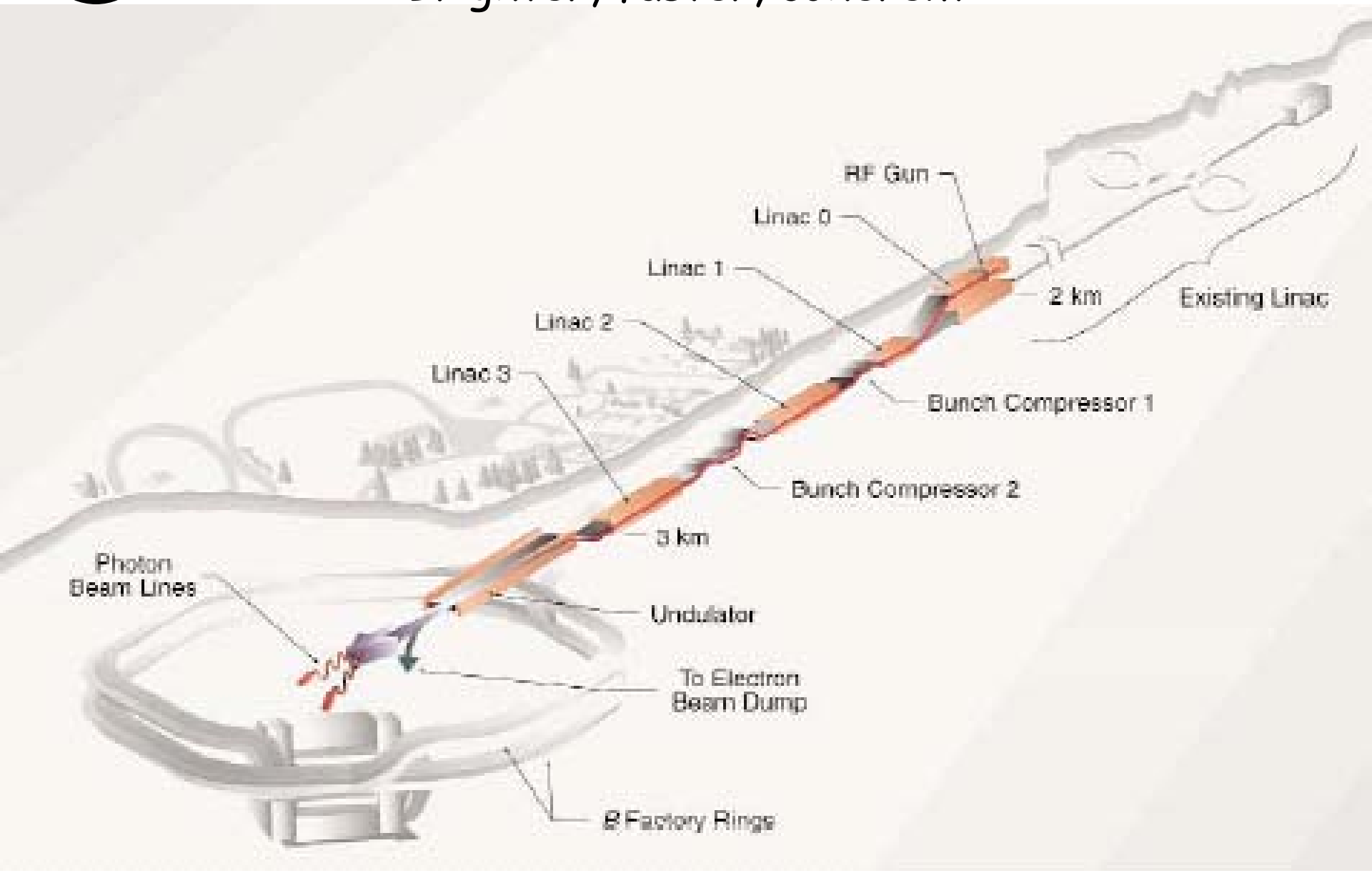




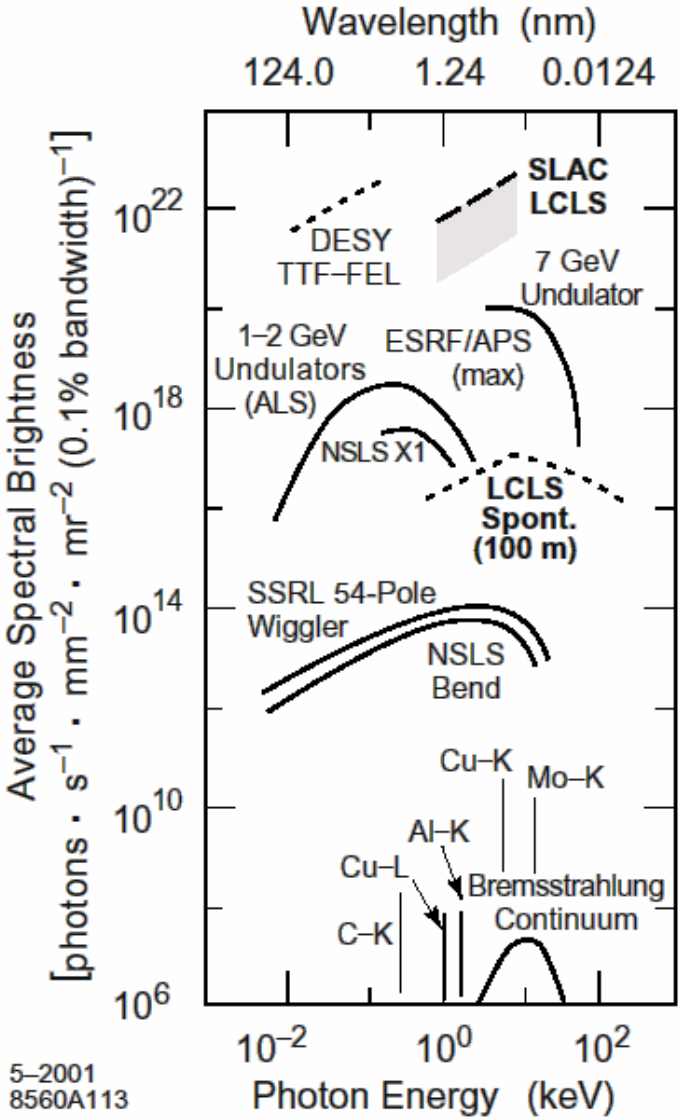
Feedback on fiber length (not shown) , holds time delay fixed.

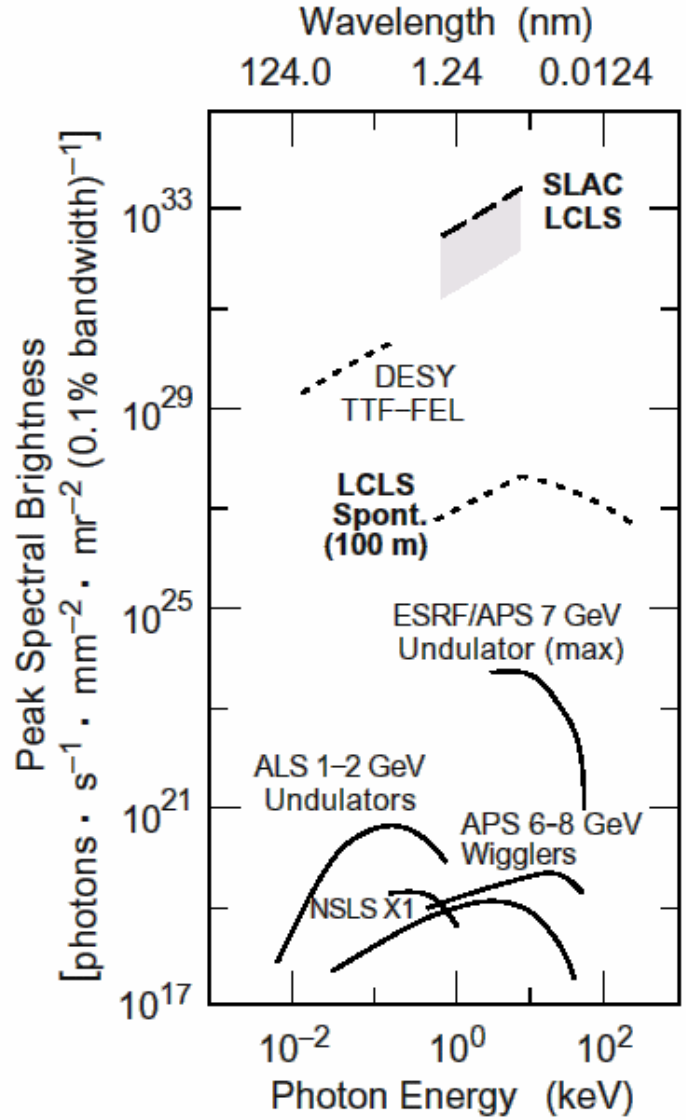
David Reis

Free-Electron Lasers
Brighter, faster, coherent



Comparing apples and eggs…





Source: LCLS CDR

On the other hand, spectral brightness is not necessarily the currency of the realm for high field x-ray physics:

XFEL parameter comparisons – here's what counts:

- Tesla TTF: currently has
  - 25 uJ
  - 1×10^{14} W/cm^2=3v/Å
  - 100 fsec
  - 100 nm (10 eV)
- LCLS: will have:
  - 1 mJ
  - >1×10^{18} W/cm^2? = 300v/Å ← intense
  - 200 fsec ← short
  - 1 nm (1keV)

Even XFEL's have their limitations: e.g. SASE

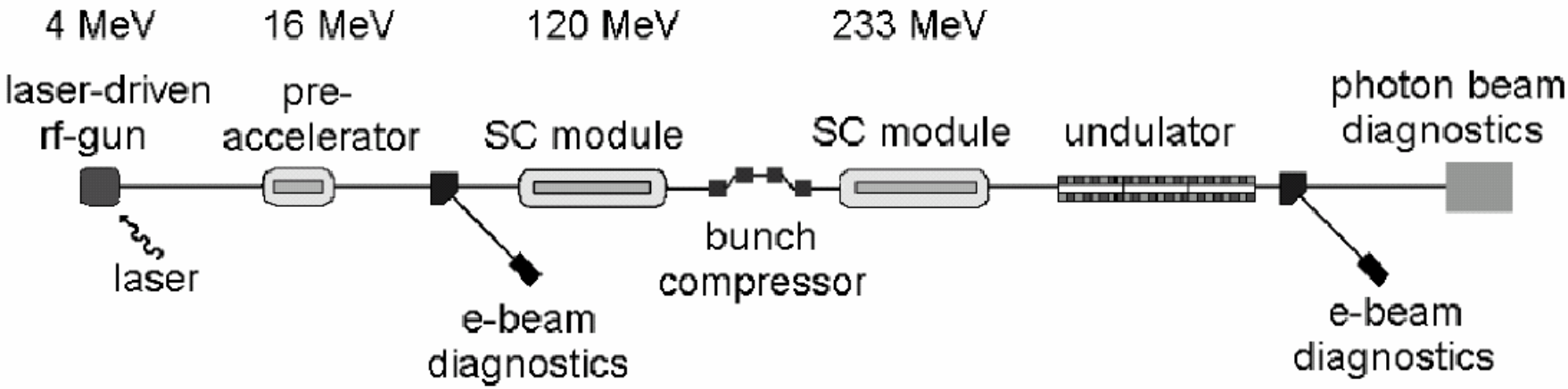
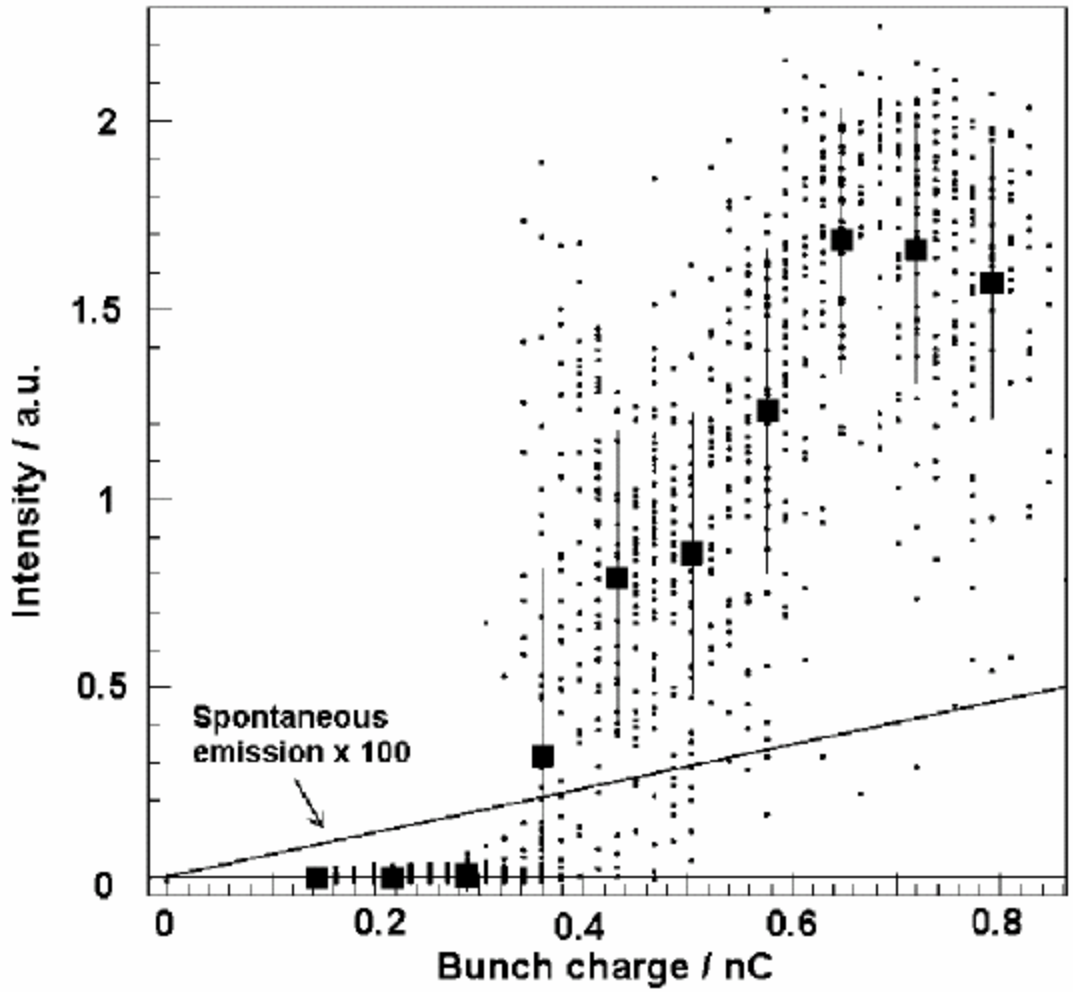


FIG. 1. Schematic layout of phase 1 of the SASE FEL at the TESLA Test Facility at DESY, Hamburg. The linac contains two 12.2 m long cryogenic modules each equipped with eight 9-cell superconducting accelerating cavities [20]. The total length is 100 m.

Source: Andruszkow, et al., Phys. Rev. Lett. 85, 3825 (2000)

The "statistical character" of SASE (i.e. noise is horrible)



Source: Andruszkow, et al., Phys. Rev. Lett. 85, 3825 (2000)

Attosecond pulses, at random times

Source:
LCLS design report

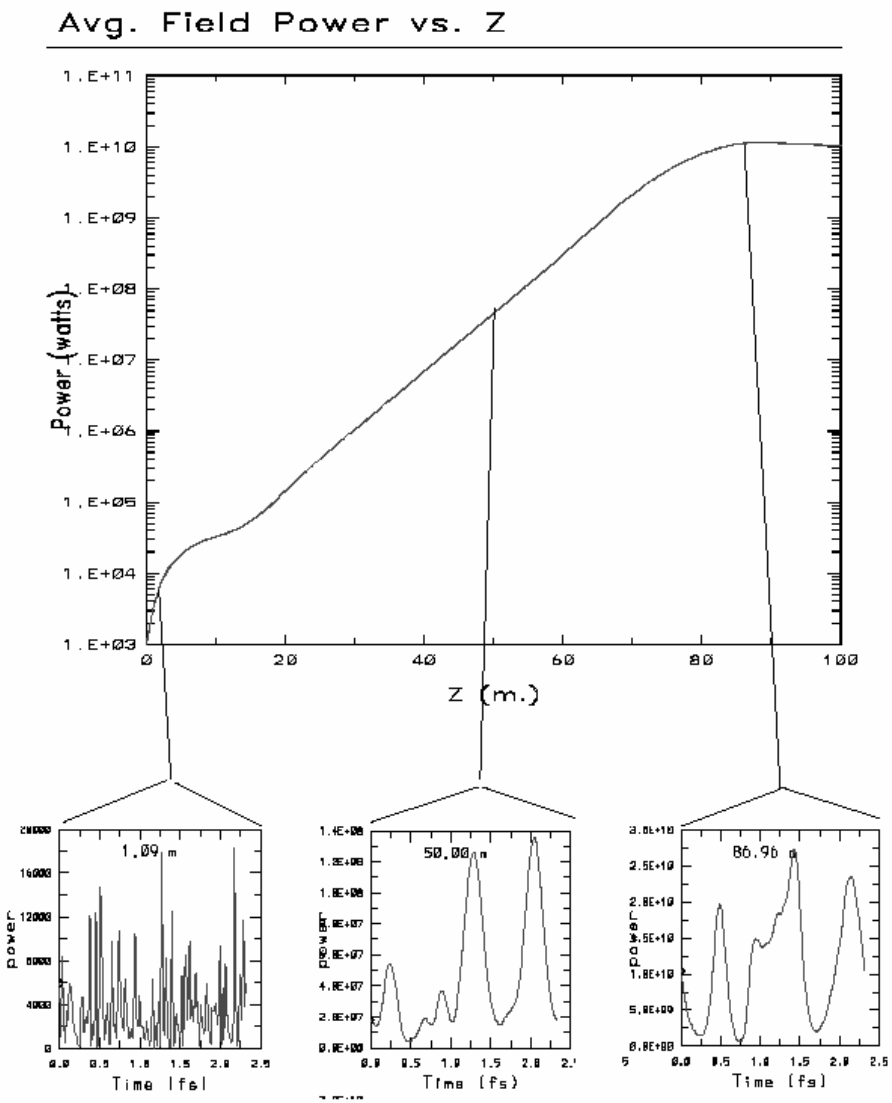


Figure 5.6-1. FEL output power pattern along the bunch.

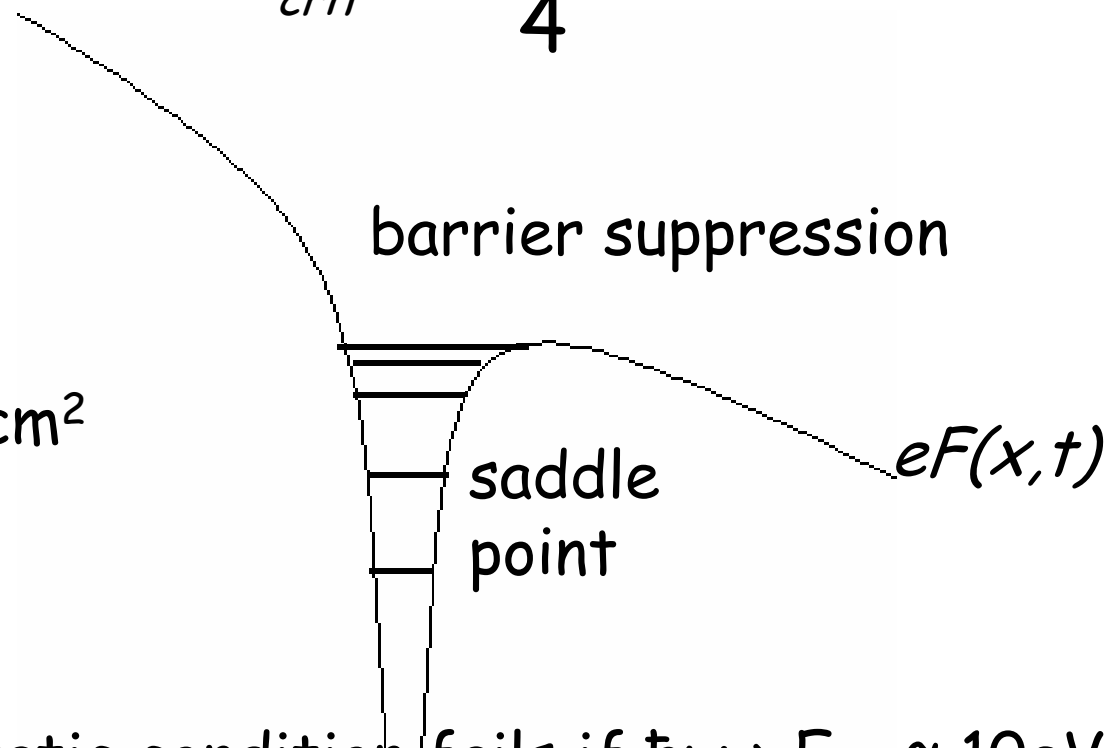
What will happen at high fields

- Lasers are already there, what do they see?
  - Above threshold ionization
  - Non-sequential ionization, barrier suppression
  - High Harmonic radiation, attophysics
  - Relativistic plasma processes
  - Extreme acceleration
  - Antimatter formation
- X-rays won't be like this at all!

A simple rule of thumb for when a SLOW field gets "strong": Field Ionization

$$eF_{crit} = \frac{E_{I.P.}}{4}$$

Strong:
$F_{crit} \geq 3$ v/Å
$I \geq 10^{14}$ W/cm^2



But the static condition fails if $\hbar\omega \geq E_{I.P.} \approx 10$eV;
So x-rays are not good for field ionization!

A different definition of Strong:

Ponderomotive Potential (wiggle energy)

$$U_p = \frac{e^2 F^2 \lambda^2}{8\pi m c^2}$$

$U_p \geq \hbar\omega$ defines the strong field limit.

for $\lambda = 1\mu m\ (\approx 1\text{eV}),\ I_{strong} \geq 10^{13} W/cm^2$;

for $\lambda = 1nm\ (\approx 1\text{keV}),\ I_{strong} \geq 10^{22} W/cm^2$!

X-rays are not a good way to wiggle electrons…

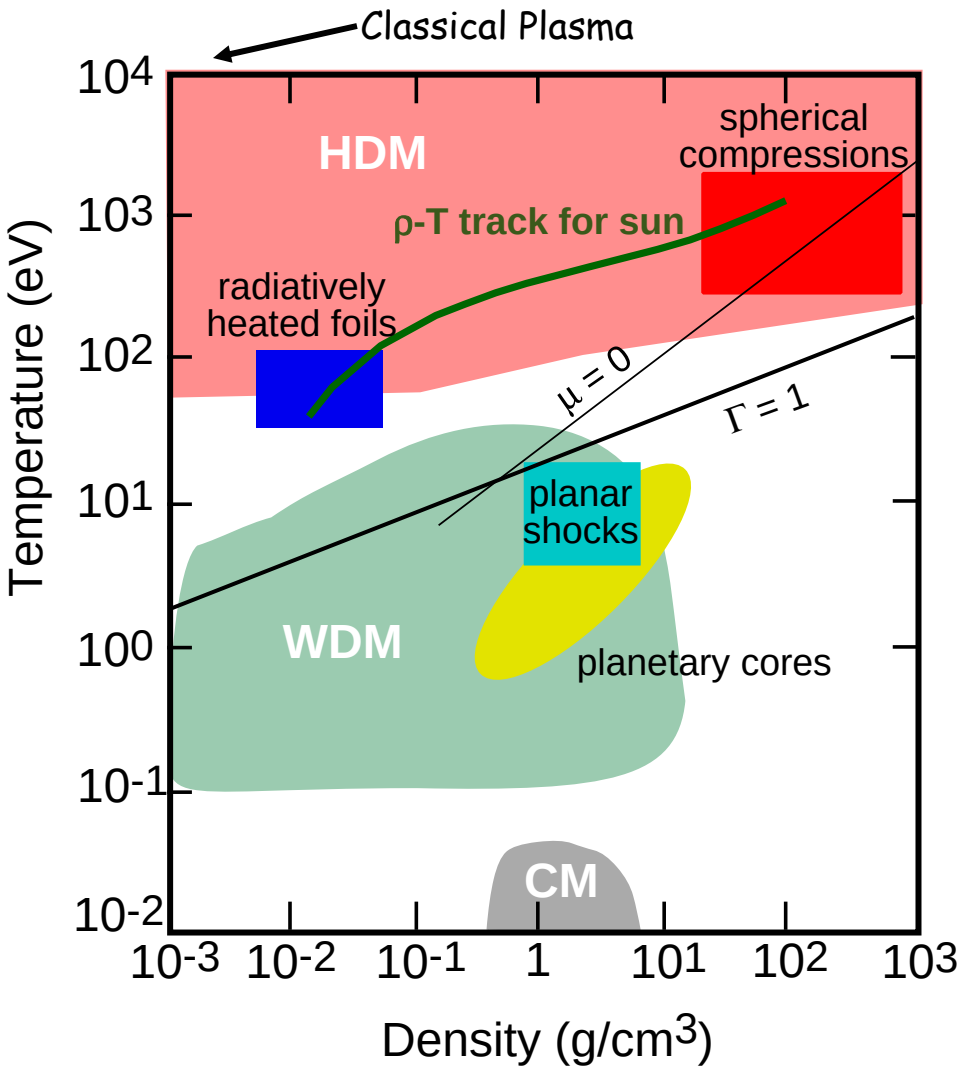
What are XFEL's good for?

"Transparency" when $\omega \geq \sqrt{\dfrac{4\pi N e^2}{m}}$

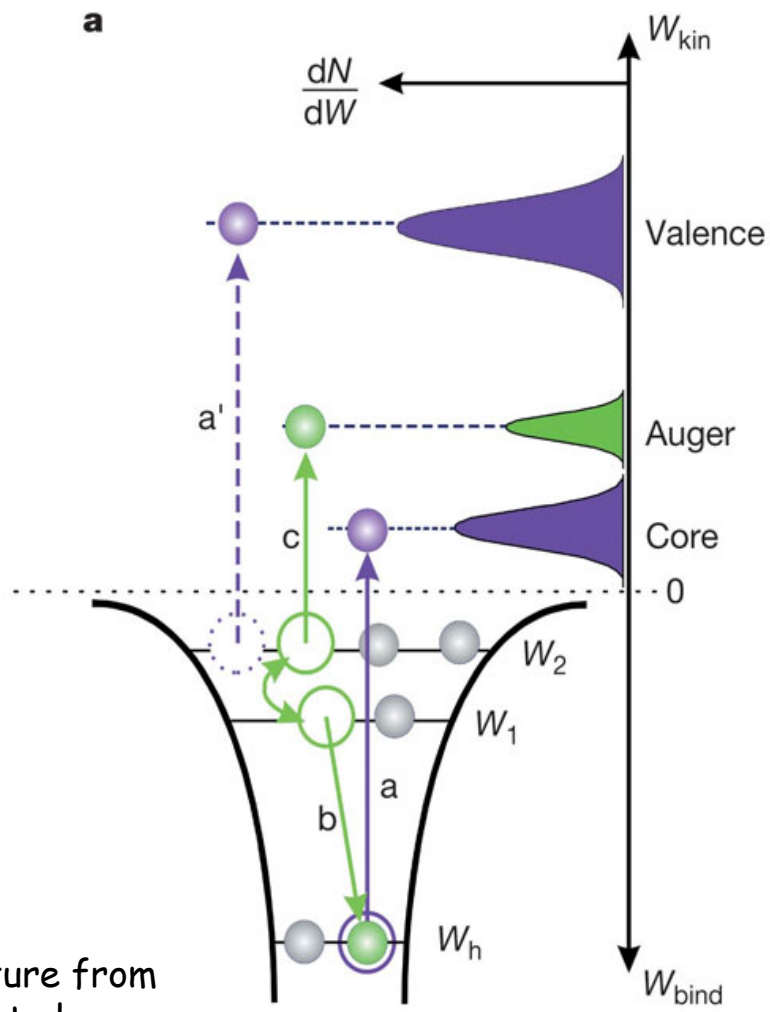
Heating stuff fast

- **Hot Dense Matter (HDM):**
  - Supernova, stellar interiors, accretion disks
  - Plasma devices: laser produced plasmas, Z-pinches
  - Directly driven inertial fusion plasma
- **Warm Dense Matter (WDM):**
  - Cores of large planets
  - Systems that start solid and end as a plasma
  - X-ray driven inertial fusion implosion

Source: Dick Lee, LLNL



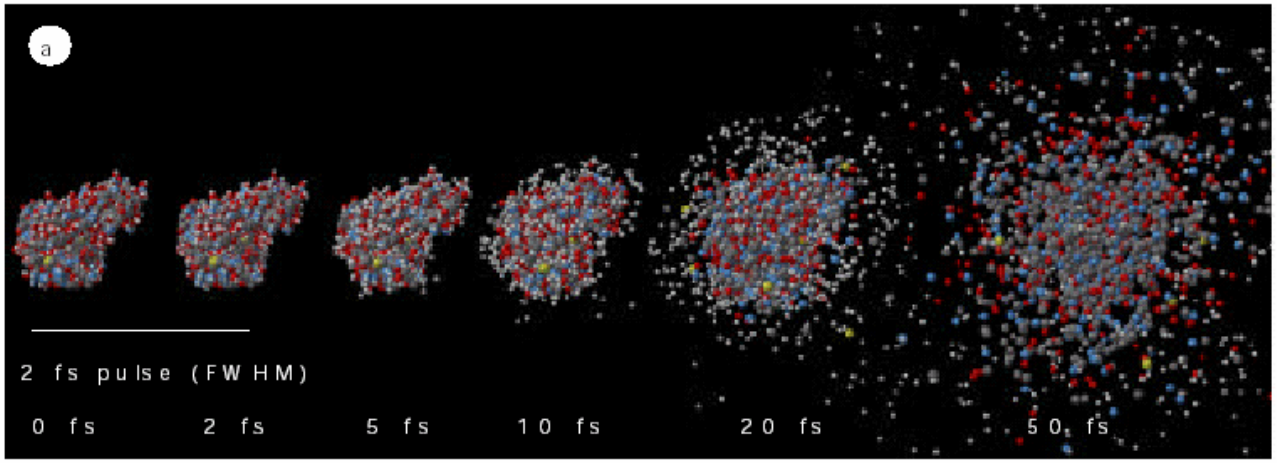
Ionizing atoms inside out: Multiphoton AND Multielectron ("Hollow atoms")

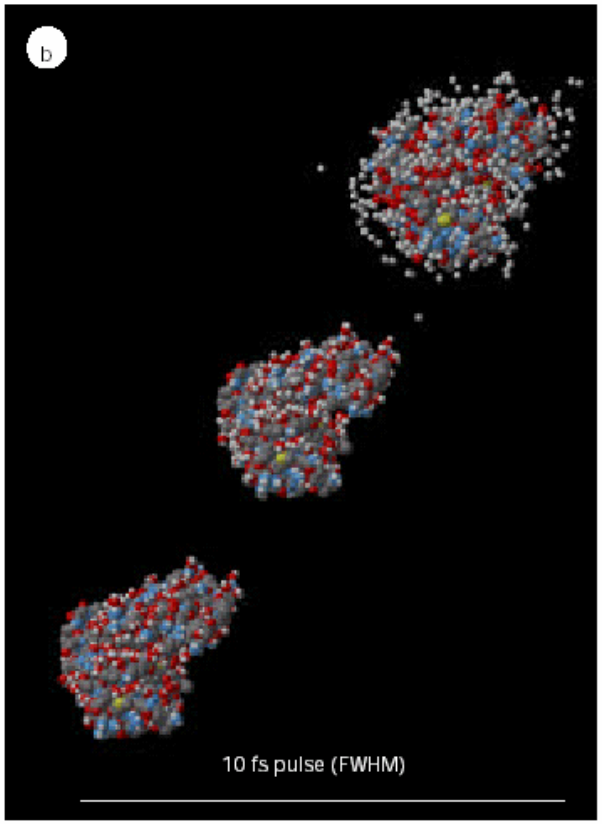


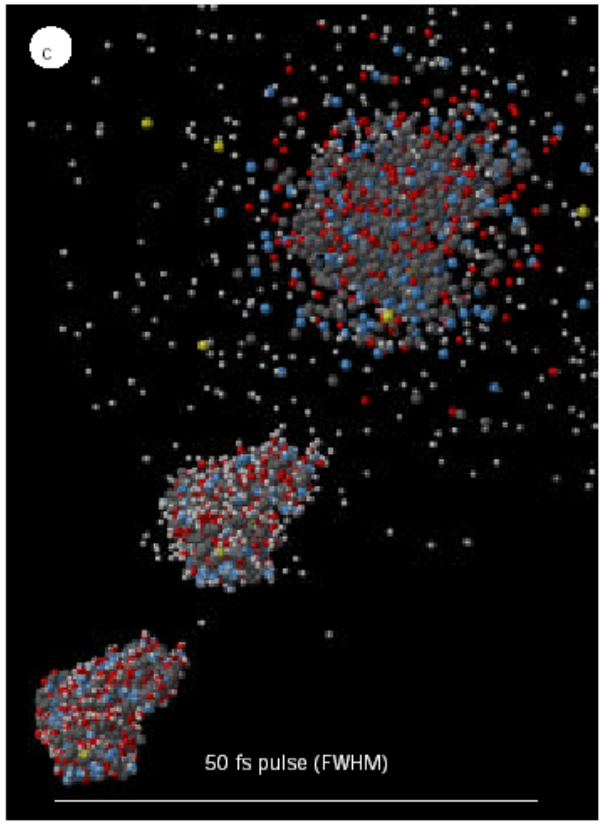
Pretty picture from Drescher et al., Nature **419** (2002)

Single-shot X-ray Flashbulb

LCLS: First Experiments







Ultrafast X-ray Science

- SPPS and TTF are helping to build new expertise in the community
- Lots of new ideas, eg: slicing, parametric processes
- Lots of new challenges as well
- The bar keeps rising: attophysics brings new opportunities to ultrafast x-ray science