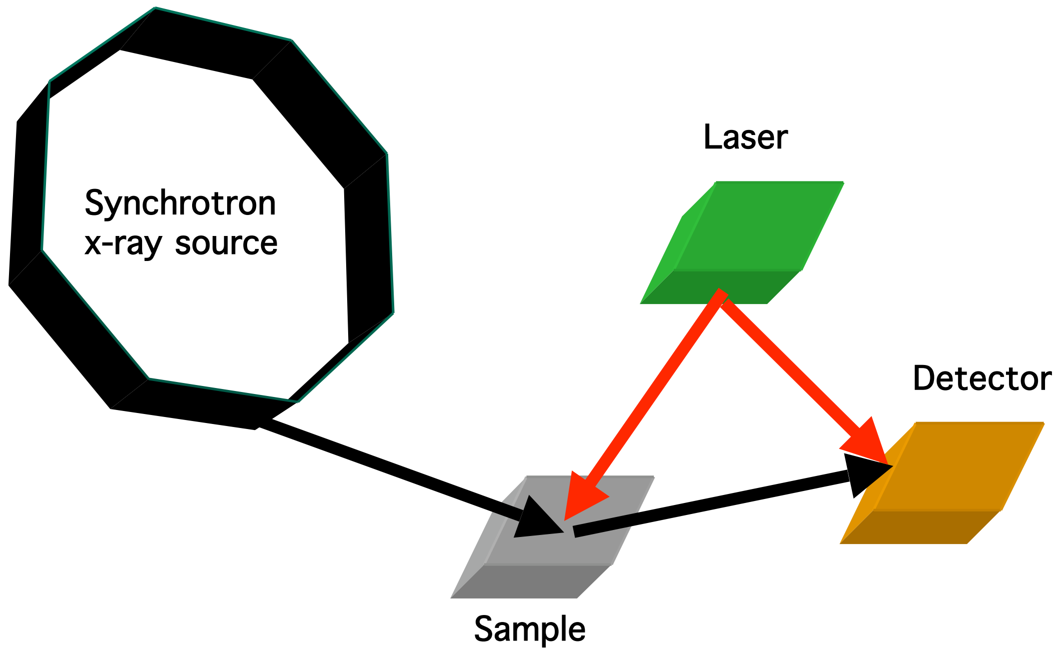
Time-resolved X-Ray Scattering Experiments at the ALS, SPPS, and Beyond

Roger Falcone
UC Berkeley, LBNL and SLAC

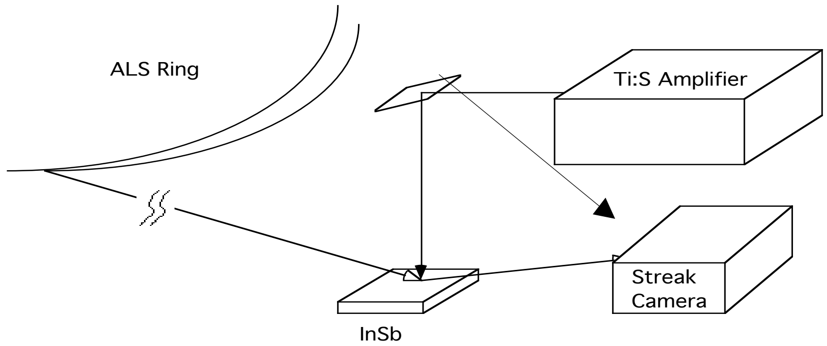
and ALS and SPPS Collaborations

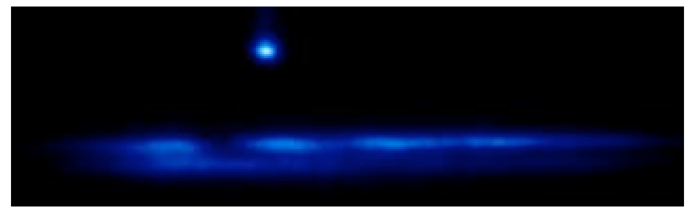
TCSDM Meeting Paris March 2005

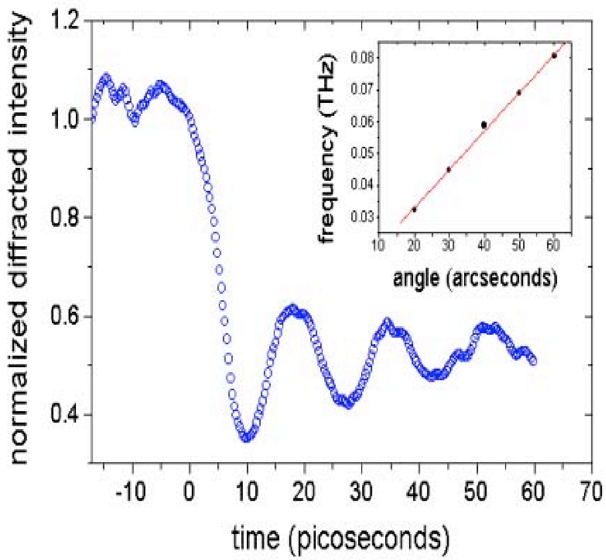
Short-pulse-lasers and synchrotron x-ray sources can create and probe transient states

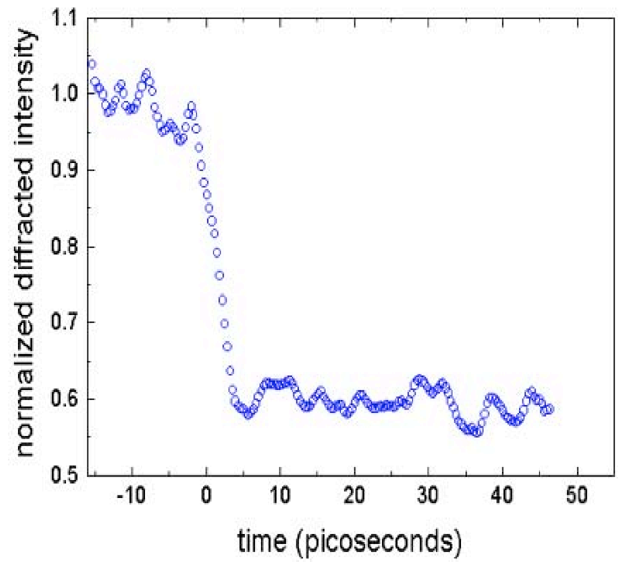


Solid-to-liquid melt transition is initiated by large lattice vibrations









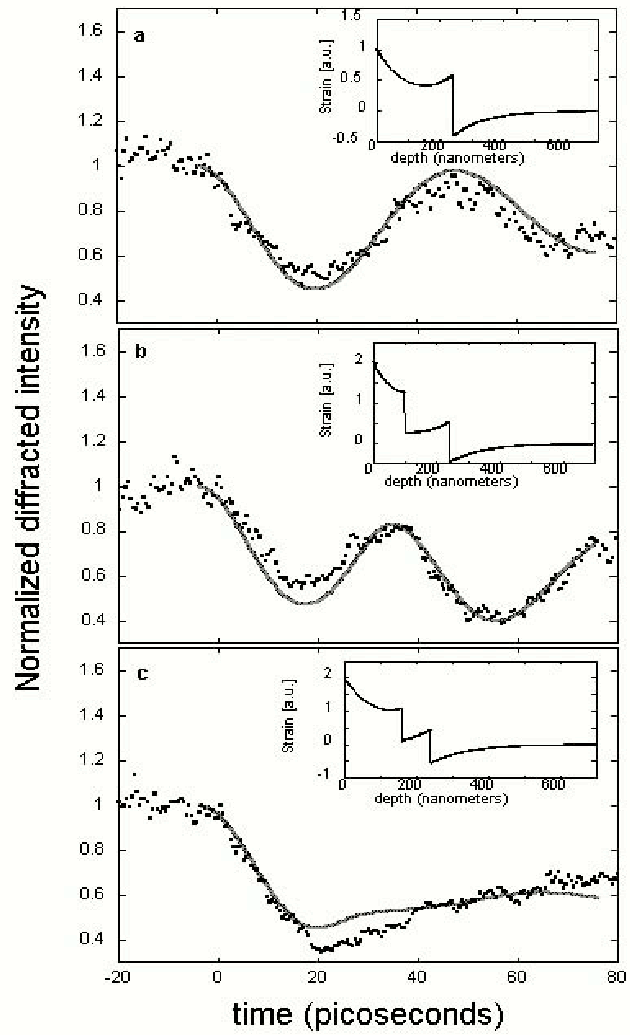
A.M. Lindenberg *et al.*, Phys Rev. Lett. 84, 111 (2000)

Mode selective excitation is observable using multiple pump pulses

Single-pulse excitation

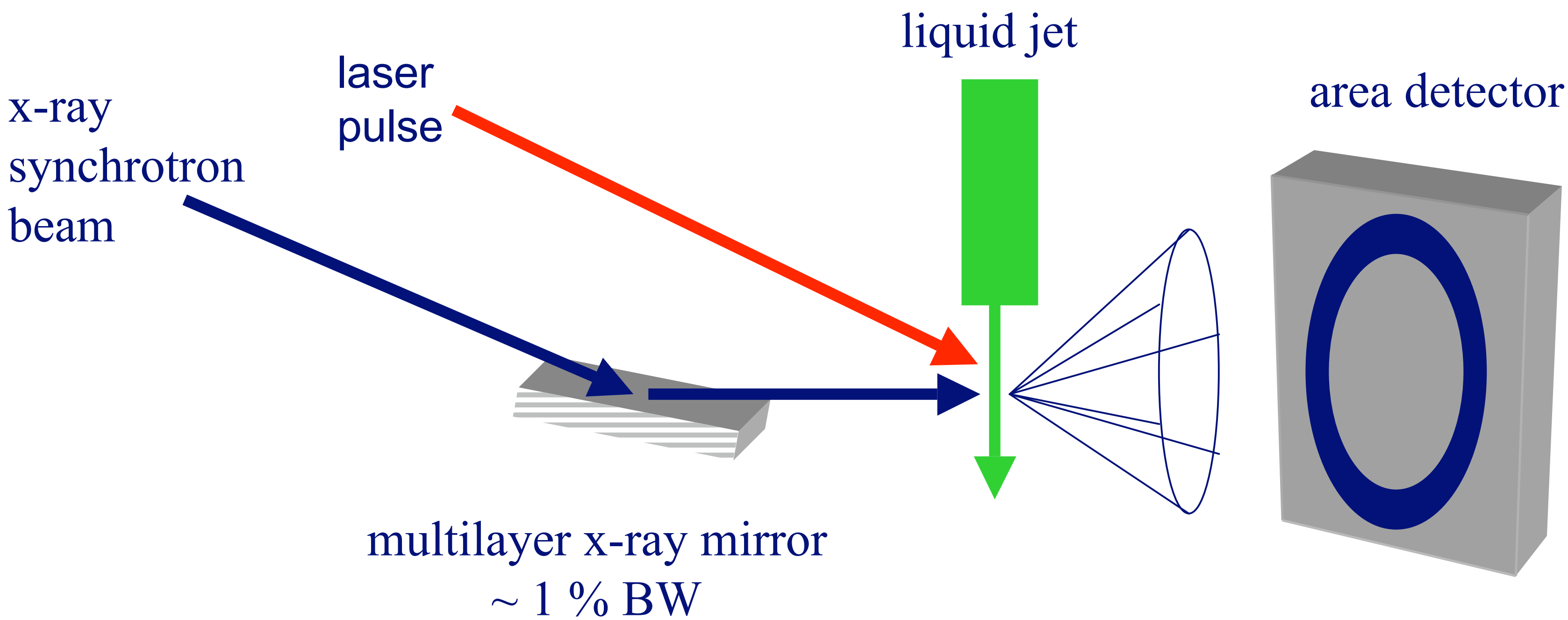
Multiple pulse excitation: constructive interference (Δt~35 ps)

Multiple pulse excitation: mode cancellation mode (Δt~18 ps)



A.M. Lindenberg *et al*. Optics Letters, **27**, 869 (2002)

Perturbed liquid state structure and dynamics can be probed by small angle x-ray scattering

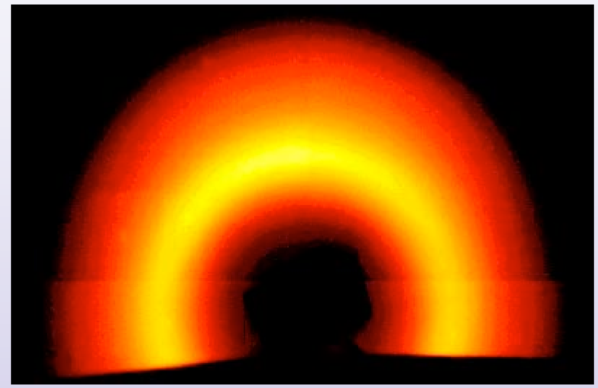


X-ray scattering as a function of angle indicates local structure of perturbed matter

(intramolecular term) (intermolecular term)

$$I(Q) = \sum_{i,j} x_i x_j f_i(Q) f_j(Q) \frac{\sin Q r_{ij}}{Q r_{ij}} + \sum_{i \leq j} x_i x_j f_i(Q) f_j(Q) H_{ij}(Q)$$

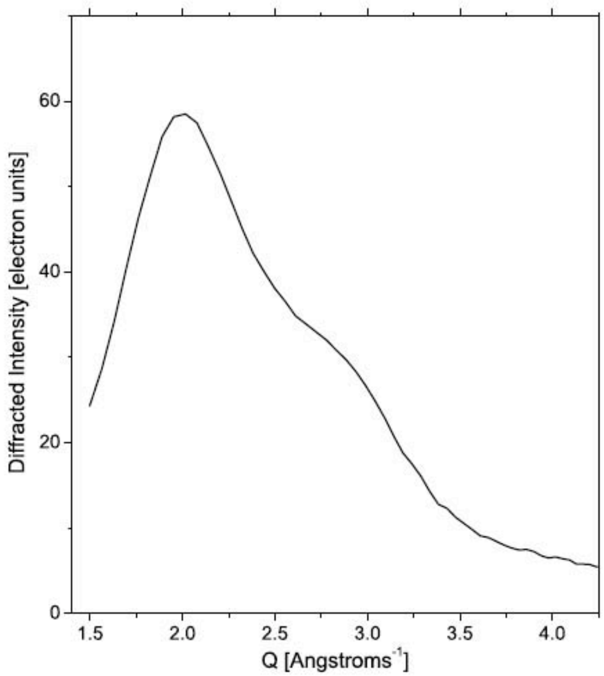
$$H_{ij}(Q) = 4\pi\rho \int_0^\infty r^2 [g_{ij}(r) - 1] \frac{\sin Q r_{ij}}{Q r_{ij}} dr$$



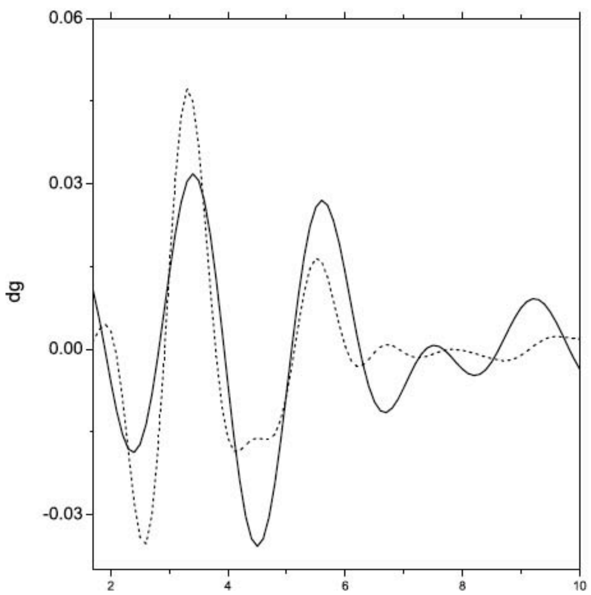
- <u>High Q</u> reflects hard-core region, short length-scales
- <u>Intermediate Q</u> reflects lattice spacing; can be compared with EXAFS pair correlation function
- <u>Low Q</u> reflects long-range, mesoscopic properties

Time-resolved structural changes are seen in H_2O upon charge injection

Static scattering signal



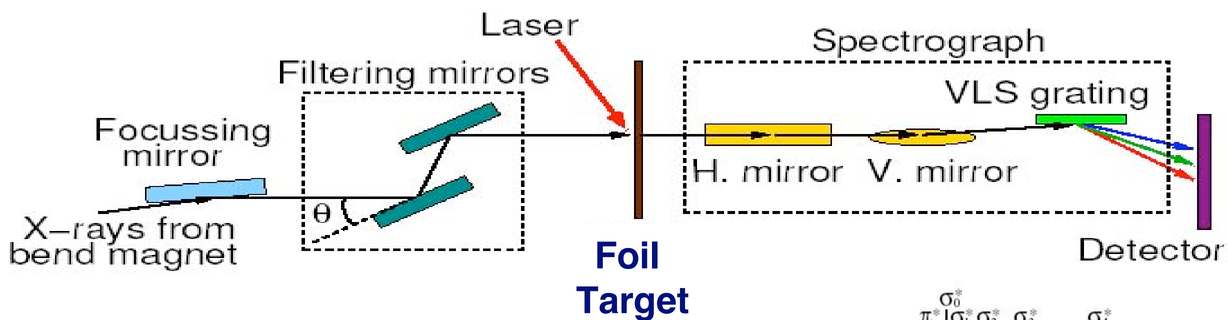
Difference signal PCF at 700 ps, compared with thermal



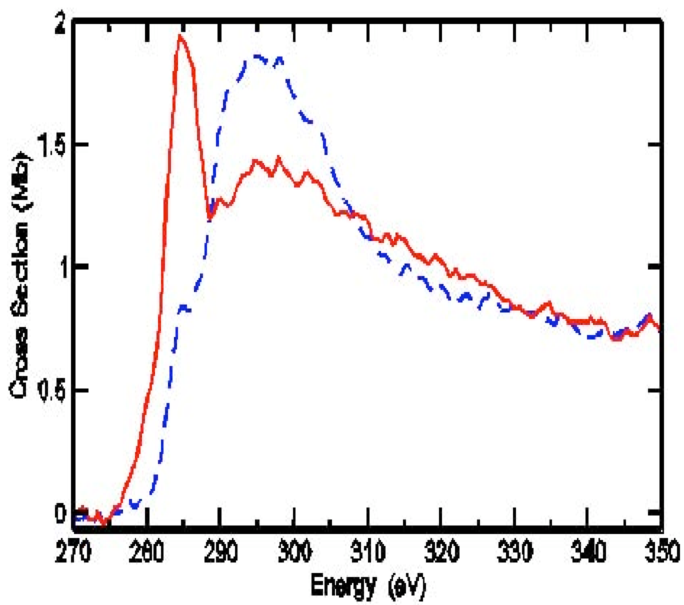
- Implies molecular re-orientation around injected charge with similarities to thermally induced changes

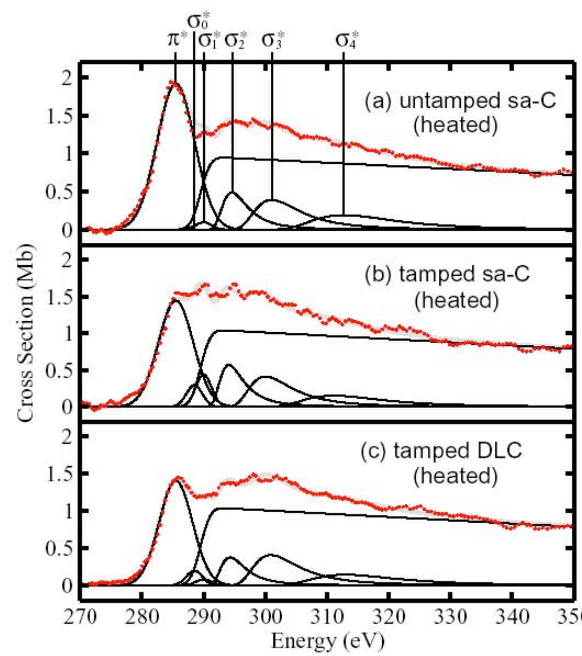
A.M. Lindenberg, *et al*, J. Chem. Phys. (to be published)

X-ray absorption spectroscopy probes bonding in transient states



solid / liquid carbon

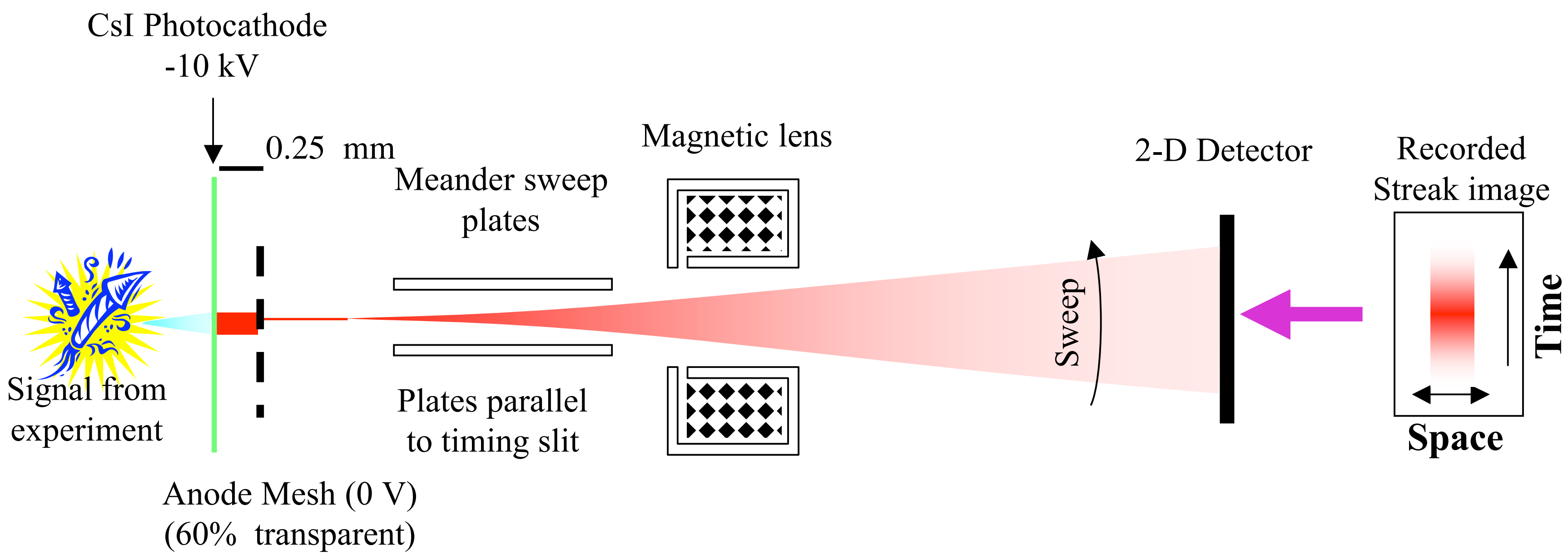




e.g., supports calculations indicating that the low-density phase of liquid carbon is predominately sp-bonded

S.L. Johnson, *et al*
Silicon: PRL **91**, 157403 (2003)
Carbon: PRL **94**, 057407 (2005)

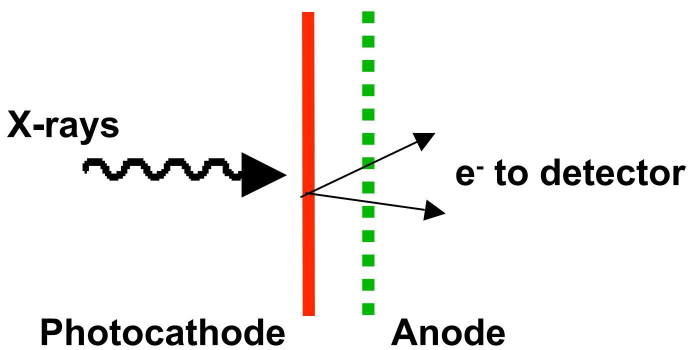
Ultrafast x-ray streak camera detectors enable ultrafast x-ray science

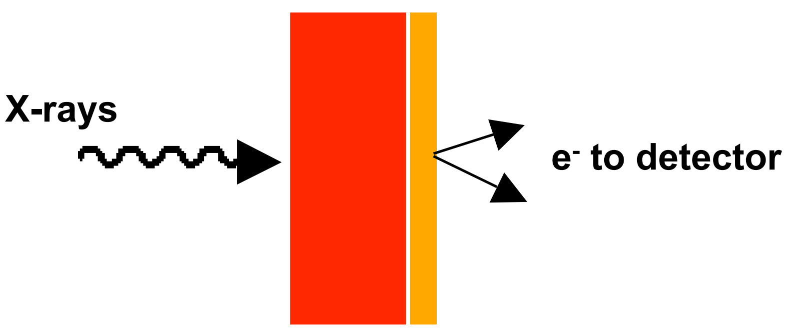


… but detection quantum efficiency is typically low

QE in Normal Incidence vs. Grazing Incidence

Transmission Photocathode

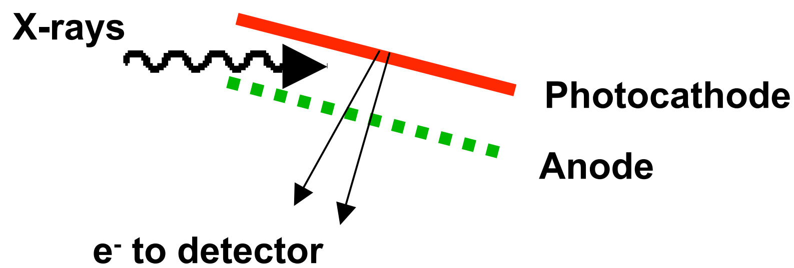


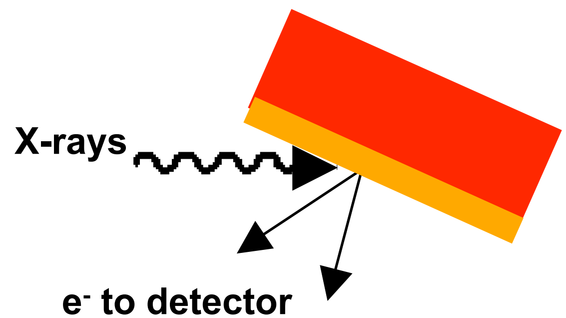


Thickness optimization

QE ~ 0.1 - 10 %

Reflection Photocathode

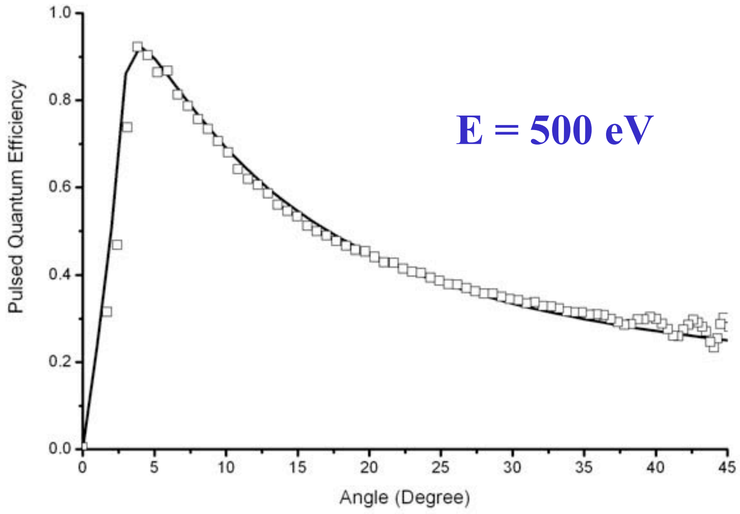


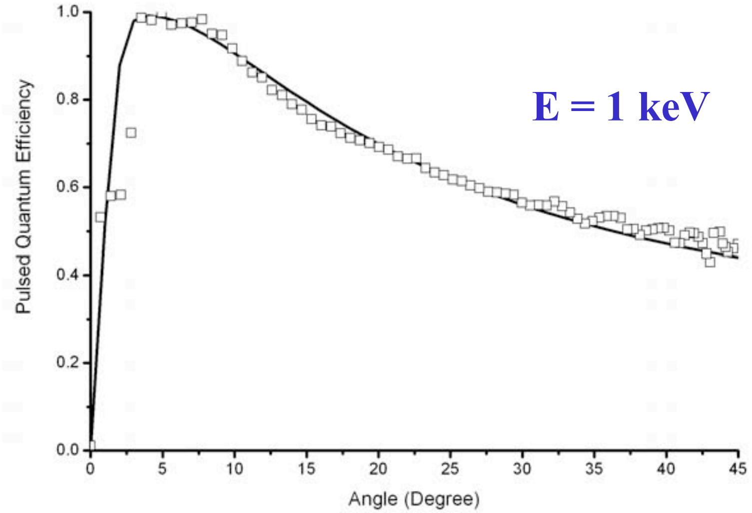


Match X-ray penetration and secondary electron escape depths

QE ~ 100 %

Reflection Photocathode: QE Characterization

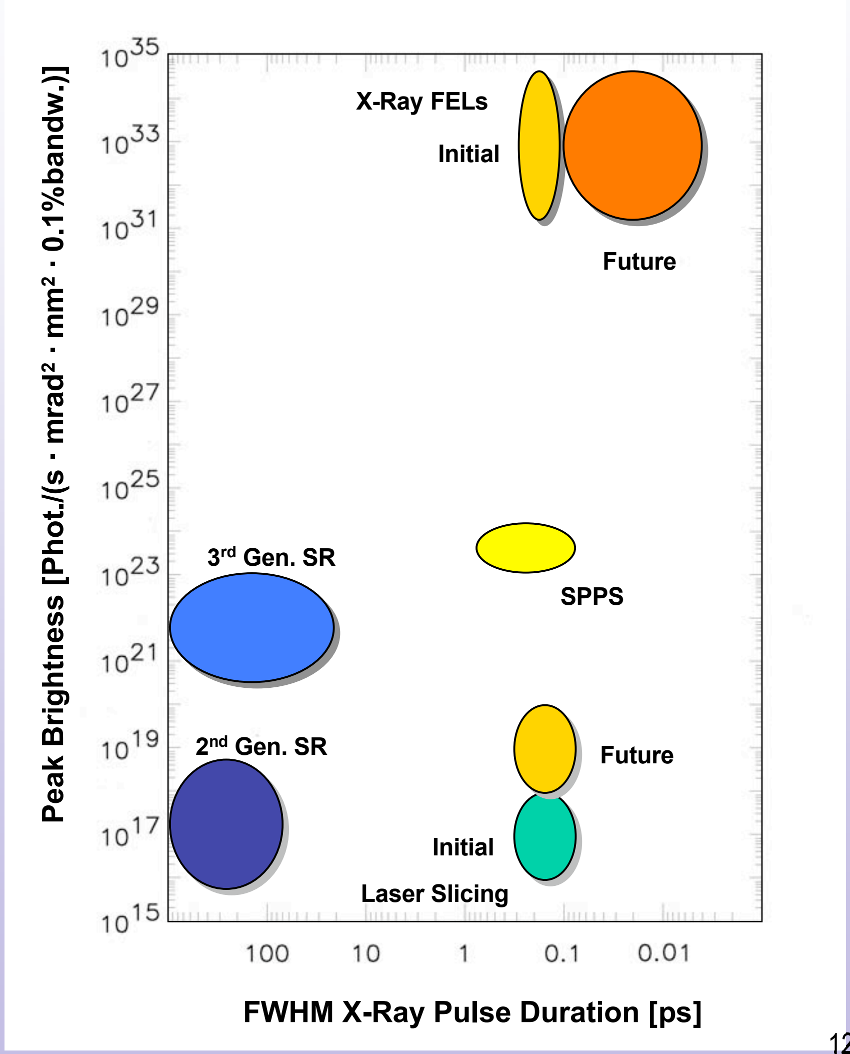




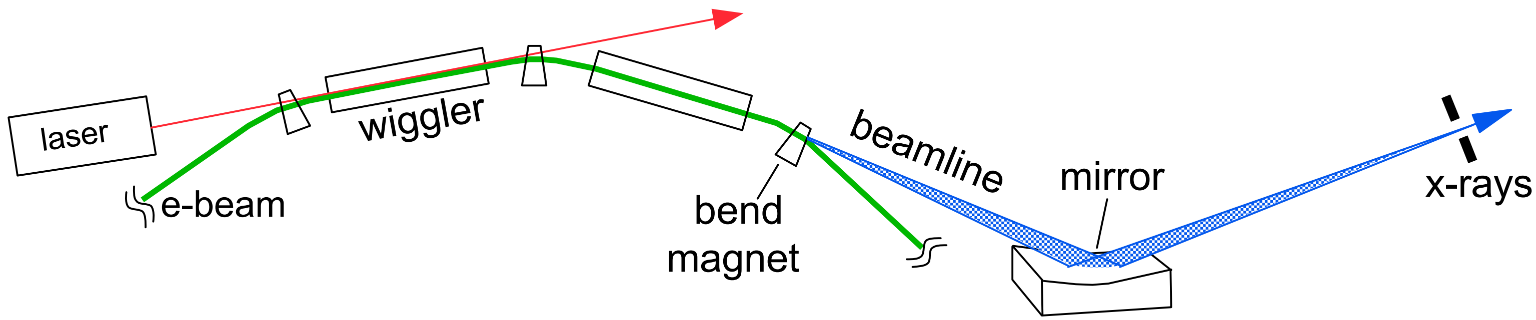
- **Unity Pulsed Quantum Efficiency demonstrated at 1 keV. Near-unity at 500 eV.**
- **Angular dependence of PQE different to TEY:**
  - **Batch escape probability compared to single electron escape probability**
- **No significant field dependence observed.**

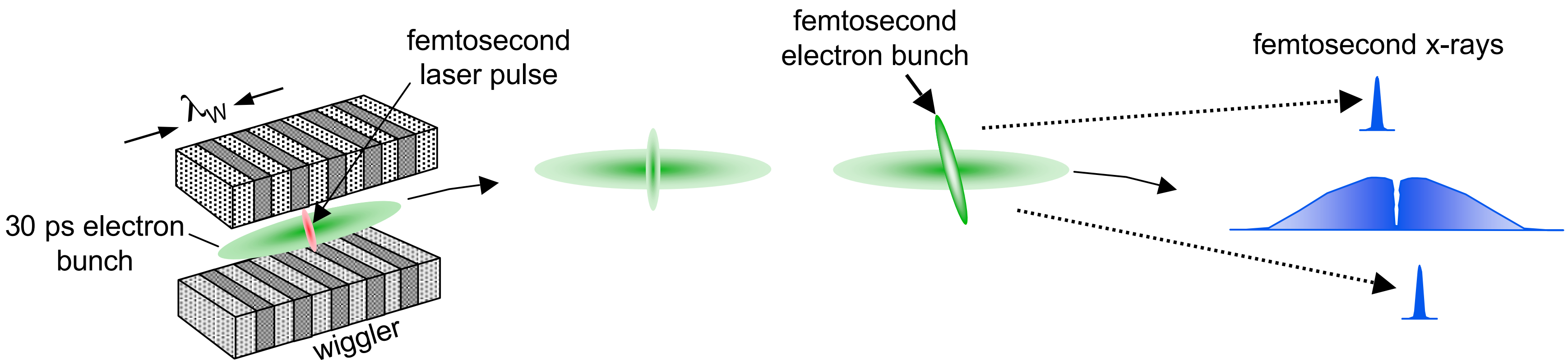
D.P. Lowney, *et al*, Rev. Sci. Inst. **75**, 3131 (2004)

Next-generation x-ray sources will play an important role in HED studies



"Sliced" ultrashort x-ray pulses at ALS Beamline





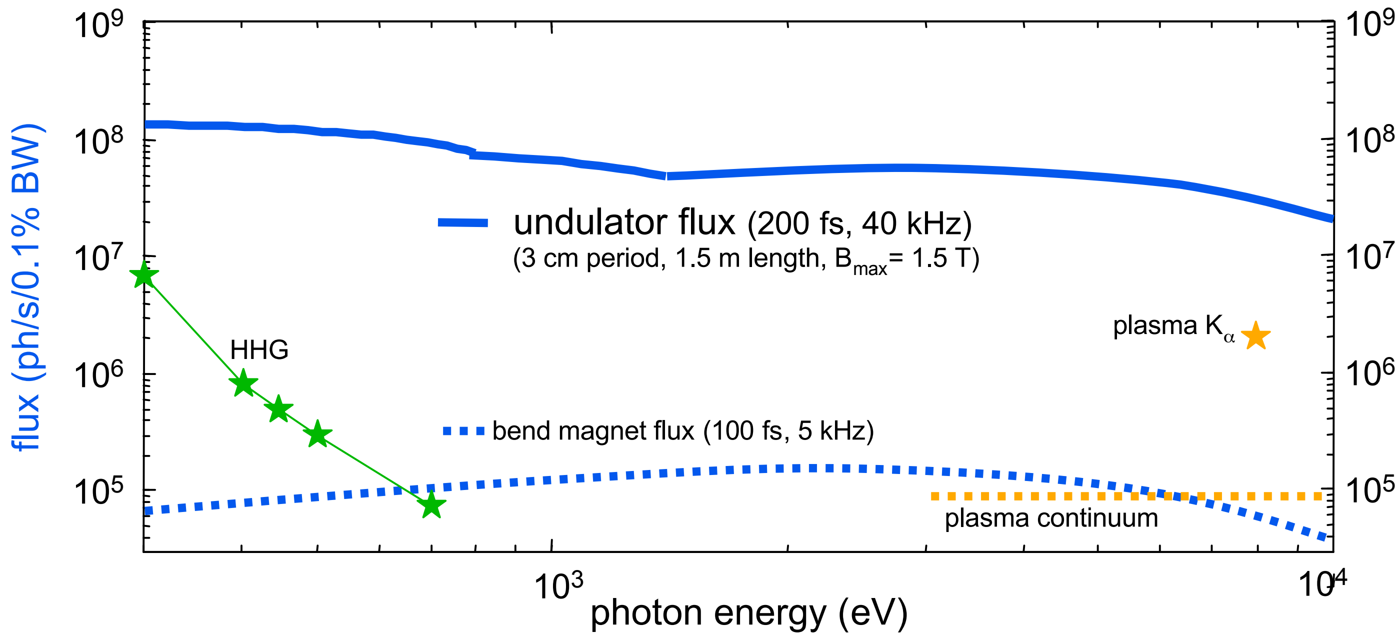
electron-photon interaction in wiggler

spatial separation dispersive bend

bend magnet beamline

Proposed by Zholents and Zolotorev, Phys. Rev. Lett., 76, 916,1996

Comparison of Ultrafast X-Ray Sources



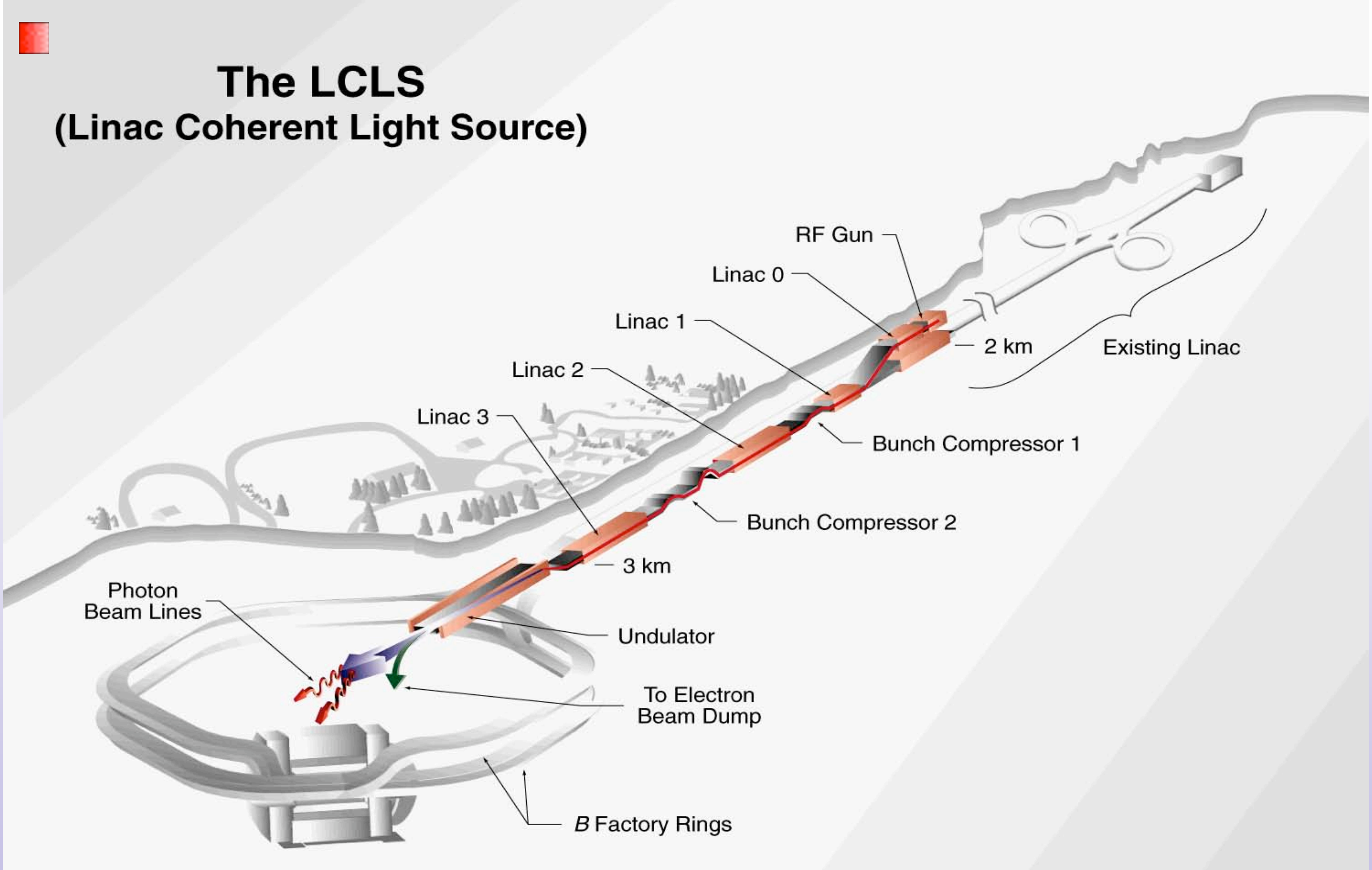
★ HHG flux from F. Krausz, laser: 10 fs, 3 mJ/pulse, 60 W

★ Plasma source flux in mrad^2 from Rose-Petruck, laser: 40 fs, 1 mJ/pulse, 60 W (continuum includes projected 10^5 improvement)

Cu K_α - 10^{10} ph/s/4π (proj. 10^{12} with Hg target)
cont. 6x10^7 ph/s/4π (integ. from 7-8 keV)

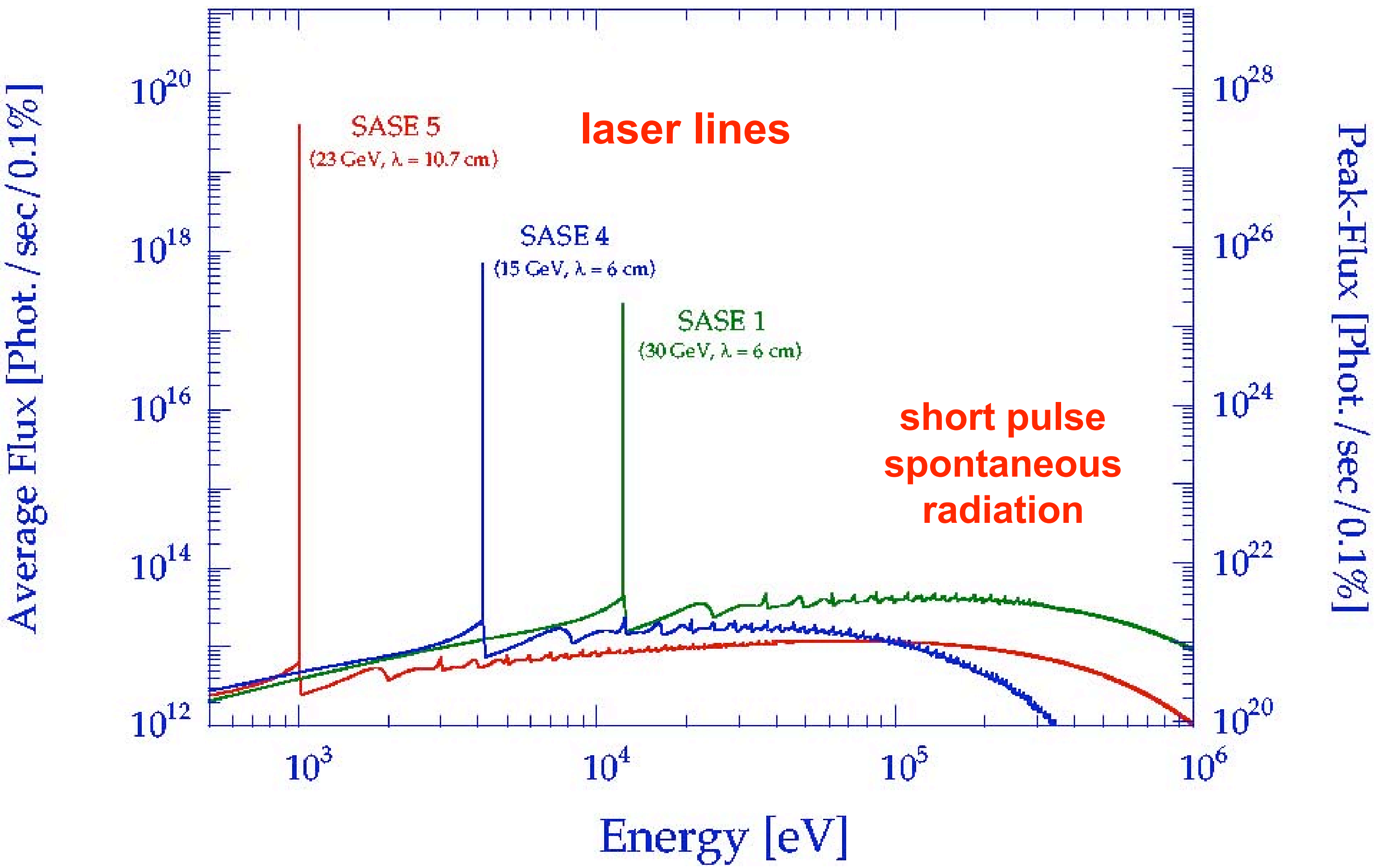
typical average ALS x-ray flux
undulator ~10^{15} ph/s/0.1% BW
bend-magnet ~10^{13} ph/s/0.1% BW

From R.W. Schoenlein LBNL



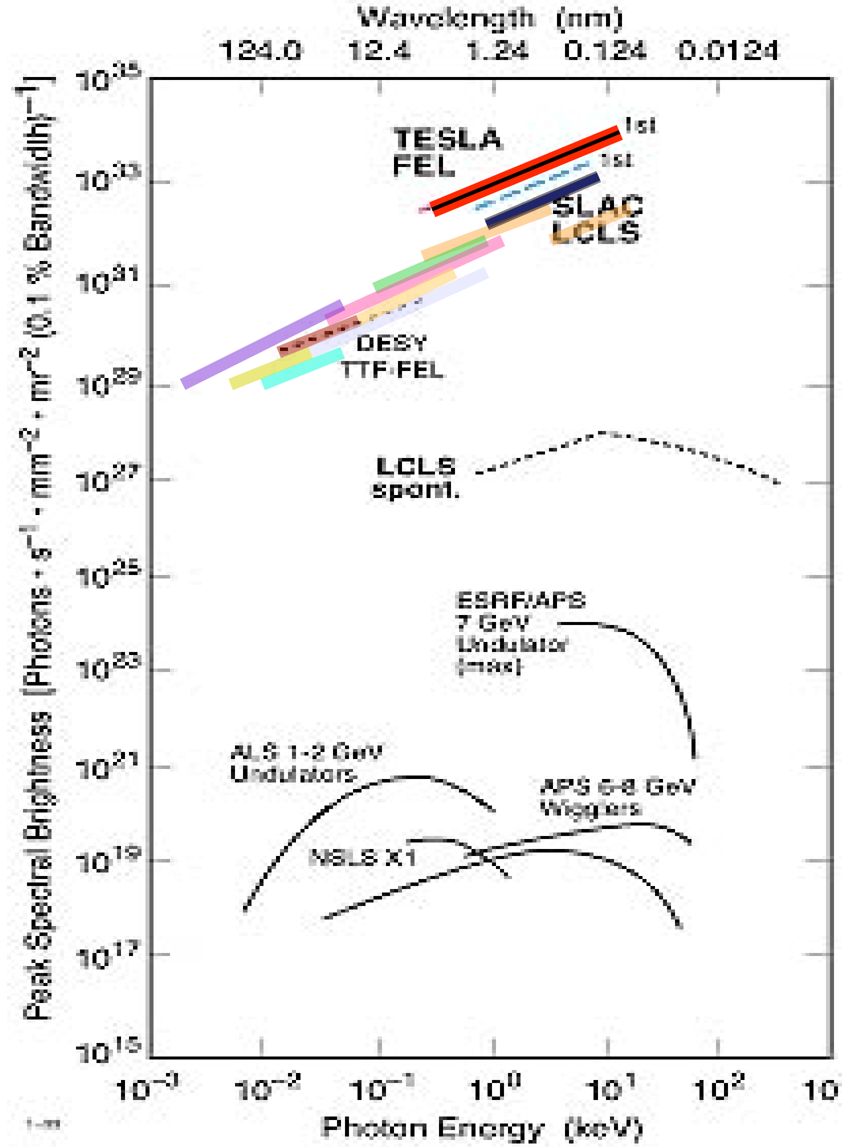
2008-09 Expected commissioning

Spectral properties of SASE FEL

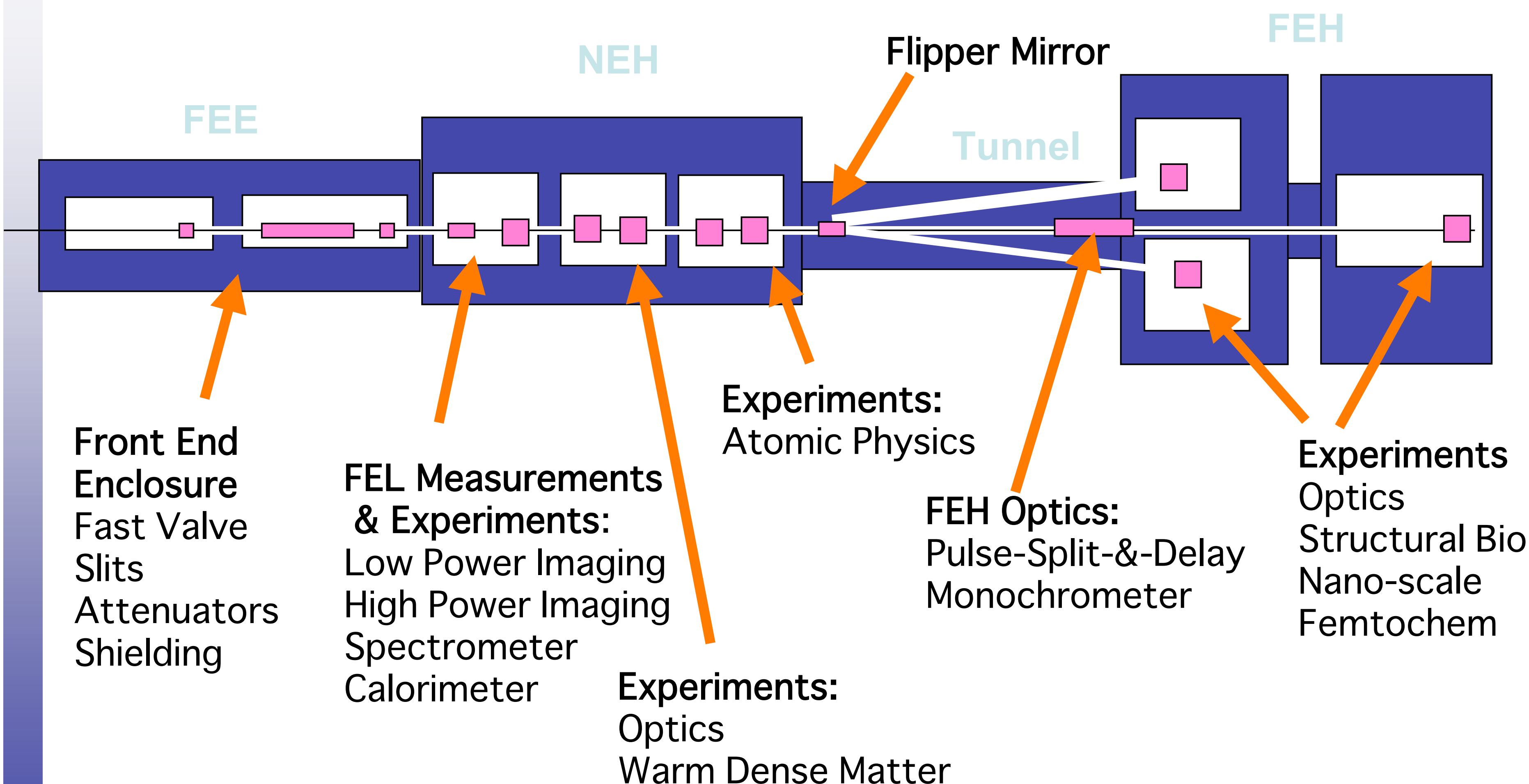


New FEL Sources

- PAL FEL
- SPARX
- SCSS
- FERMI@ELLETRA
- BESSY
- LEUTL
- DUVFEL
- 4GLS
- LUX
- MIT



Possible Layout: X-ray Transport, Optics, and Experiments



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Nanoscale Dynamics in Condensed Matter

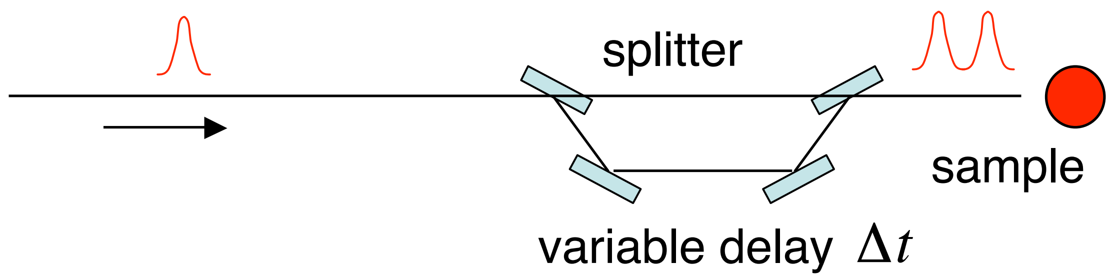
Requirements: **Maximum transverse coherence**

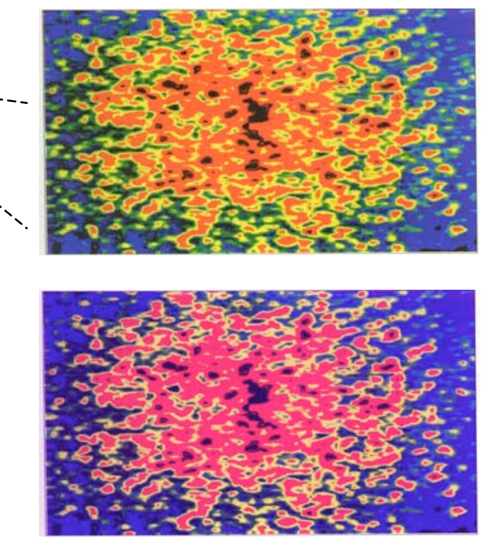
230 fs pulse

<8-24 keV x-rays (3rd harmonic)

Fast Array detectors

In **picoseconds - milliseconds** range

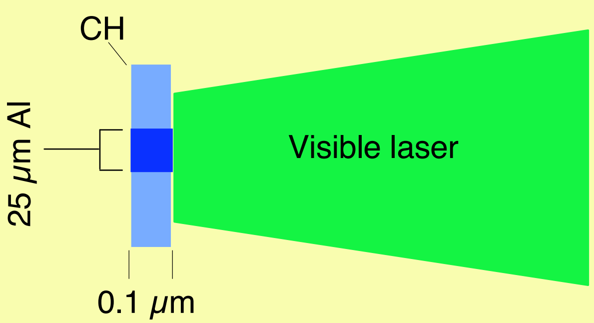




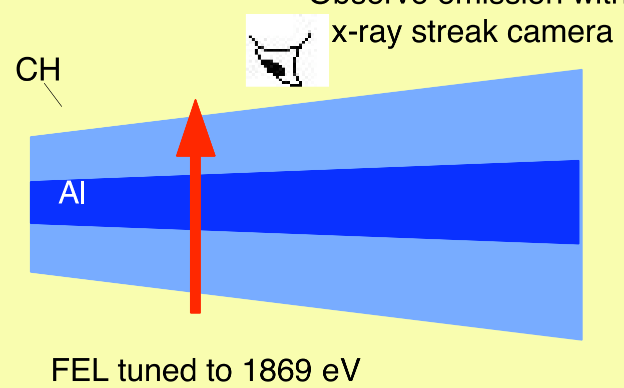
Analyze contrast as function of delay time
Observe time dependence of ordering in materials

X-ray FEL will create excitation levels of high energy density material that are observable in emission

• Schematic experiment

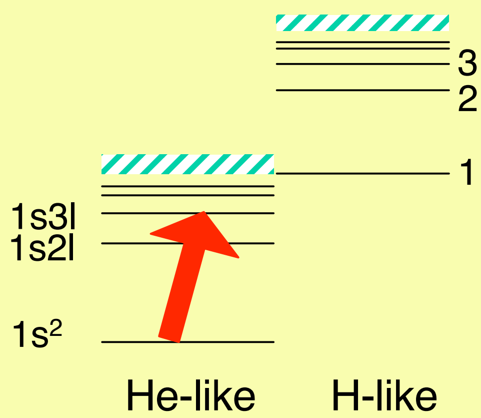


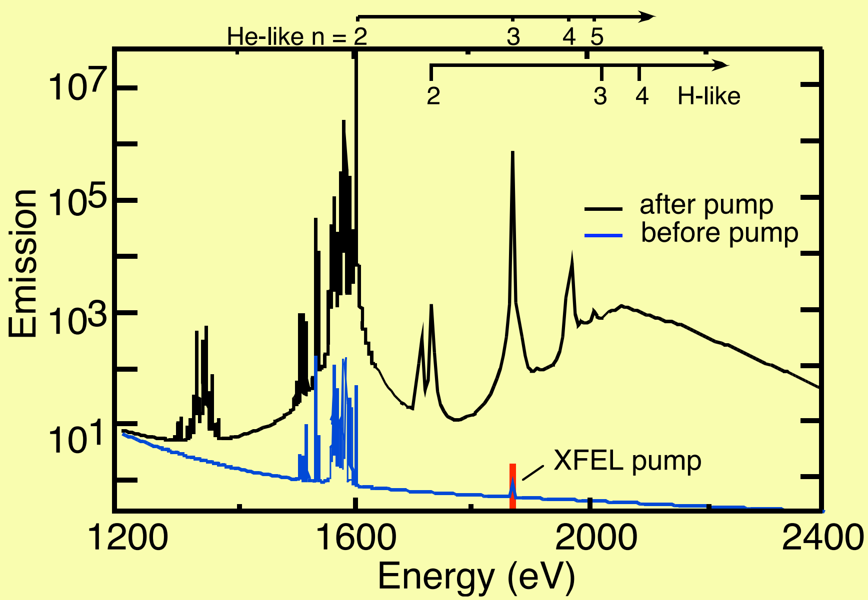
• **t = 0 laser irradiates Al dot**



• **t = 100 ps FEL irradiates plasma**

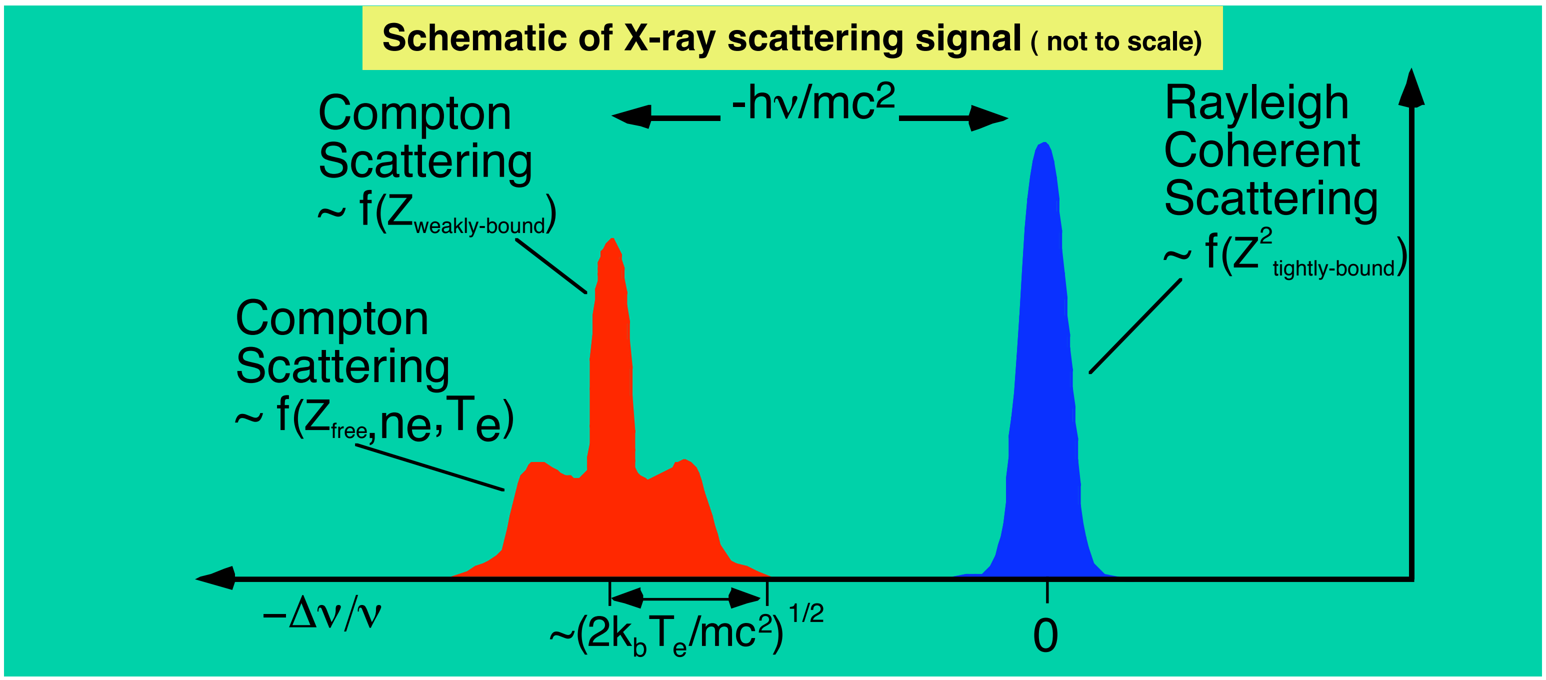
• Simulations





Scattering of the XFEL will provide data on free, tightly, and weakly bound electrons

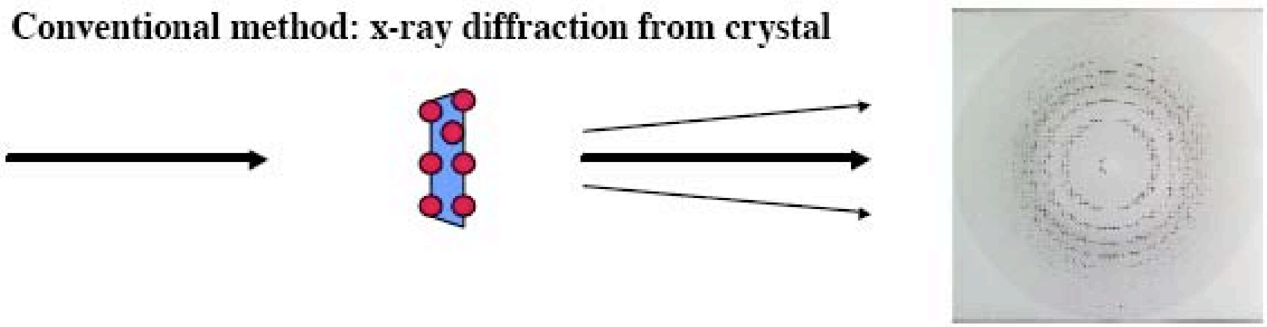
- Weakly-bound and tightly-bound electrons depend on their binding energy relative to the Compton energy shift



- For a 25 eV, 4×10^{23} cm^{-3} plasma the XFEL produces 10^4 photons from the free electron scattering
- Can obtain temperatures, densities, mean ionization, velocity distribution from the scattering signal

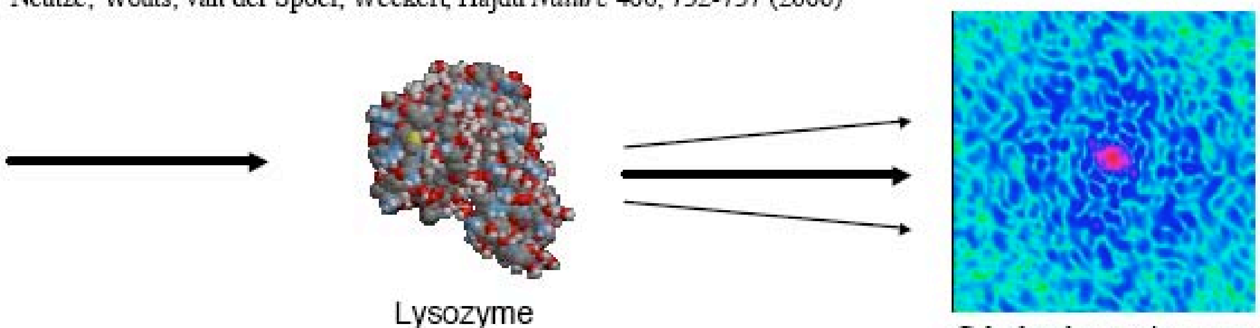
Single molecule imaging has been proposed using short pulse FELs

Conventional method: x-ray diffraction from crystal



Proposed method: diffuse x-ray scattering from single protein molecule

Neutze, Wouts, van der Spoel, Weckert, Hajdu *Nature* 406, 752-757 (2000)



Calculated scattering pattern from lysozyme molecule

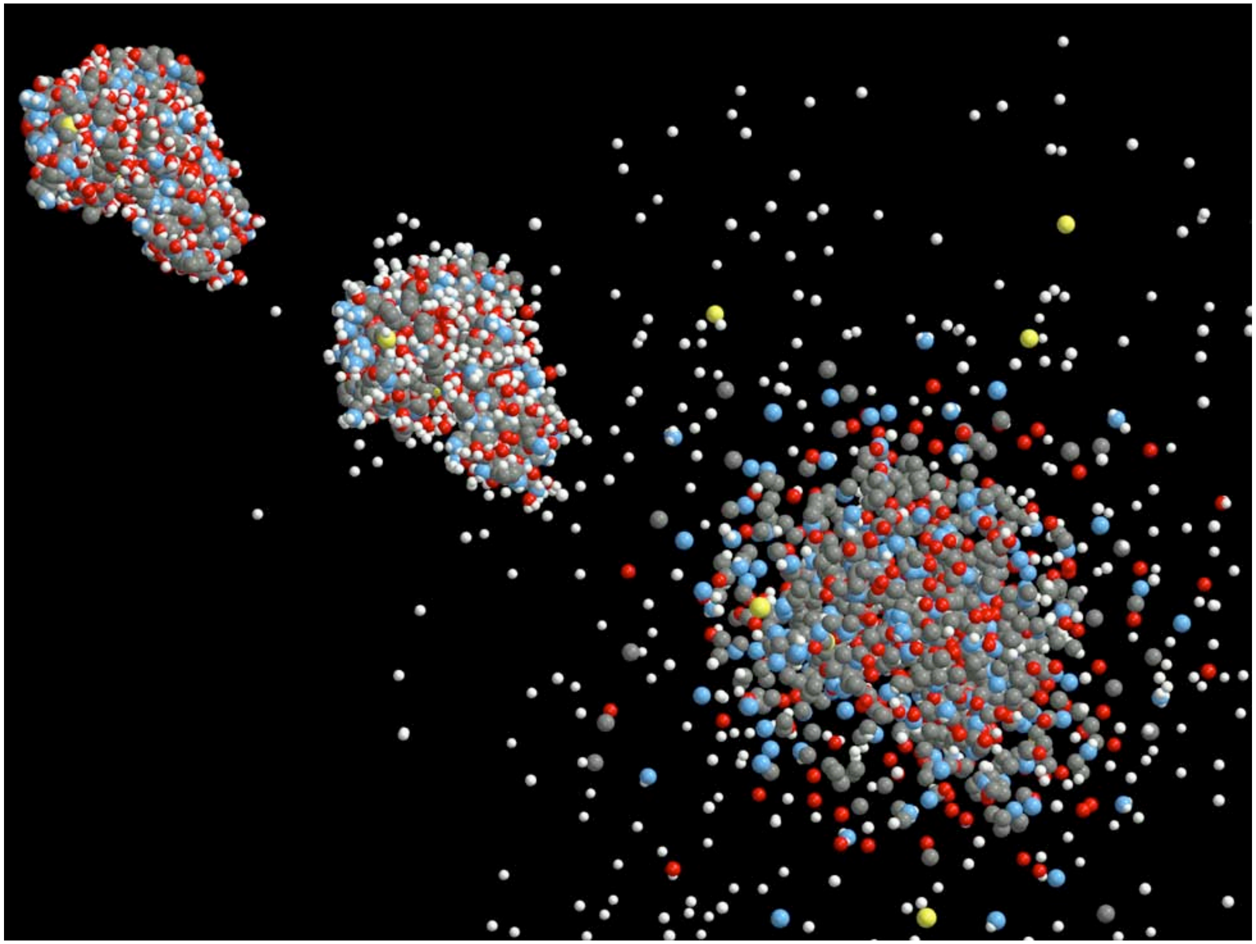
Implementation limited by radiation damage:

In **crystals** limit to damage tolerance is about **200 x-ray photons/Å^2**

For **single protein molecules** need about **10^{10} x-ray photons/Å^2** (for 2Å resolution)

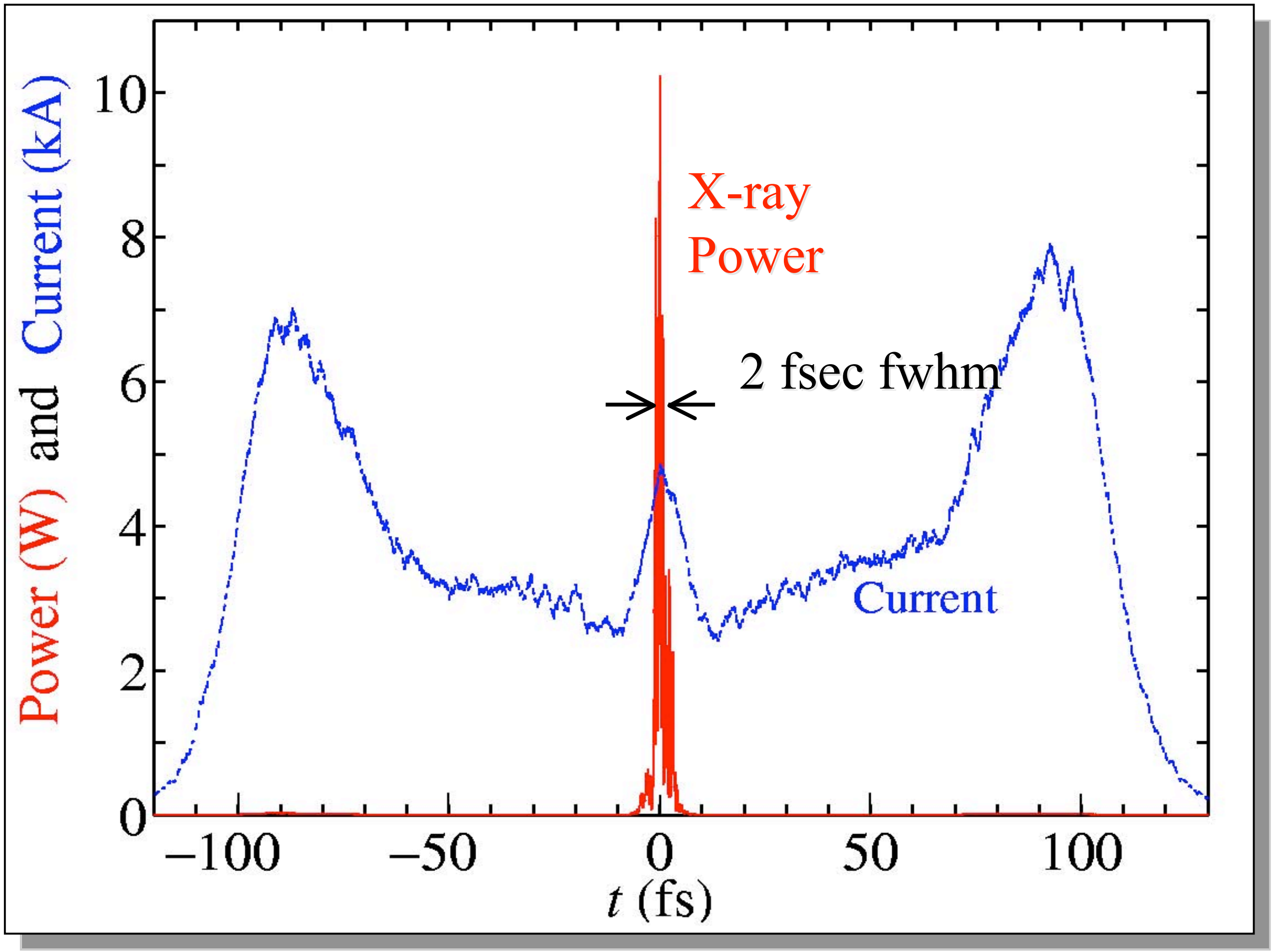
Coulomb explosion modeling (lysozyme)

50 fs
$3x10^{12}$ photons
100 nm spot
12 keV



from Neutze, Wouts, van der Spoel, Weckert, & Hajdu, Nature **406**, 752 (2000)

Emittance spoiling is a possible way to produce ultrashort pulses



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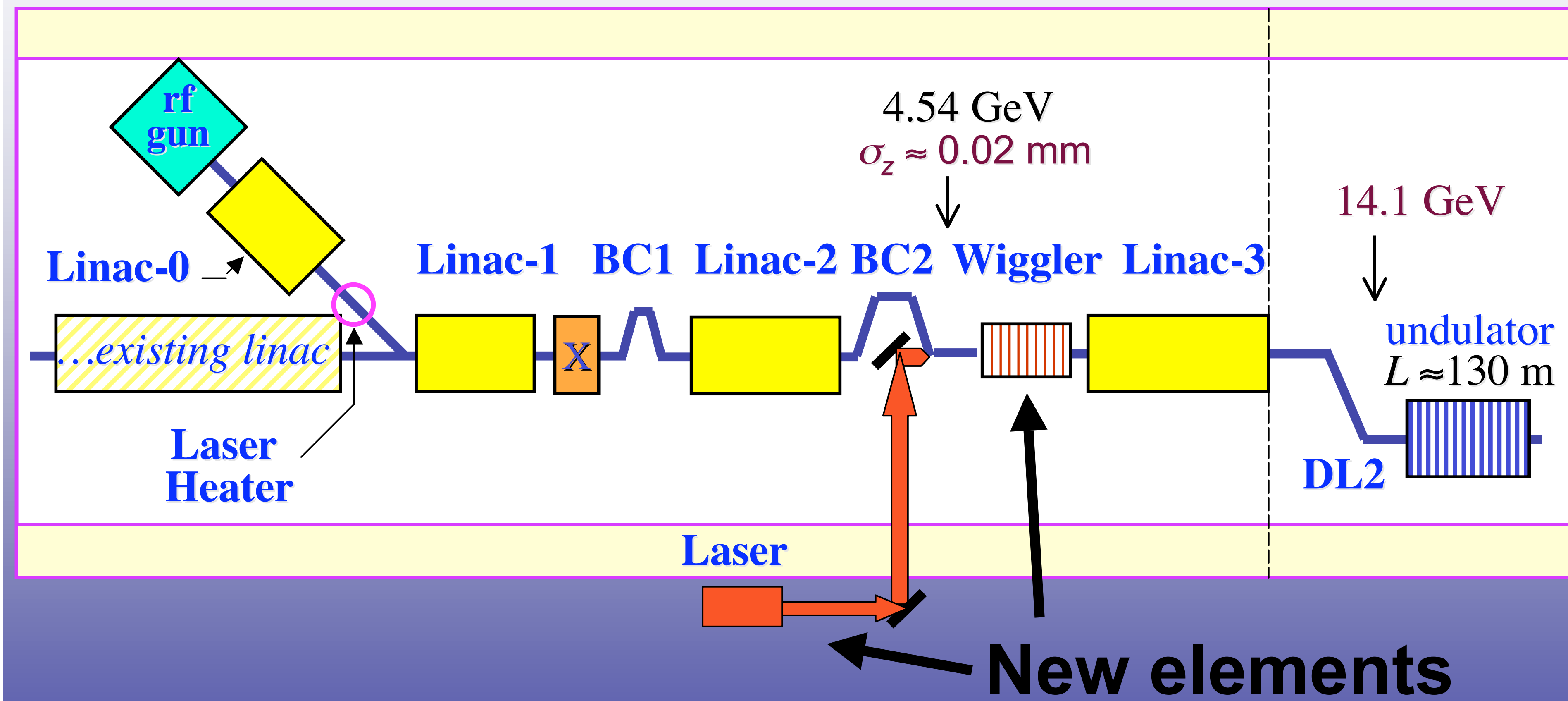
Enhanced SASE scheme may benefit LCLS users



ESASE offers advantages

1) Shorter gain length, high peak power, comparable average power.
2) Easy tunability for a duration of x-ray pulse by laser pulse shaping.
3) Nearly temporally coherent and Fourier transform limited radiation within the spike with random carrier phase between spikes; a solitary attosecond x-ray pulse.
4) Absolute synchronization between laser pulse and x-ray pulse.
5) Relaxed emittance requirement.
6) Shorter x-ray wavelengths.

ESASE Proposal: John Corlett, Sasha Zholents, et al, LBNL CBP

Basedline LCLS Design Parameters

FEL Radiation Wavelength	**1.5**	**15**	**Å**
Electron Beam Energy	14.3	4.5	GeV
Normalized RMS Slice Emittance	1.2	1.2	mm-mrad
Peak Current	3.4	3.4	kA
_ Parameter	4×10^{-4}	8×10^{-4}	
Bunch/Pulse Length (FWHM)	≤230	≤ 230	fs
Relative Slice Energy Spread @ Entrance	<0.01	0.025	%
Saturation Length	87	25	m
FEL Fundamental Saturation Power @ Exit	8	17	GW
FEL Photons per Pulse	1	29	10^{12}
Peak Brightness @ Undulator Exit	0.8	0.06	10^{33} *
Transverse Coherence	Full	Full	
RMS Slice X-Ray Bandwidth	0.06	0.24	%
RMS Projected X-Ray Bandwidth	0.13	0.47	%

*** photons/sec/mm^2/mrad^2/ 0.1%-BW**

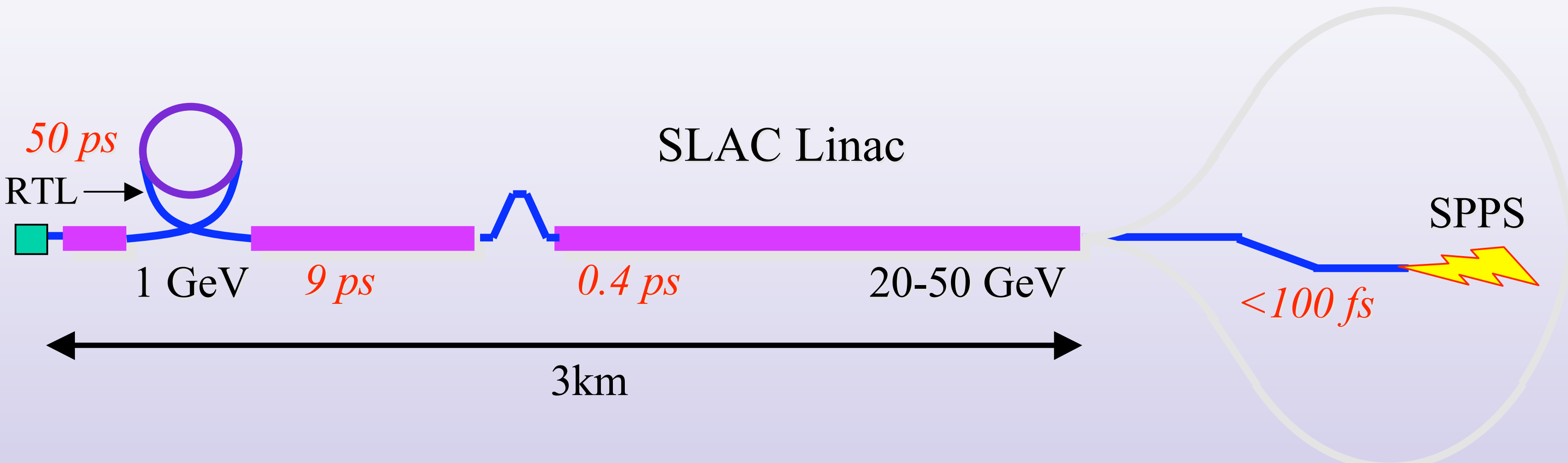
Storage Ring *vs.* Linac *vs.* Recirculating Linac X-Ray Sources

- Storage rings provide ~ 100-ps duration pulses
  - of spontaneous x-ray radiation
  - with high average brightness at high repetition rate
  - and can be "sliced" to provide ultrashort pulses at moderate repetition rate

- Linacs provide ultrashort pulses
  - of soft and hard x-ray FEL radiation
  - with high peak brightness
  - at low repetition rate

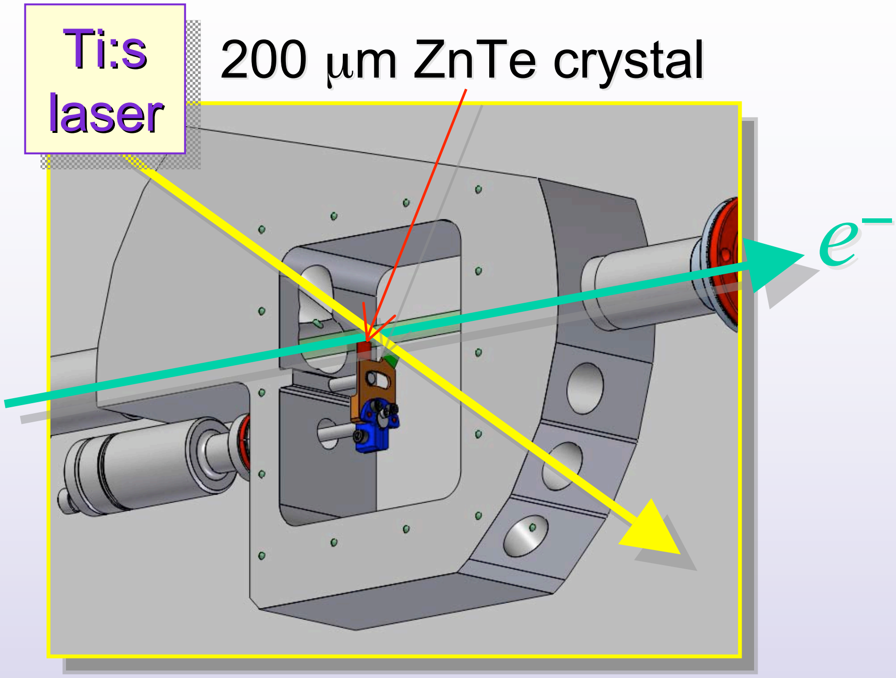
- Recirculating Linacs provide ultrashort pulses
  - of soft x-ray FEL or HGHG radiation, and hard x-ray spontaneous radiation
  - at moderate repetition rate

The Sub-Picosecond Pulsed Source (SPPS) is an R&D facility for the LCLS FEL

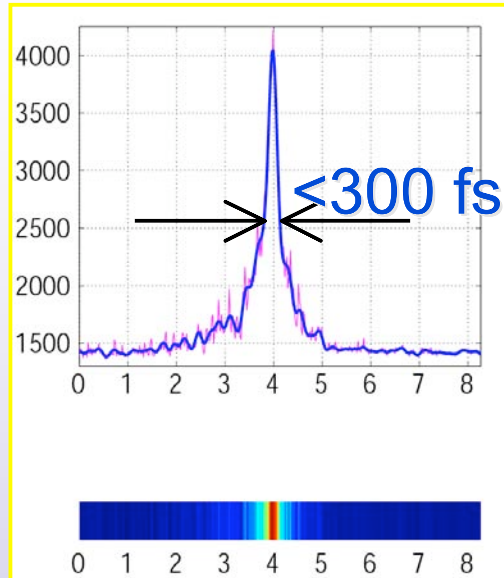


- **Electron bunches generated now at SPPS are 80 fs in duration, comparable to the bunches that will drive future x-ray FELs such as LCLS**
- **A 2m undulator delivers 80 fs duration hard x-ray pulses**

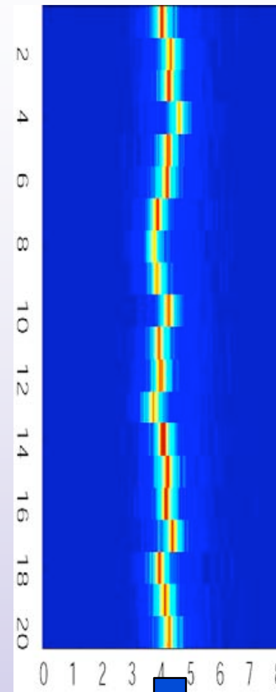
Electro-Optical Sampling for timing at the SPPS

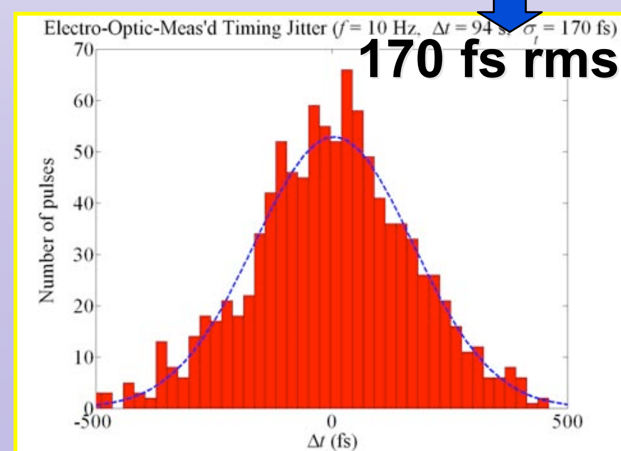


Single-Shot



Timing Jitter



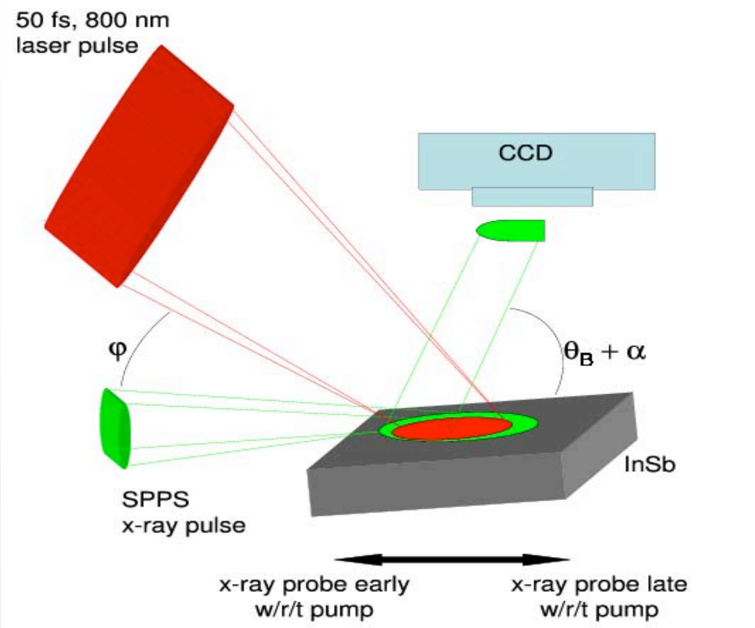


e^- temporal information is encoded on transverse profile of laser beam

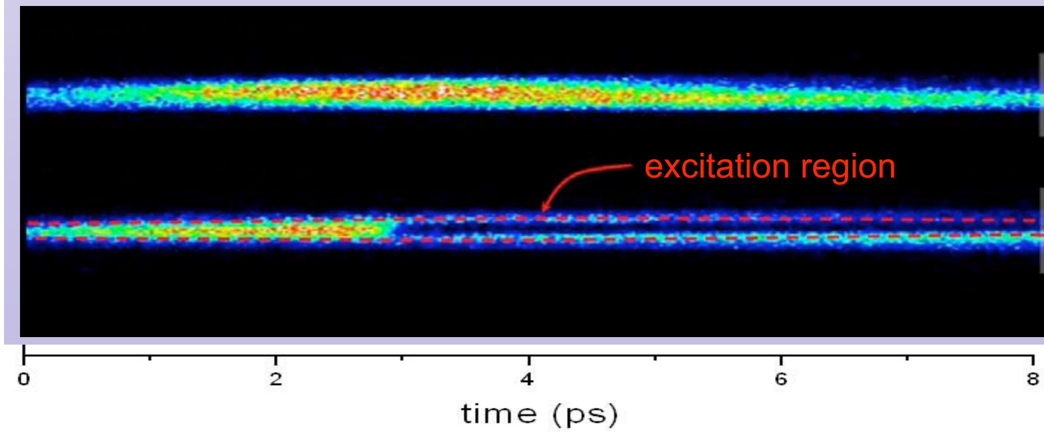
A.W. Cavalieri, *et al.*, Phys.Rev. Lett. 94, 114801 (2005)

Sub-Picosecond Pulse Source

Crossed-Beam Topography for timing at the SPPS



A. Lindenberg, *et al* (to be pub. *Science*)

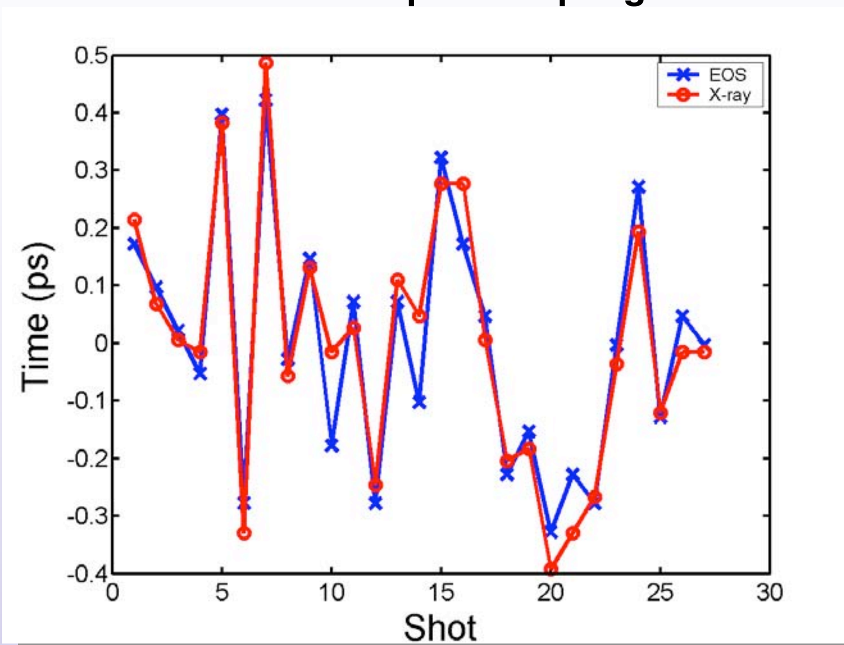


- Crossed-beam technique transforms temporal information into spatial information.
- Measures complete time history in a single shot.
- Position of edge indicates x-ray/laser timing

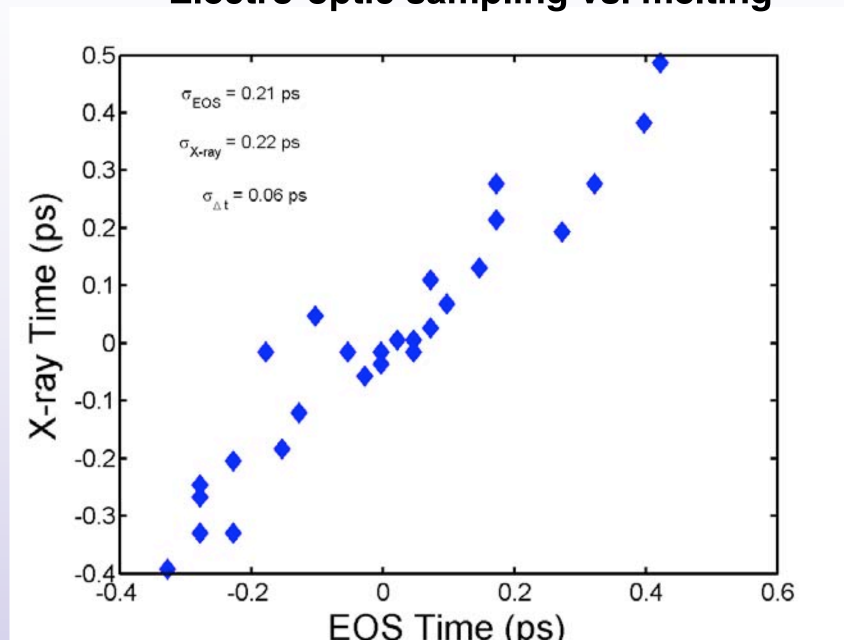
Sub-Picosecond Pulse Source

Jitter of synchronized laser with respect to x-rays at SPPS: EO / melting

Electro-optic sampling



Electro-optic sampling vs. melting



- 30 shots recorded at 1Hz rate
- EO timing accuracy: ~ 30 fs
- Melting timing accuracy: ~ 50 fs
- Agreement between two measurements ~ 60 fs RMS

ALS BL 6 Collaboration

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DESY
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LCLS Collaboration

Five science teams