X-Ray Scattering Studies of Materials Using Ultra-Short Pulses

Roger Falcone

UC Berkeley

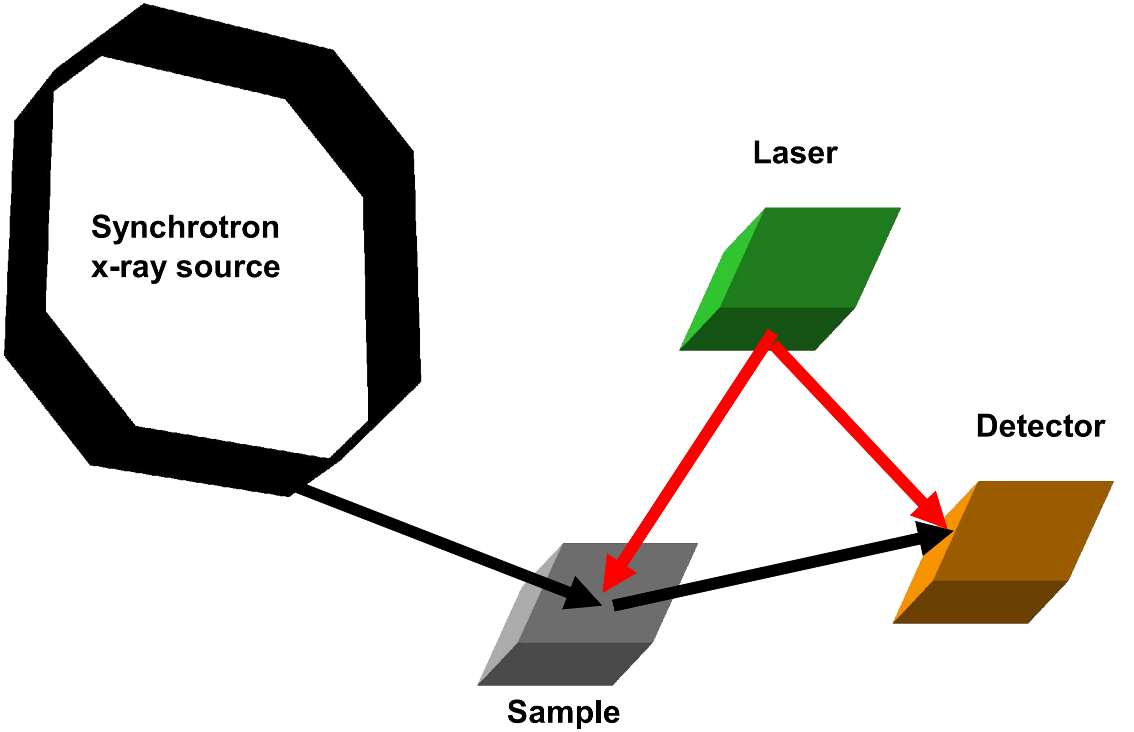
LBNL

SLAC

and the ALS and SPPS Collaborations

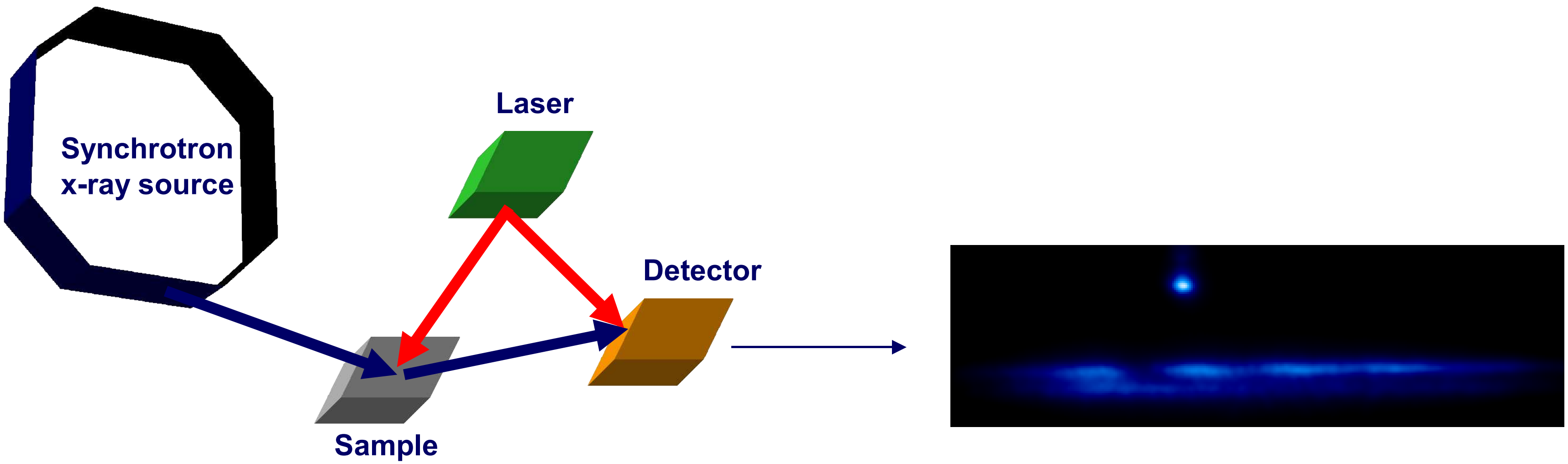
“Workshop on 4th Generation Light Sources and Ultrafast Phenomena” December 2005

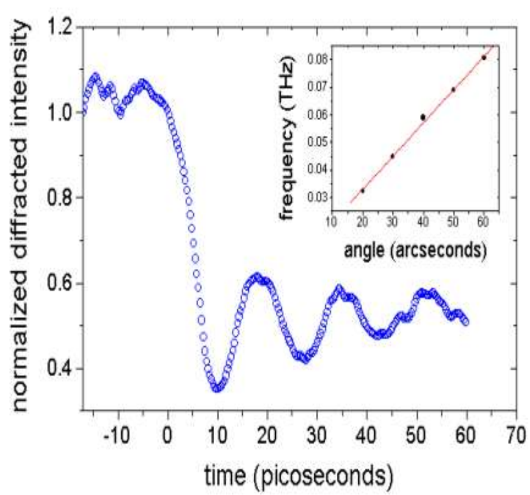
Short pulse lasers and **synchrotron x-ray sources** can **create** and **probe** dynamic and transient states of matter

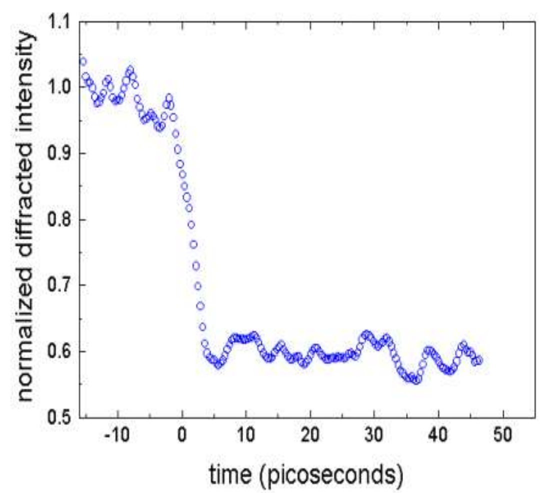


Current facilities: APS, ESRF, SLL, BESSY, ALS

Laser-initiated bond-breaking, heating, and strain can result in phase transitions and be probed by time-resolved x-ray diffraction







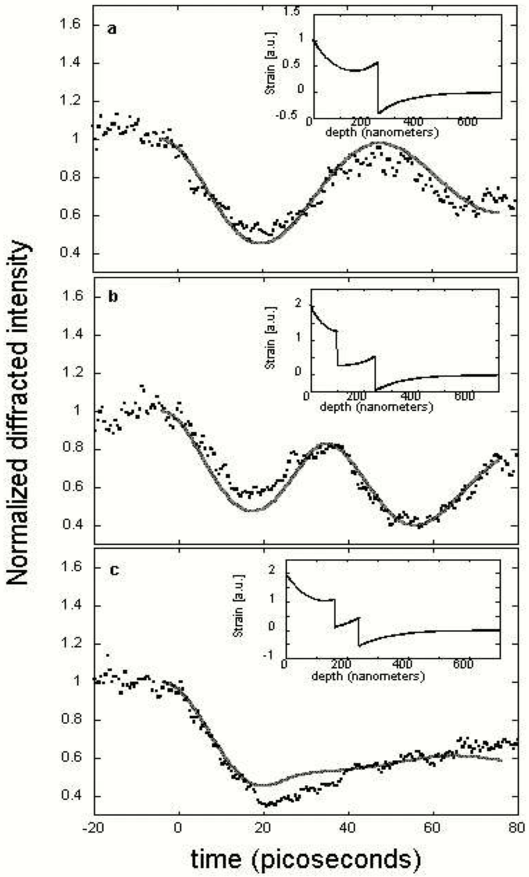
Lindenberg *et al., Phys Rev. Lett*. 84, 111 (2000)

Transient strain can be generated and controlled using multiple ultrashort pump pulses

Single-pulse excitation

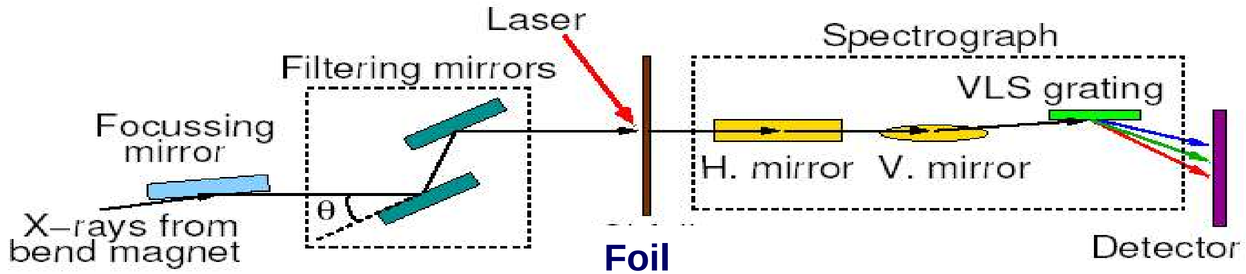
Multiple pulse excitation: constructive interference ($\Delta t \sim 35$ ps)

Multiple pulse excitation: mode cancellation mode ($\Delta t \sim 18$ ps)



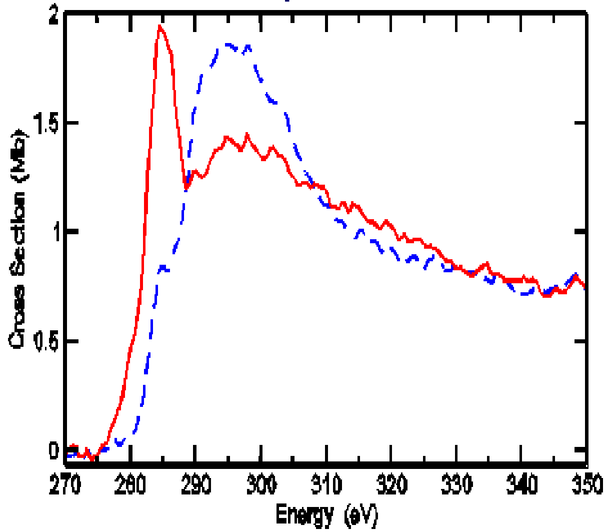
Lindenberg *et al*. Optics Letters, **27**, 869 (2002)

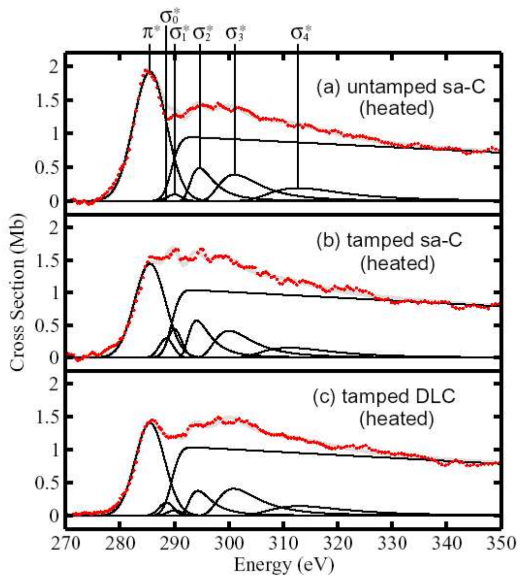
High-energy-density matter can be probed by time-resolved x-ray absorption spectroscopy



Foil Sample

solid / liquid carbon



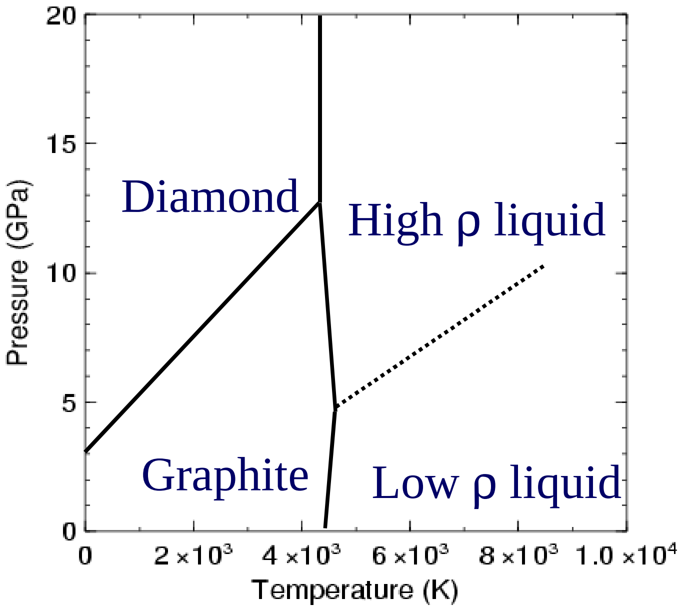


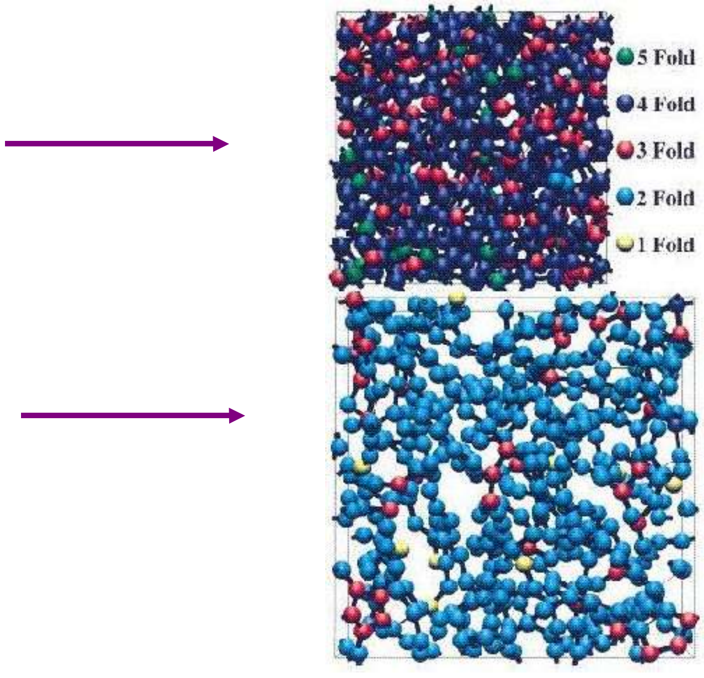
e.g., supports calculations indicating that the low-density phase of liquid carbon is predominately sp-bonded

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

X-ray absorption of liquid carbon

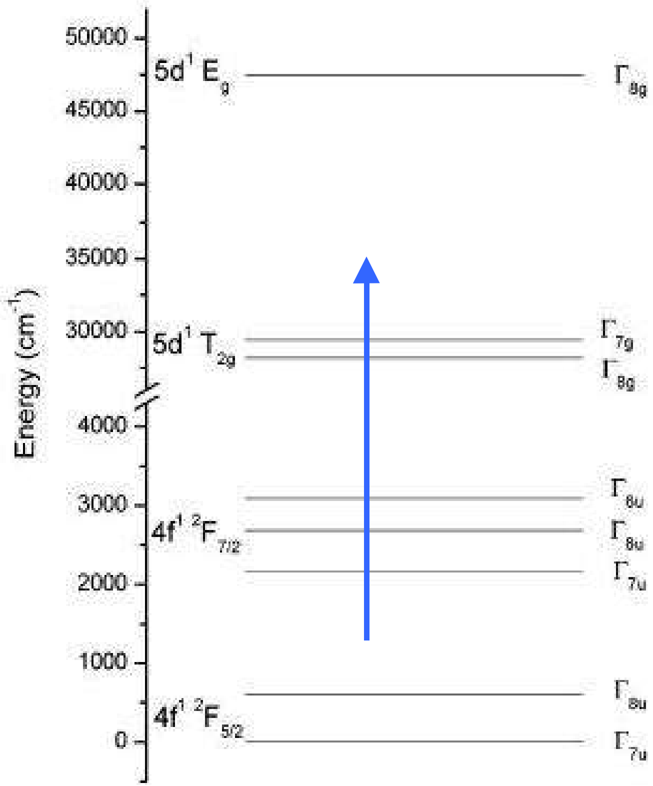
- Liquid carbon present in the interiors of planets Uranus and Neptune. Liquid carbon not stable ambient pressure.
- Molecular dynamics calculations: High density r liquid predominantly sp^3 coordination, low r liquid mainly sp, Glosli and Ree, PRL *82*, 4659 (1999)



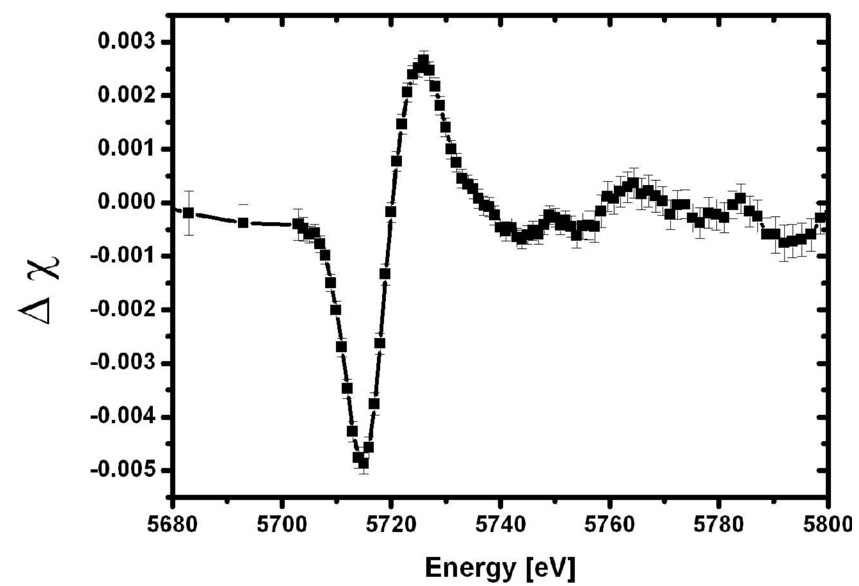


Determine structure of molecules in an excited electronic state

- D. Lowney, P.A. Heimann, T. Allison, W. Lukens, C. Booth, N. Edelstein, R.W. Falcone



$Ce^{3+}Br_6$

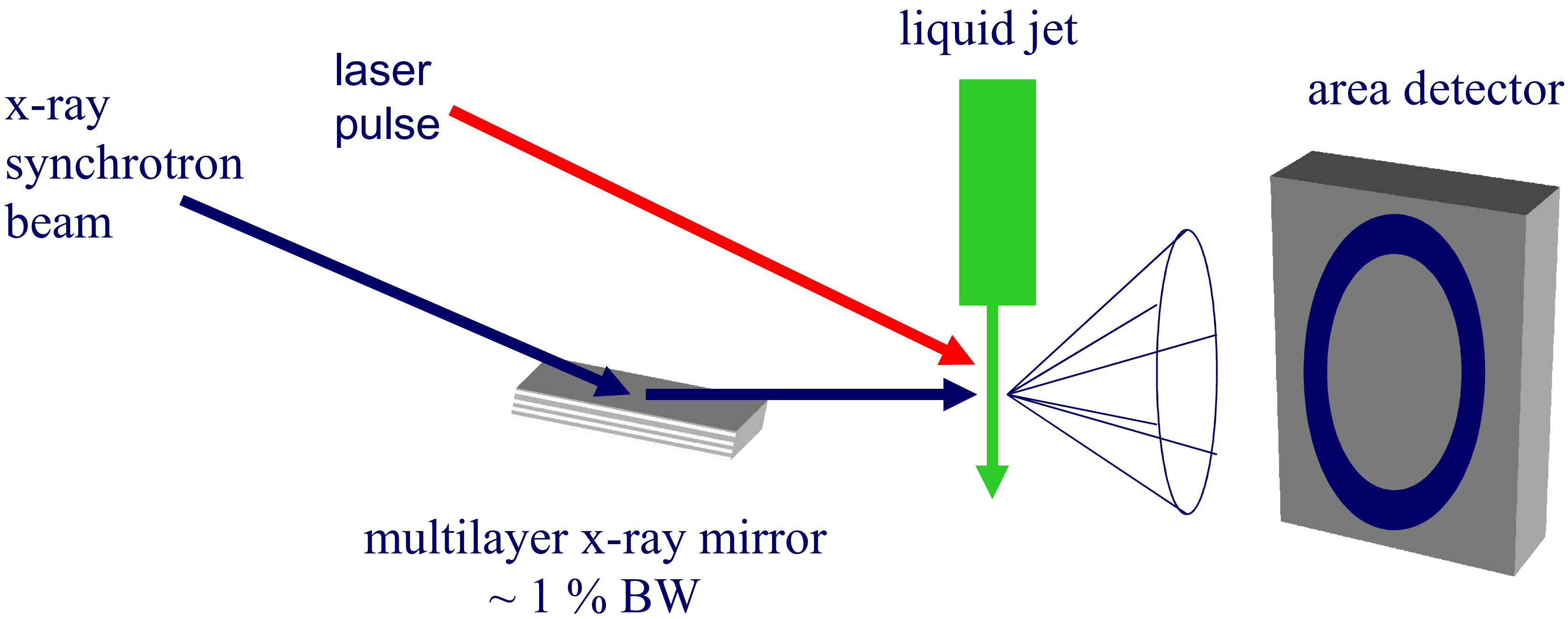


- Theory by L. Seijo and Z. Barandiaran predicts ligand-bond distances:

$$R_e[5f^{n-1}6d(t_{2g})^1] < R_e[5f^n].$$

$\Delta R = -0.65A$

Perturbed liquid structures and subsequent dynamics can be probed by time-resolved x-ray scattering

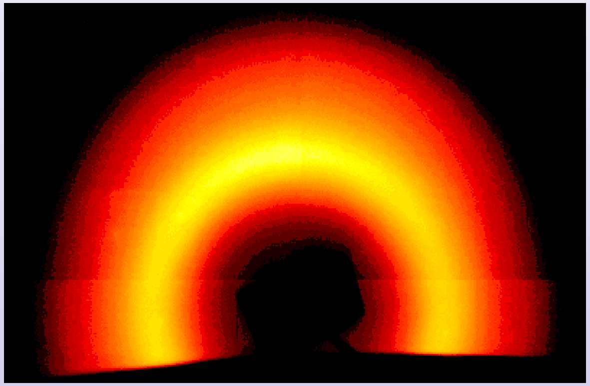


X-ray scattering as a function of angle indicates local structure of WDM

(intramolecular term) (intermolecular term)

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

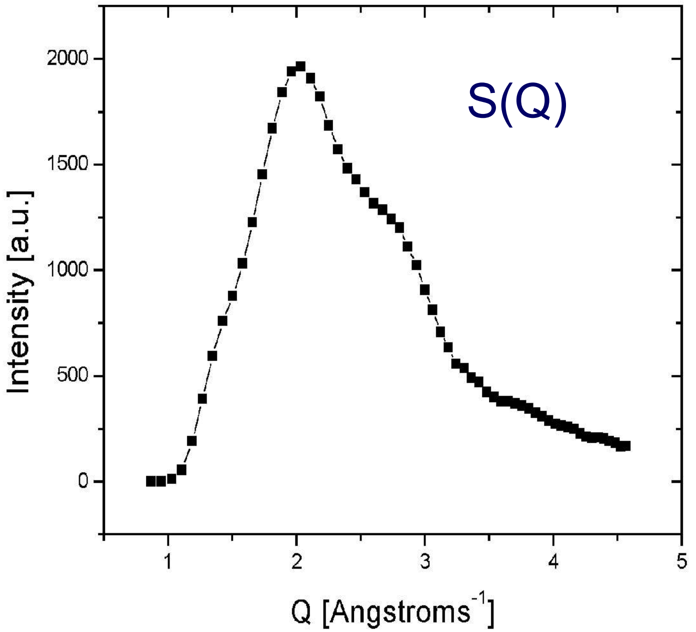
$$H_{ij}(Q) = 4\pi\rho \int_0^\infty r^2 [g_{ij}(r) - 1] \frac{\sin Q r_{ij}}{Q r_{ij}} dr$$



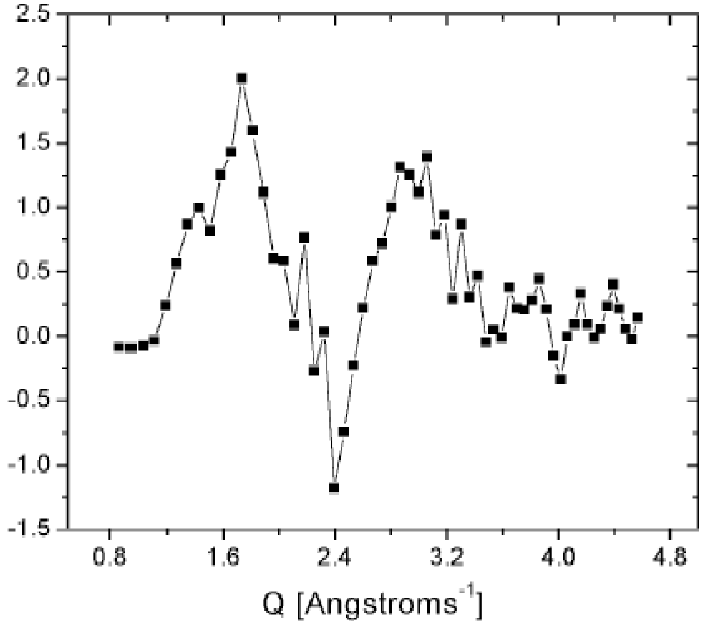
- High Q reflects hard-core region, short length-scales
- Intermediate Q reflects lattice spacing; can be compared with EXAFS pair correlation function
- Low Q reflects long-range, mesoscopic properties

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

Static scattering signal

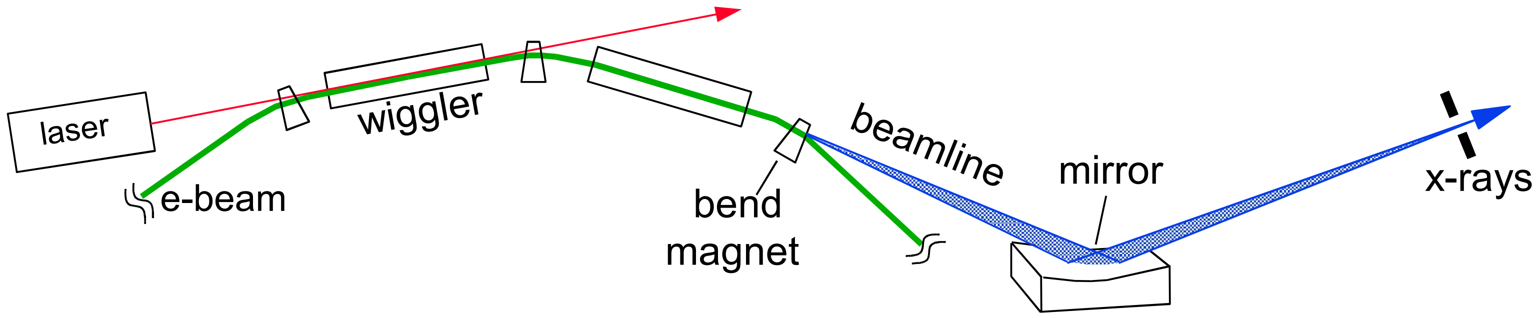


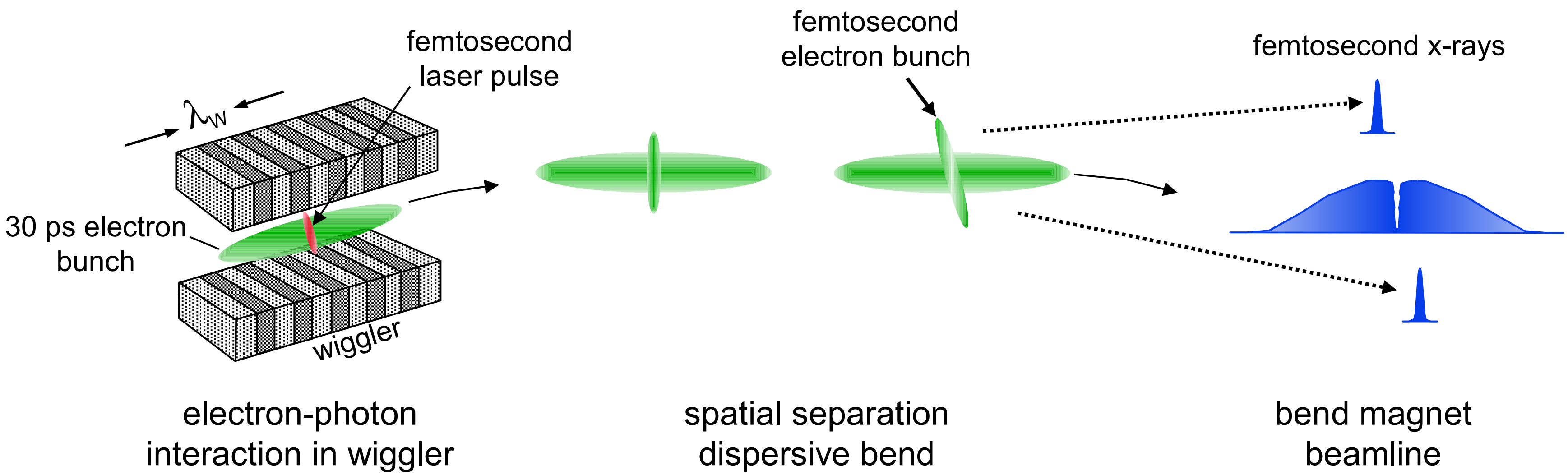
Difference signal at 100 ps following charge injection



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

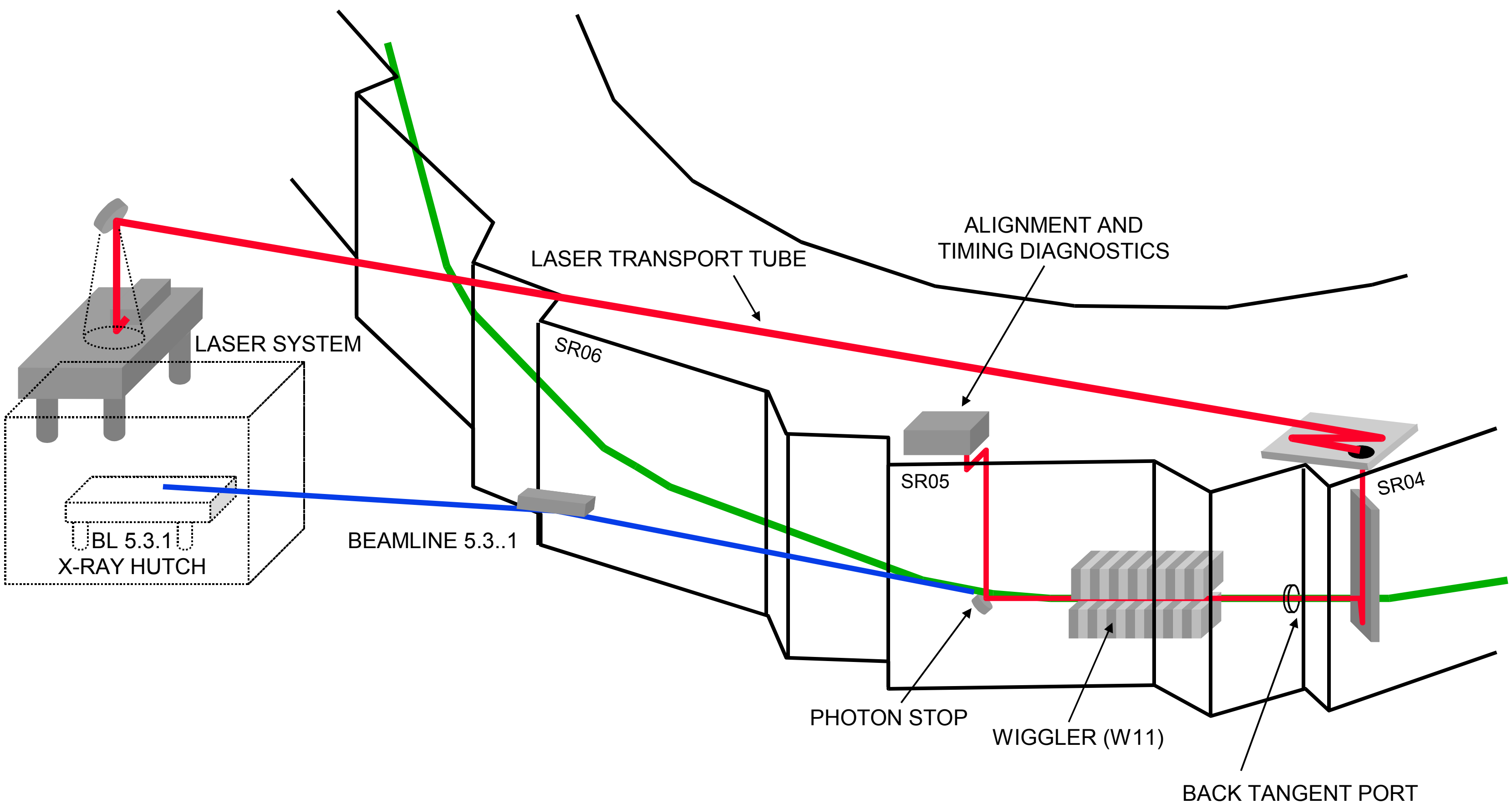
Ultrashort "sliced" x-ray pulses are available





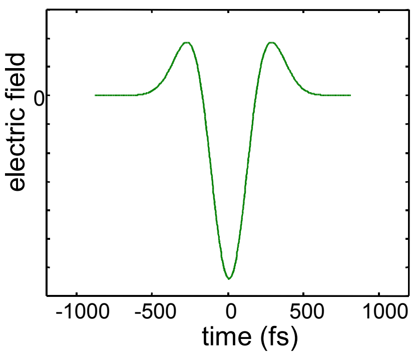
Proposed by Zholents and Zolotorev, Phys. Rev. Lett., 76, 916,1996

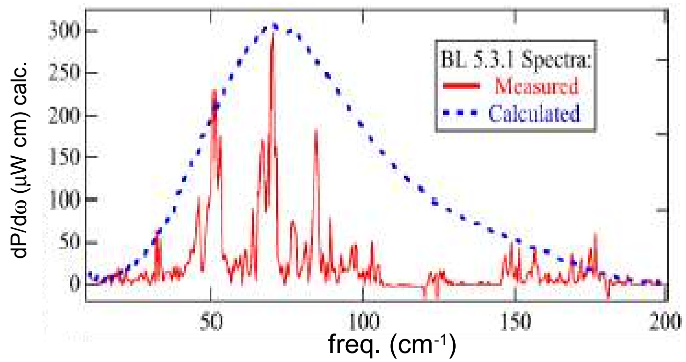
Prototype Slicing Beamline at the ALS



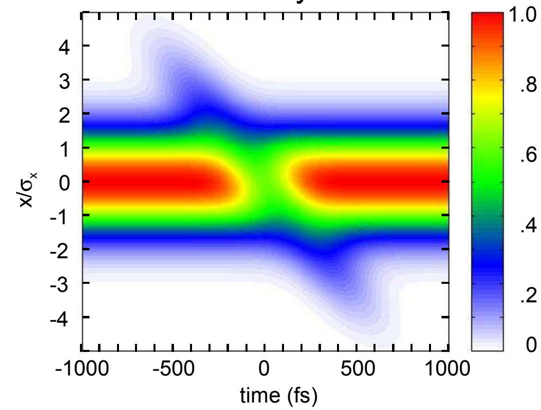
Femtosecond Pulses of Synchrotron Radiation

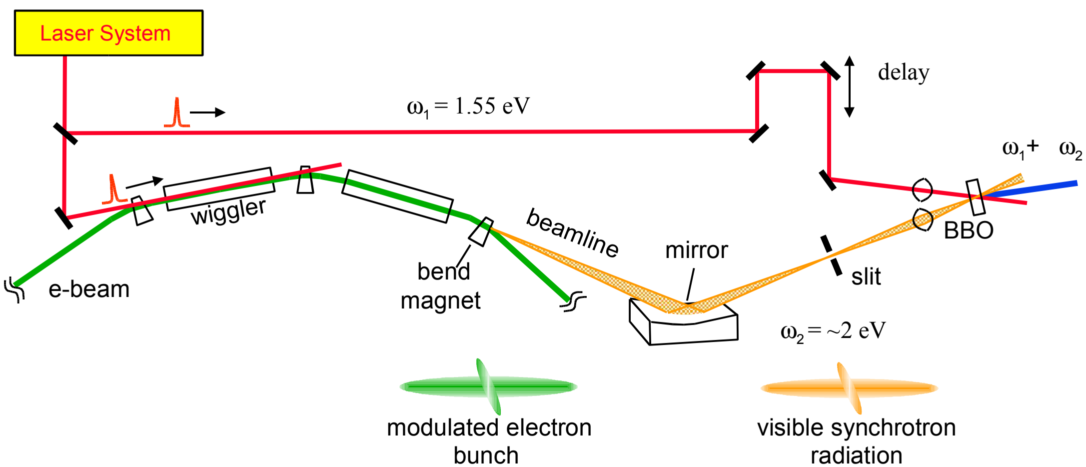
Coherent Synchrotron Radiation



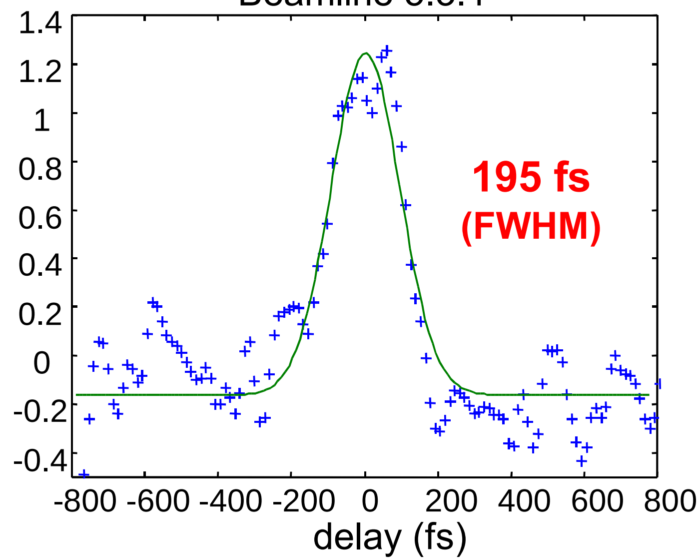


Calculated
Electron Density Distribution



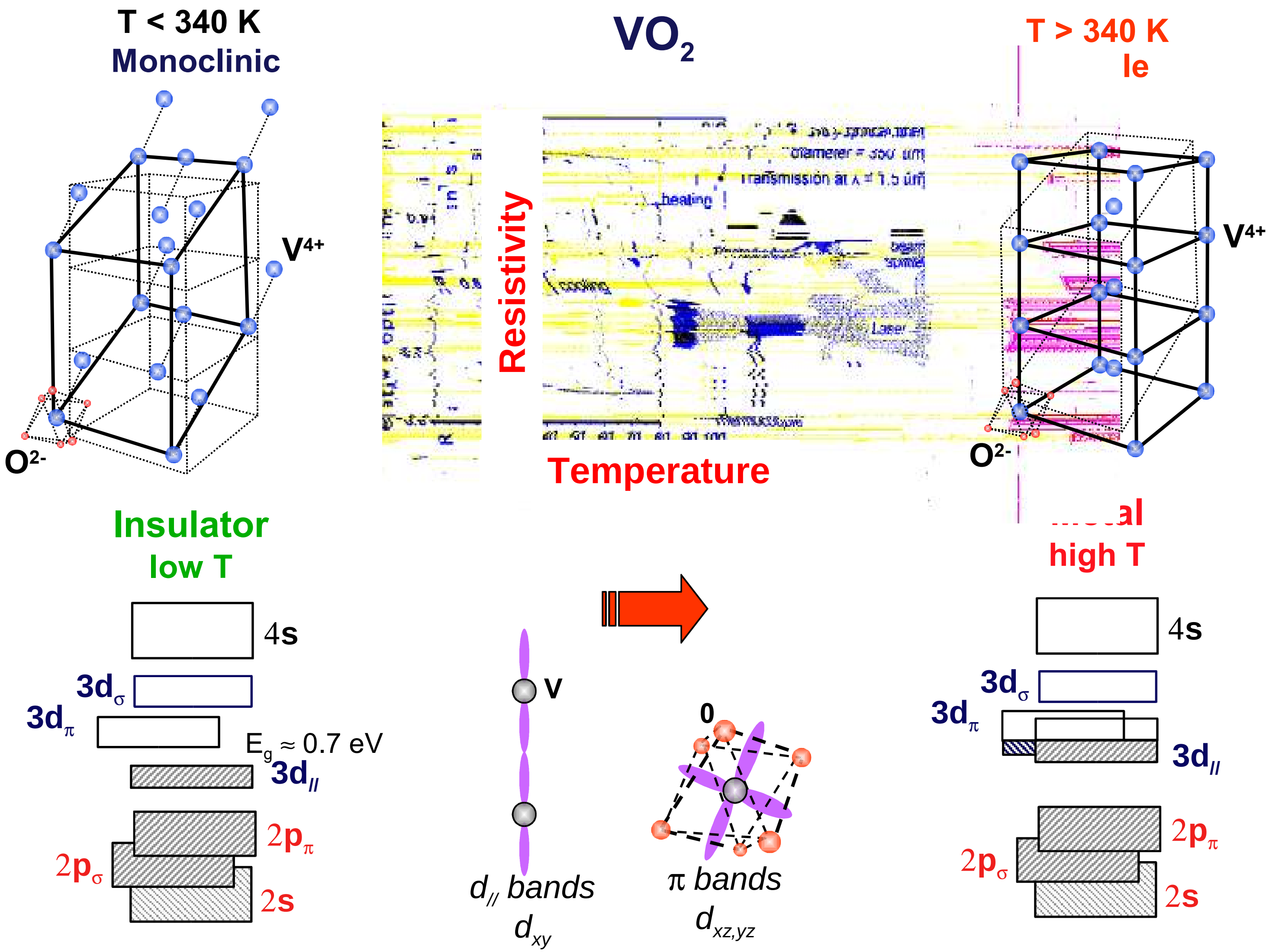


Beamline 5.3.1



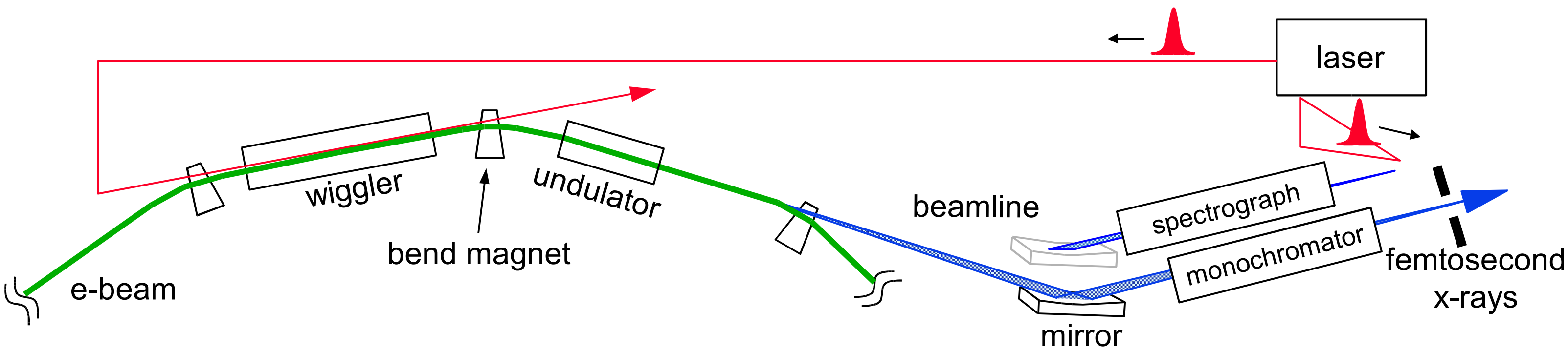
Schoenlein et al., *Science*, **287**, 2237 (2000)

Ultrafast Structural and Electronic Transitions in VO_2



See talk by Matteo Rini on Thursday

New Femtosecond Undulator Beamline 6.0 at the ALS



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

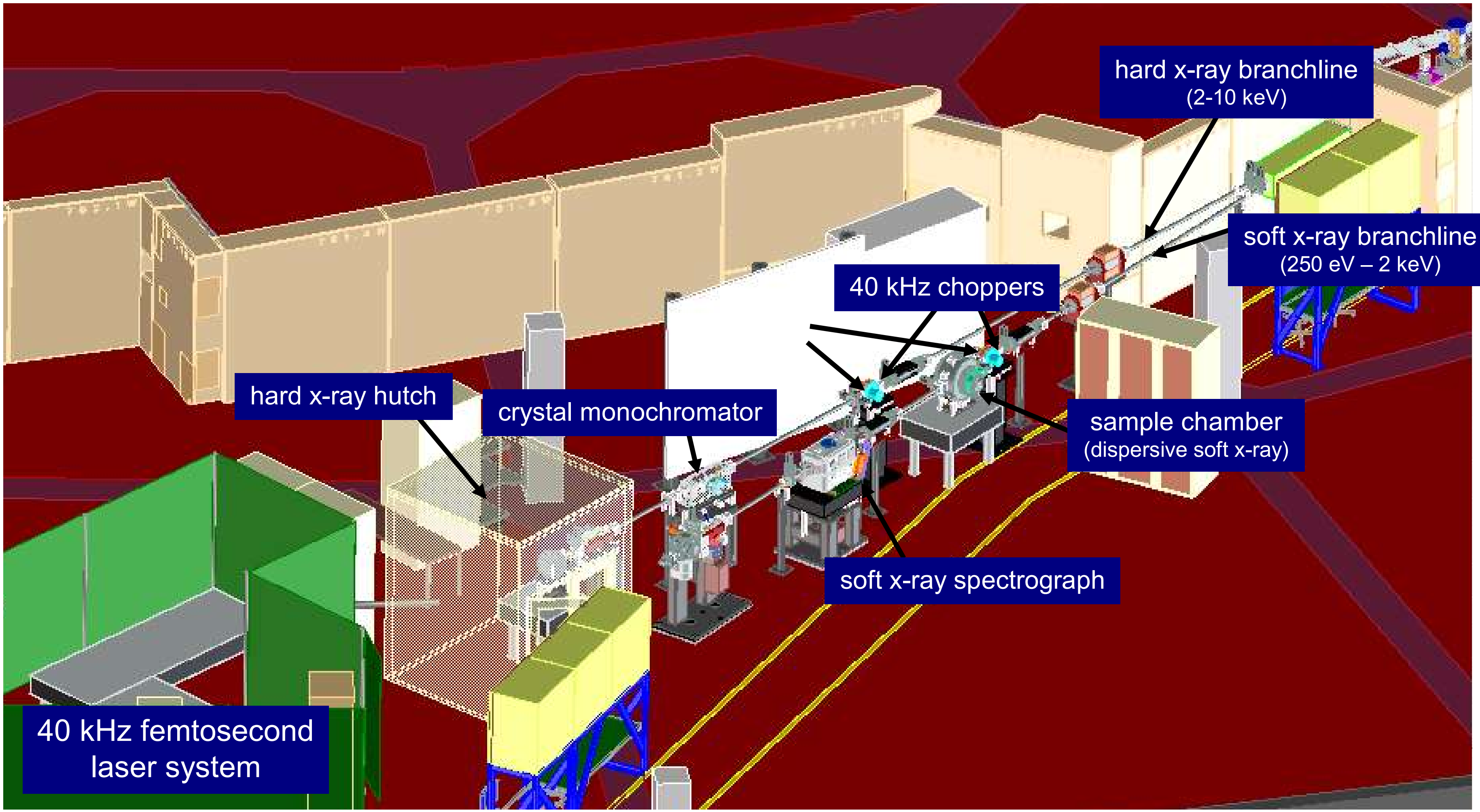
- 30 W (1.5 mJ per pulse, 20 kHz)

  x10 increase in flux

IV. Storage Ring Modifications

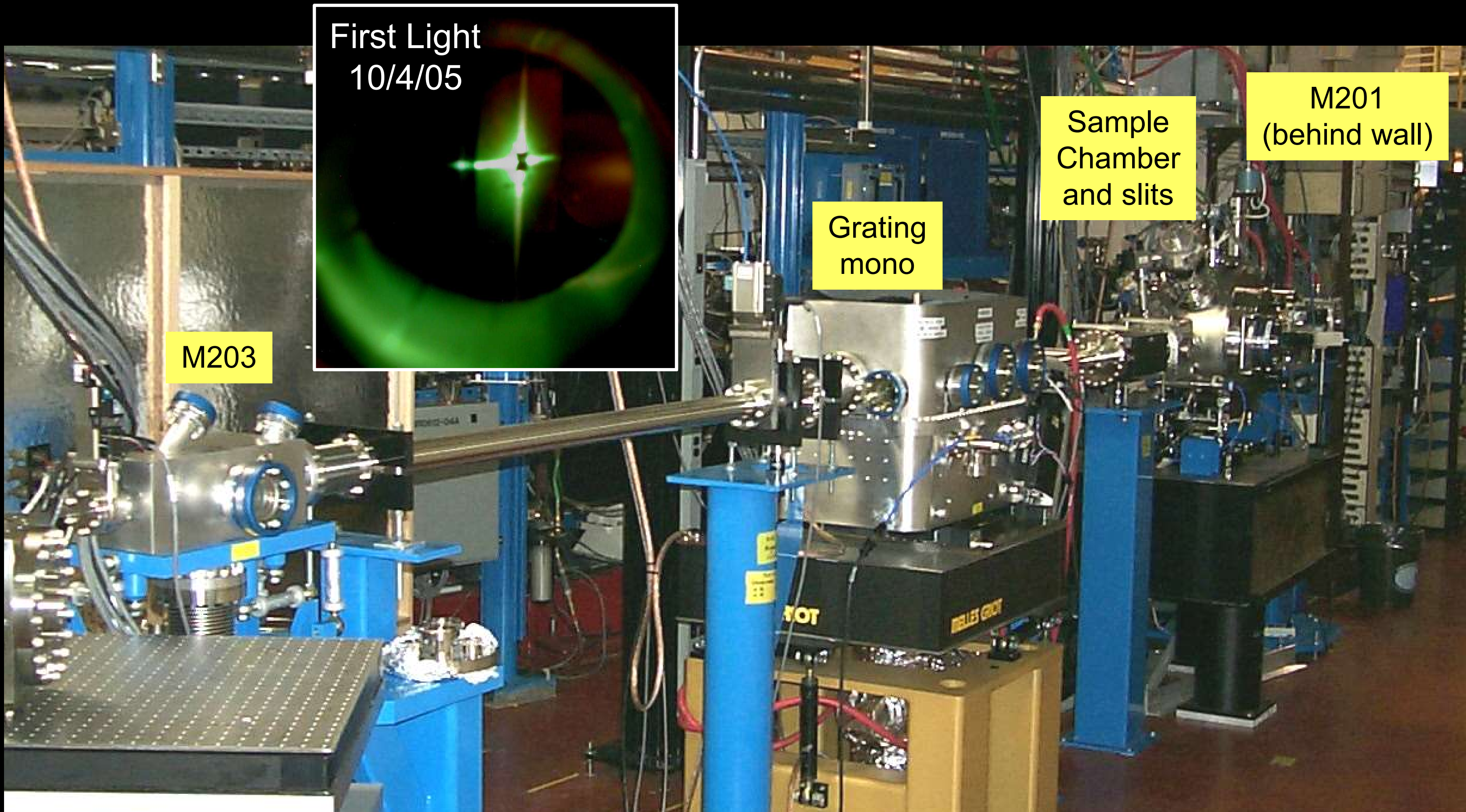
- local vertical dispersion bump – sector 6 and/or 5

New Beamline 6.0 at the ALS Synchrotron



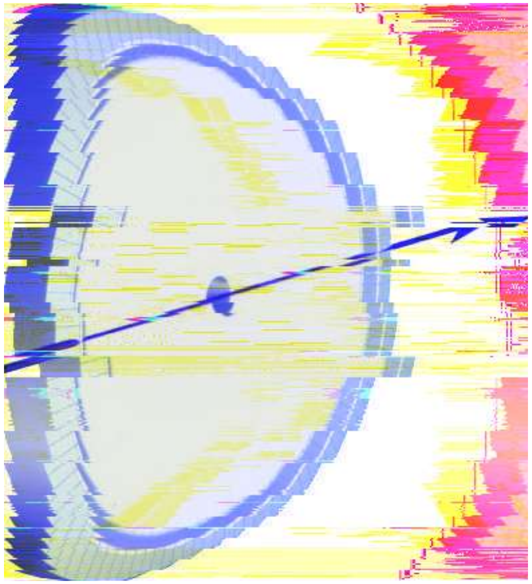
Separate beamlines for 100 fs time resolution studies using hard and soft x-rays

Femtosecond Soft X-ray Beamline 6.0.1.2



X-Ray Chopper

- Reduces power on samples and downstream optics
- Absorbed power: 440 W
- Water-cooled disk with 100 (or 200) slots spinning at 200 Hz
- Frequency: 20 kHz matched to laser repetition rate
- Opening time 2 microseconds

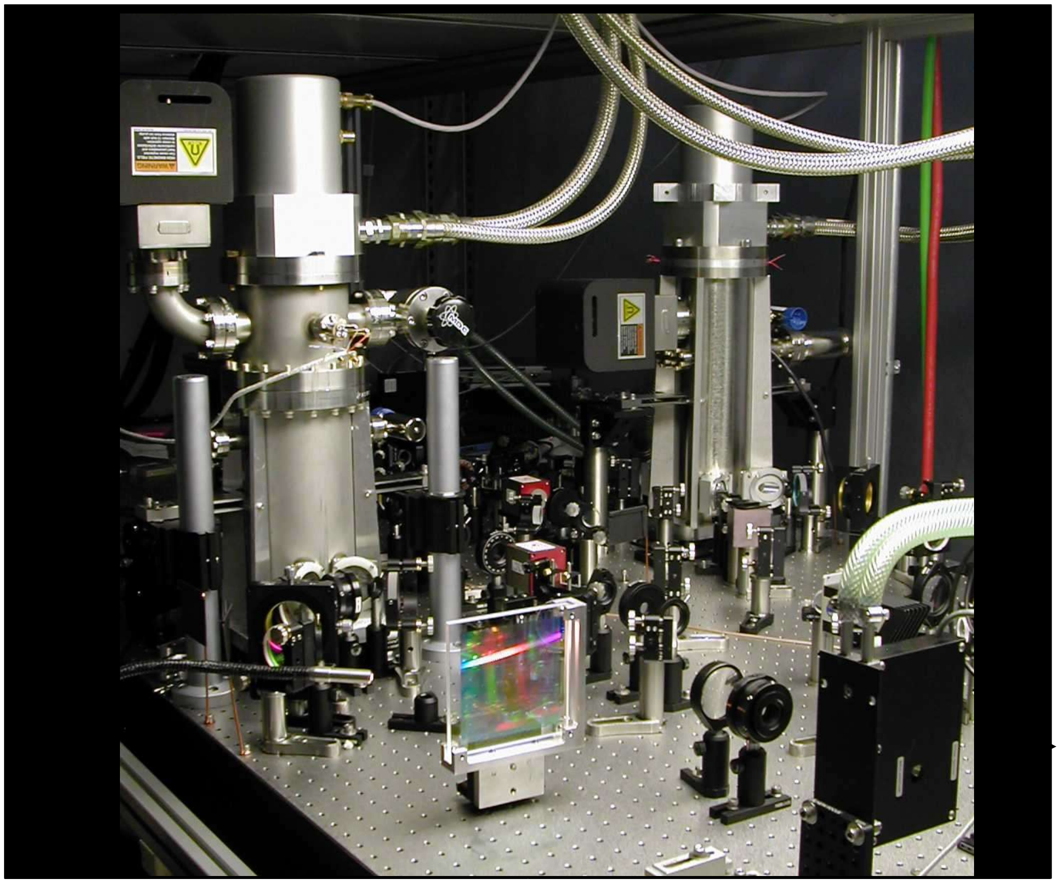


Femtosecond Laser System

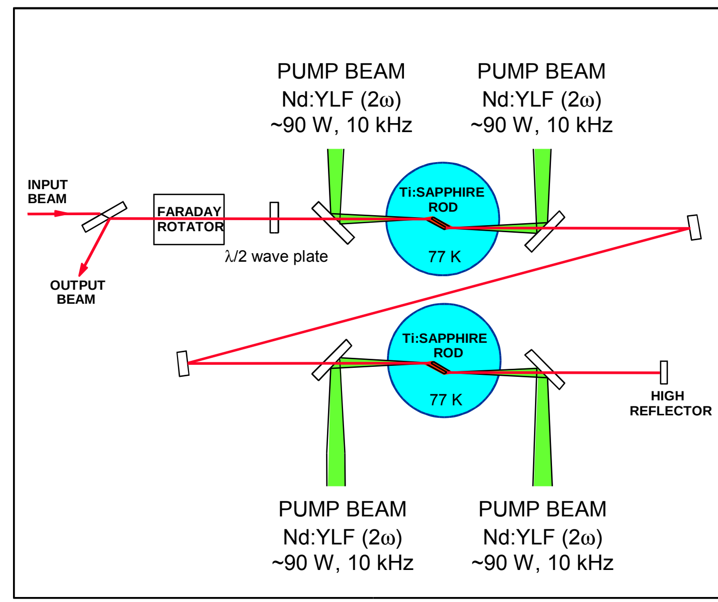
R. Wilcox, R. Schoenlein

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 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

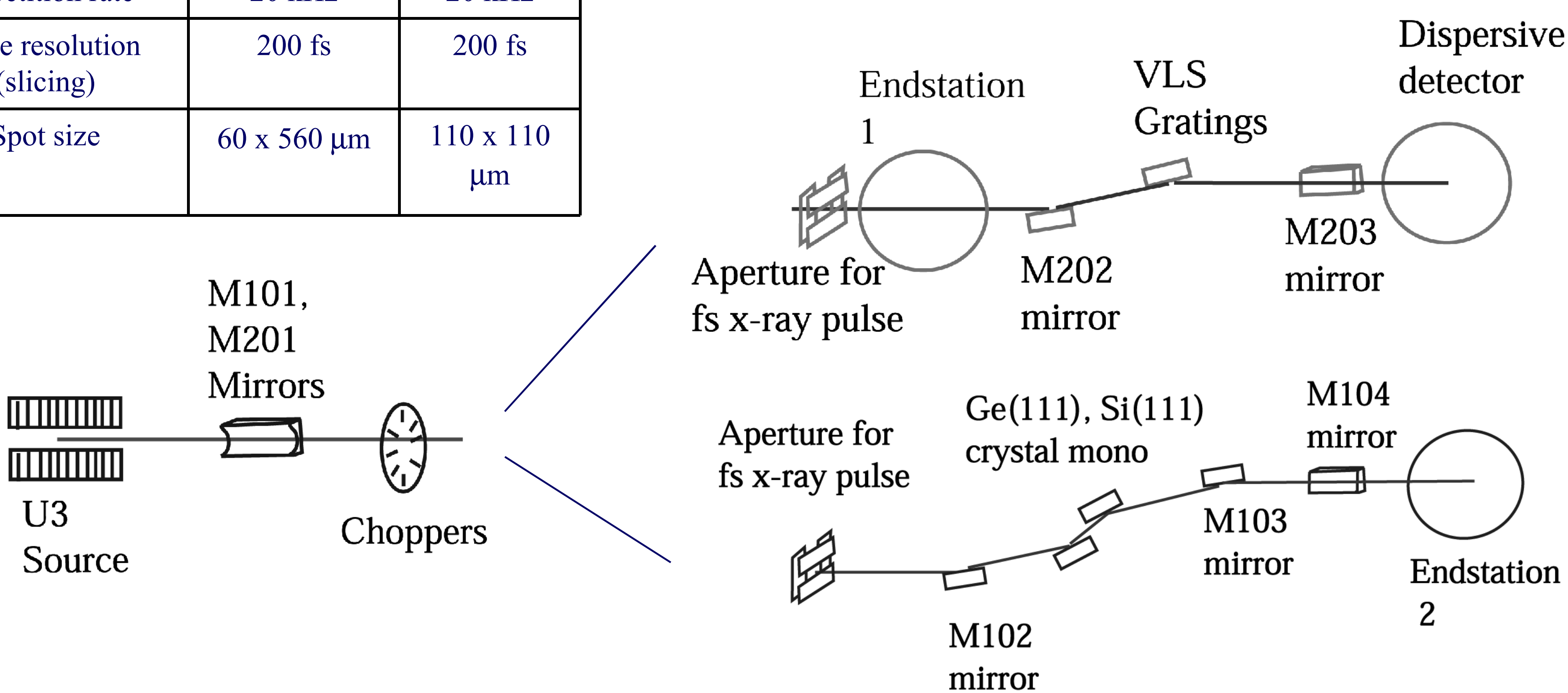


Beamline 6.0 Data Sheet

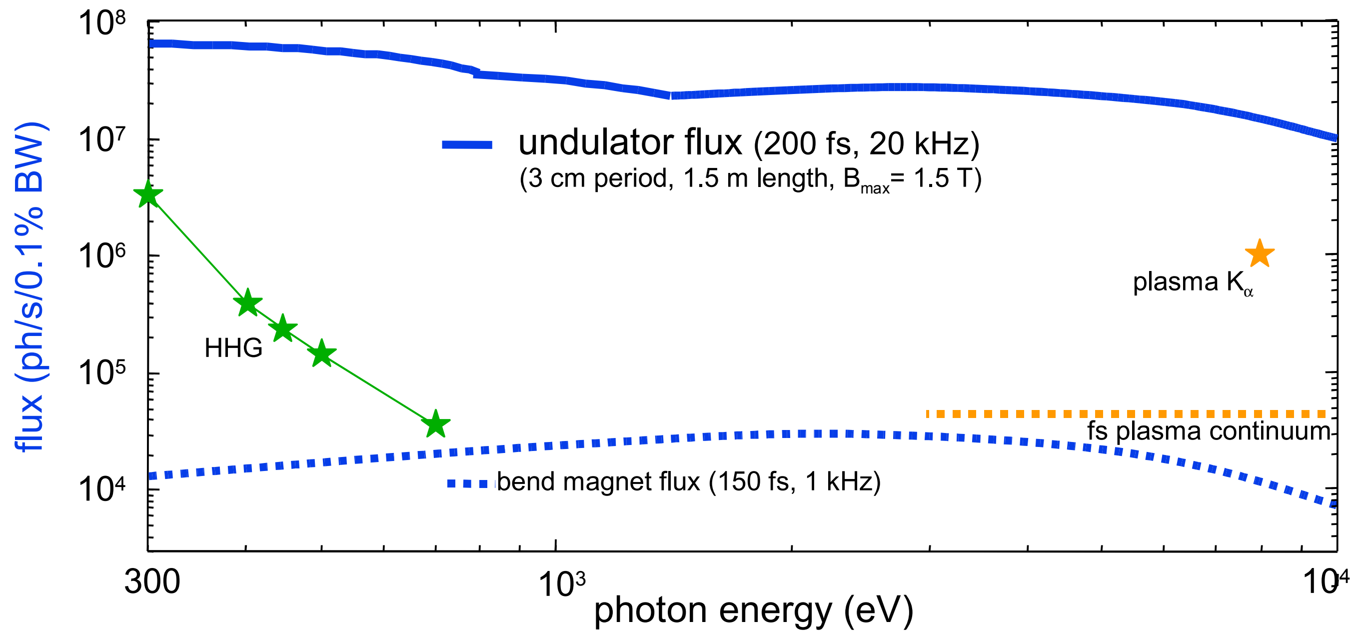
	BL 6.0.1.2	BL 6.0.1
Photon energy range	**120 – 1800** eV	**2 – 10 keV**
Flux (fs slicing pulses, at sample)	1 x 10^6 (1/s 0.1% bw)	1 x 10^5 (1/s)
Flux (70 ps pulses, at sample)	1 x 10^{10} (1/s 0.1% bw)	1 x 10^9 (1/s)
Energy resolution ($\Delta E/E$)	6 x 10^{-4}	3 x 10^{-4}
Repetition rate	20 kHz	20 kHz
Time resolution (slicing)	200 fs	200 fs
Spot size	60 x 560 µm	110 x 110 µm

	Laser system
λ	800 nm
Pulse energy	1 mJ
Repetition rate	20 kHz
Pulse duration	70 fs

For information contact: Phil Heimann at ALS, LBNL



Femtosecond X-ray Flux



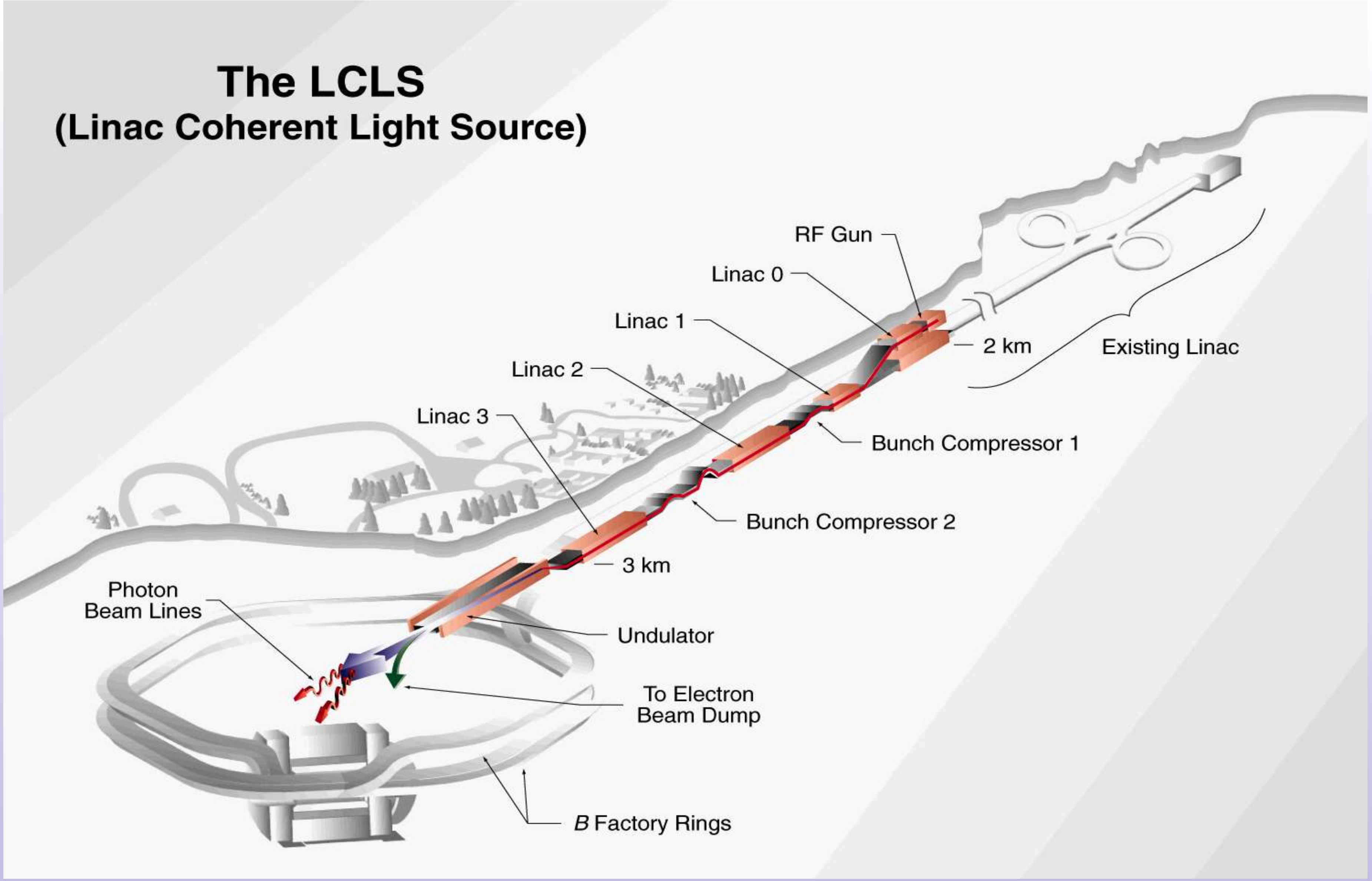
★ HHG flux from F. Krausz, laser: 10 fs, 3 mJ/pulse, 30 W

★ Plasma source flux in mrad^2 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. $6x10^7$ 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



2008-09 Expected commissioning

LCLS Science Thrust Areas

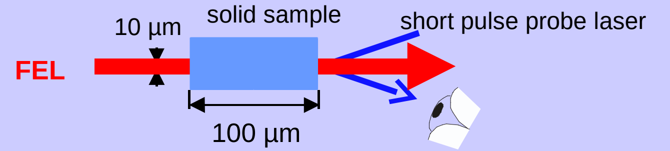
- Atomic, Molecular, and Optical (AMO) science
- High-energy-density (HED) science
- Diffraction studies of stimulated dynamics
- Coherent scattering studies of nanoscale fluctuations
- Nano-particle and single-molecule (non-periodic) imaging

See talk by Ingolf Lindau on Wednesday

High Energy Density Science: One of Five Scientific Thrusts at the LCLS XFEL

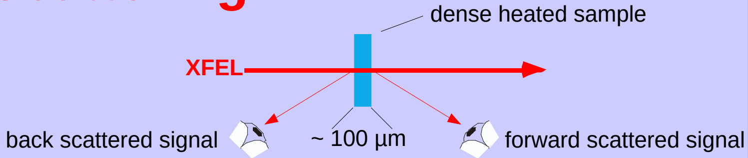
• Creating Warm Dense Matter

- Generate ~ 10 eV solid density matter
- Measure the equation of state



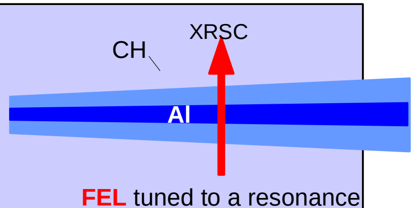
• Probing dense matter with Thomson Scattering

- Perform scattering from solid density plasmas
- Measure n_e, T_e, <Z>, *f*(v)



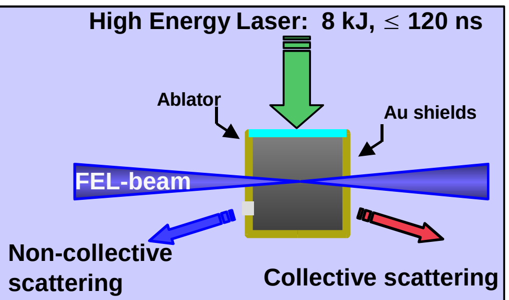
• Plasma spectroscopy of Hot Dense Matter

- Use high energy laser to create uniform HED plasmas
- Measure collision rates, redistribution rates, ionization kinetics



• Probing High Pressure phenomena

- Use high energy laser to create steady high pressures
- Produce shocks *and* shockless high pressure systems
- Study high pressure matter on time scales < 1 ps
- Diagnostics: Diffraction, SAXS, Diffuse scattering, Thomson scattering

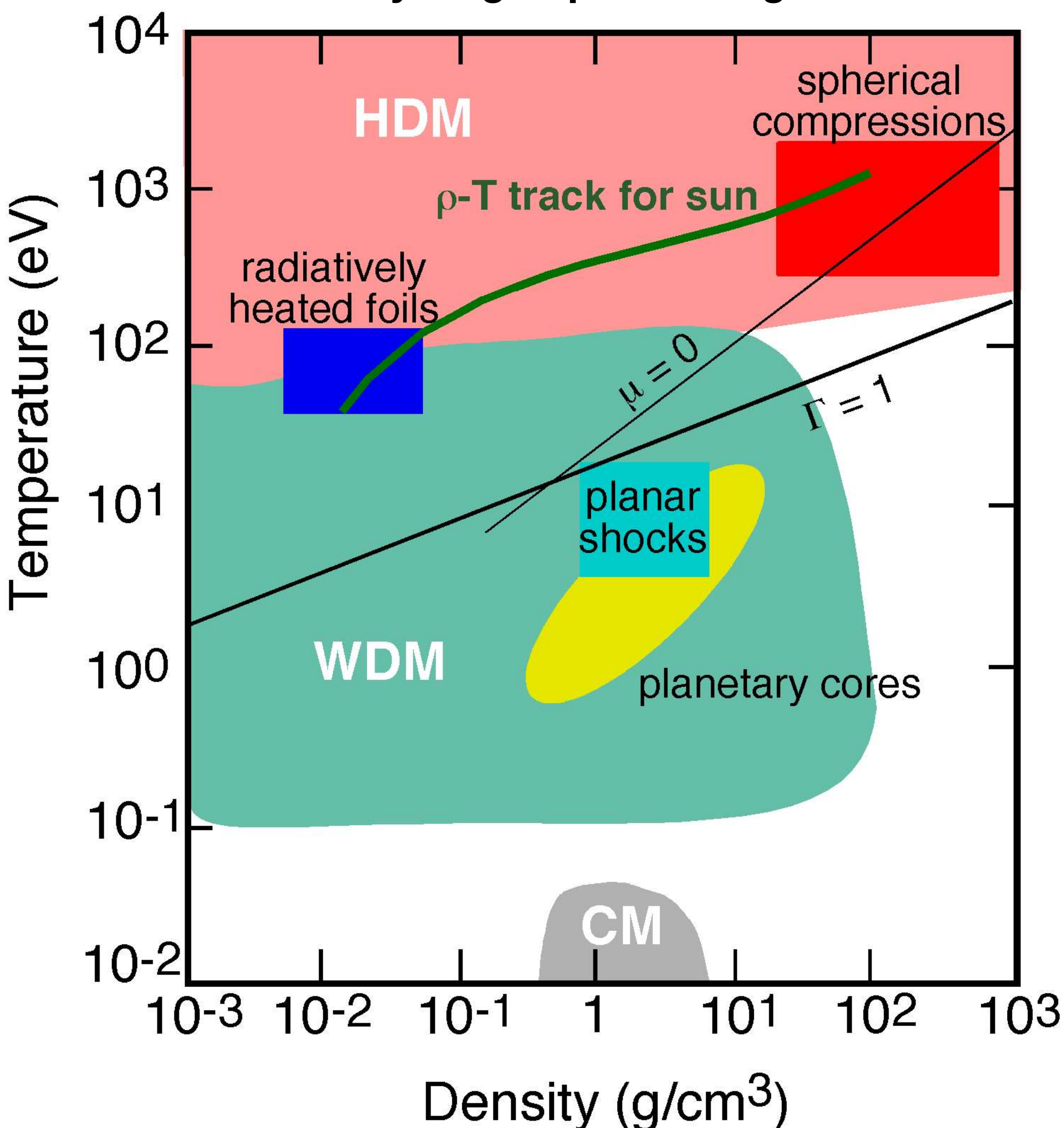


High-Energy-Density Matter Occurs Widely

- **Hot Dense Matter (HDM) occurs in:**
  - Astrophysical systems
  - Plasmas
  - Fusion experiments

- **Warm Dense Matter (WDM) occurs in:**
  - Cores of large planets
  - Systems that start solid and end as a plasma
  - Fusion experiments

Hydrogen phase diagram



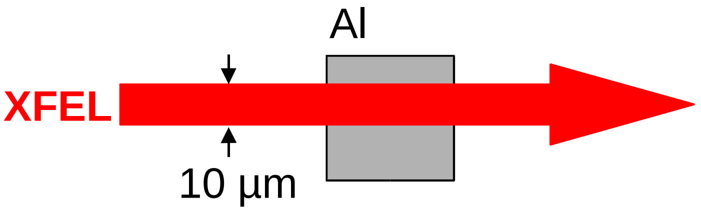
WDM created by isochoric heating will isentropically expand sampling phase space

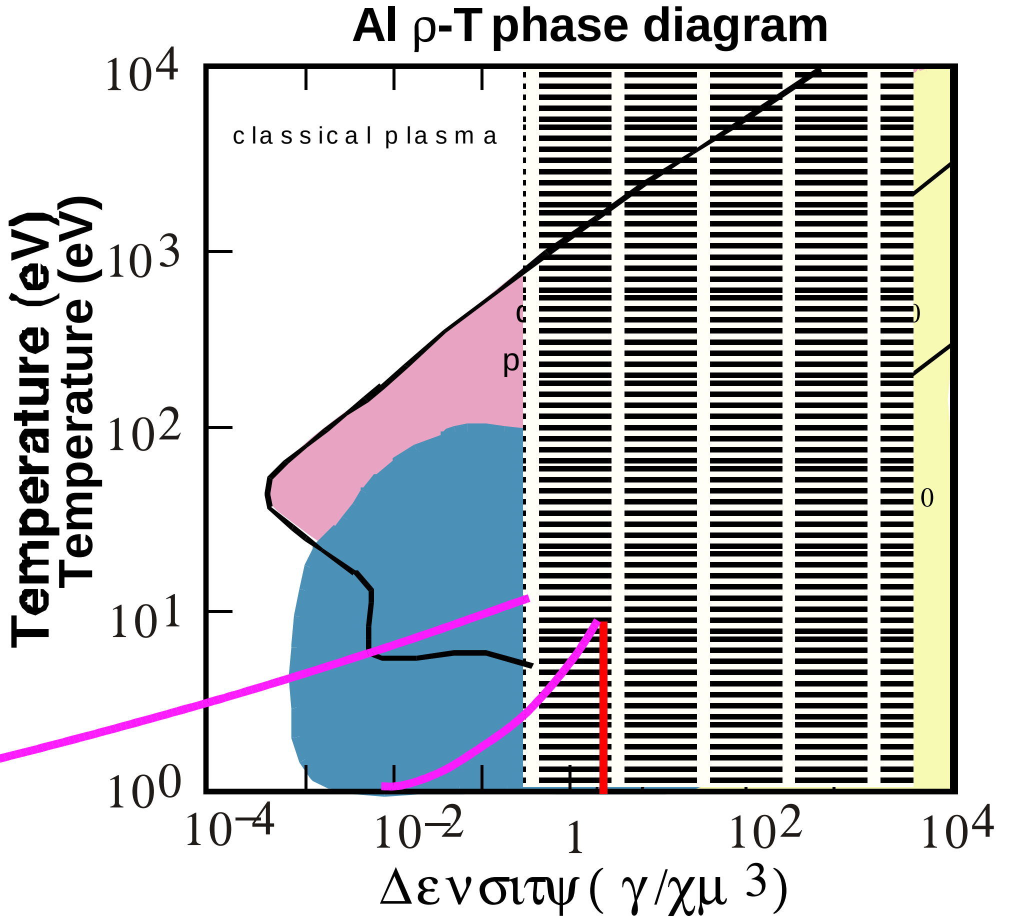
- XFEL can heat matter rapidly and uniformly to create:

- Using underdense foams allows more complete sampling

• Isochores (constant ρ)

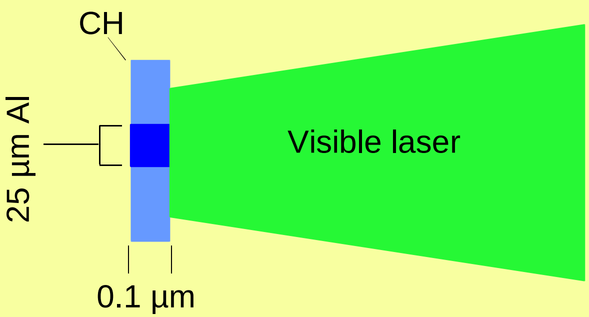
• Isentropes (constant entropy)

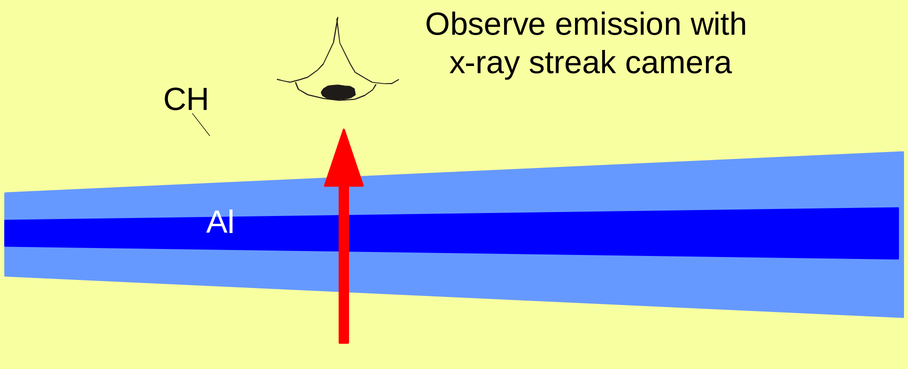




LCLS x-ray laser will enable HED spectroscopy

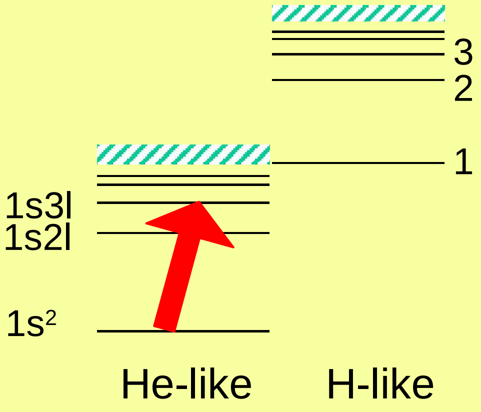
- **Schematic experiment**

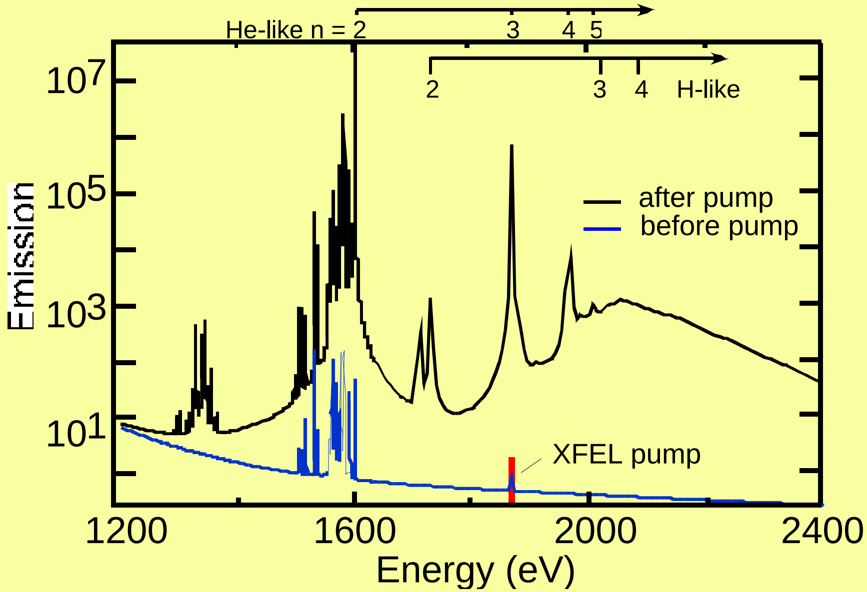




- **t = 0 laser irradiates Al dot**
- **t = 100 ps FEL irradiates plasma**

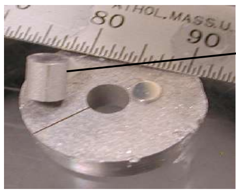
- **Simulations**





Microstate conditions of shocked, high pressure materials controls material strength, spall, and phase transitions

Current:
Post Processing x-ray scattering



Shocked and incipiently fractured single crystal Al slug

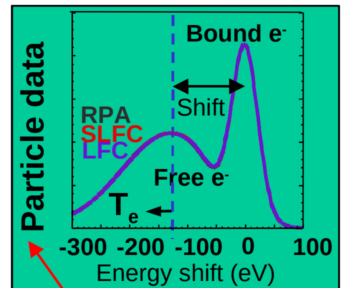
Simulated x-ray scattering

APS Beam

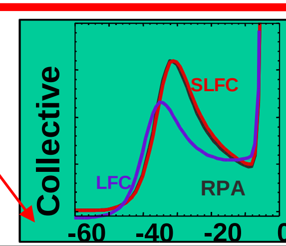
3-D x-ray tomographic reconstruction of dynamic fracture

- ***Diffraction → lattice compression and phase change***
- ***SAXS → sub-micron defect scattering***
- ***Diffuse → dislocation content and lattice disorder***

Future:
Measure during pressure pulse



LCLS



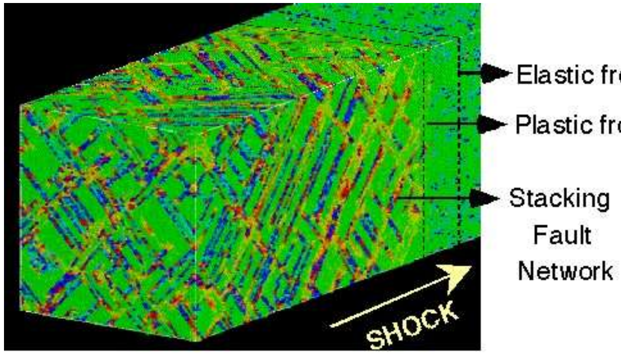
- ***LCLS will provide unprecedented fidelity to measure dynamics of the microstate with sub-picosecond resolution***

- **LCLS HEDS end station will have sufficient signal to perform measurements during the high pressure phase**

see Belak, Lee, et al

Challenge is to match experimental and theoretical capabilities for HEDS studies

Current Status
Simulation



- MD simulation of FCC Cu taking 400,000 CPU hours (From LLNL)

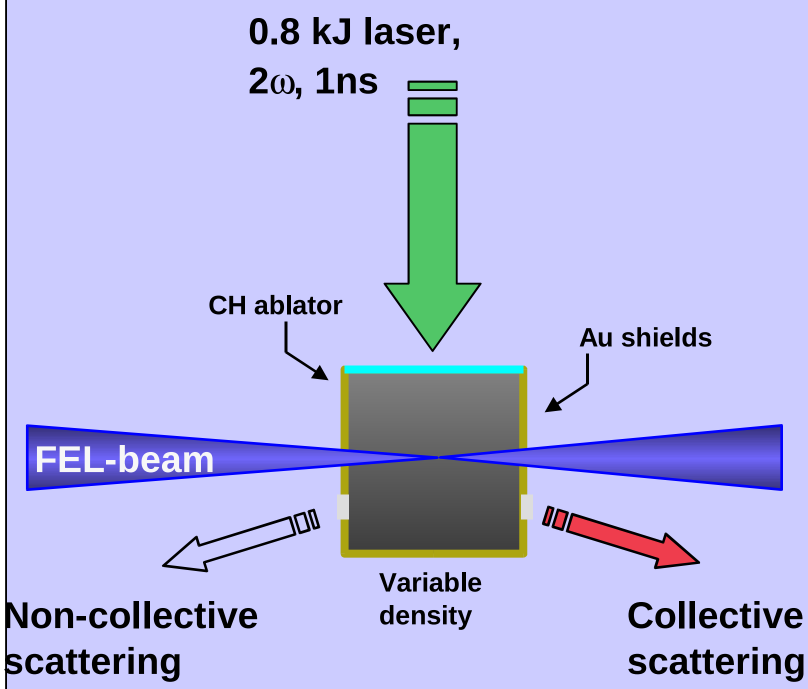
Future with LCLS
Dynamic Experiments

- **Imaging capability**
  - **Point projection imaging**
    - **Phase contrast**
    - **High resolution (sub-μm)**
  - **Direct determination of density contrast**
- **Diffraction & scattering**
  - **Detection of high pressure phase transitions**
  - **Lattice structure, including dislocation & defects**
  - **Liquid structure**
  - **Electronic structure**
    - **Ionization**
    - **T_e, f(v)**

These complement the standard instruments, e.g., VISAR and other optical diagnostics

Future capabilities at LCLS will reduce uncertainties in EOS experiments

Future



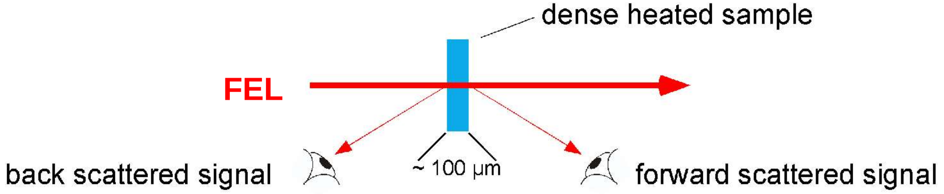
All current measurements maintained. Add direct, *in situ*, measurement of temperature and densities.

LCLS will offer unique capabilities for *in situ* real time probing

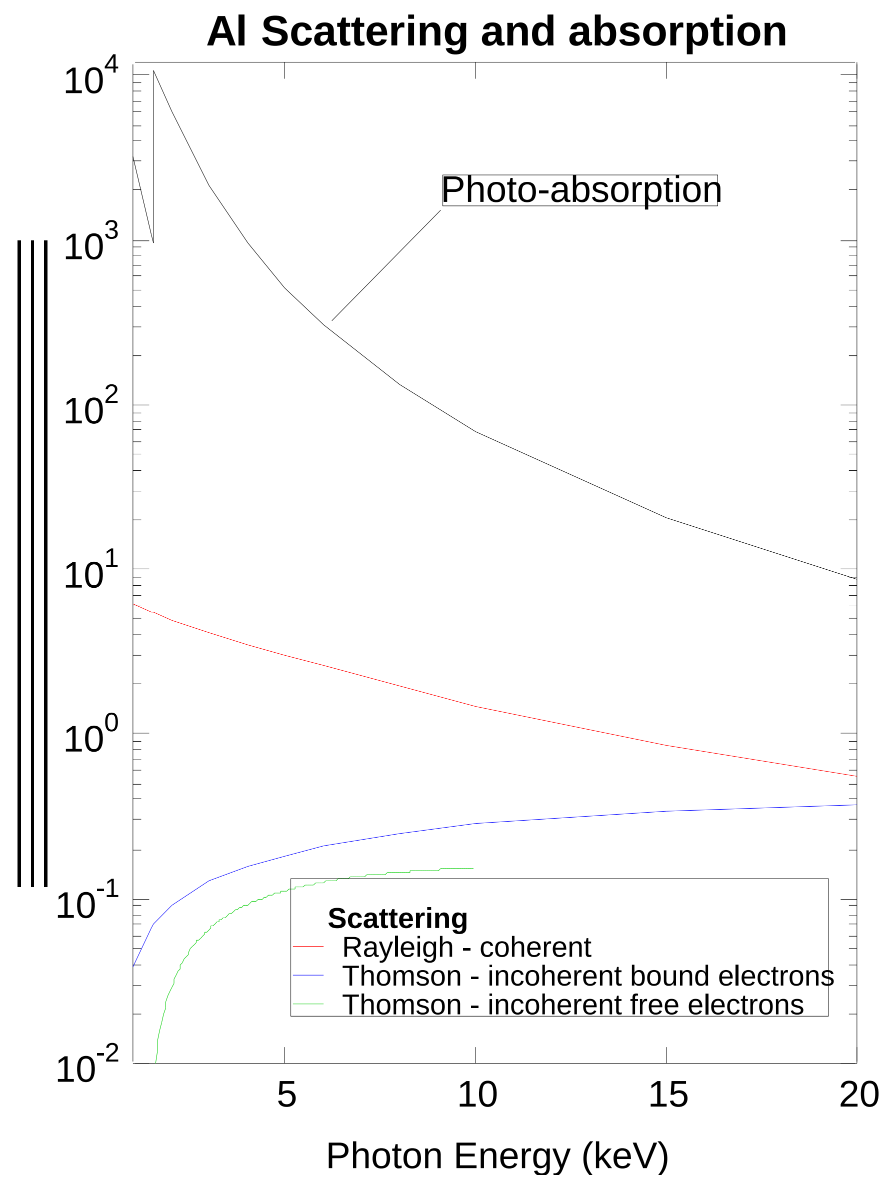
- **Imaging capability**
  - **Point projection imaging**
    - **Phase contrast**
    - **High resolution (sub-μm)**
  - **Direct determination of density**
- **Diffraction and scattering**
  - **Detection of high pressure phase transitions**
    - Detection of melt under shock is very difficult and results have lead to ambiguity and controversy
  - **Liquid structure**
  - **Electronic structure**
    - **Ionization**
    - **T_e, velocity distribution**

These complement the standard instruments, e.g., VISAR and other optical diagnostics

X-ray FEL will be used to probe HED matter

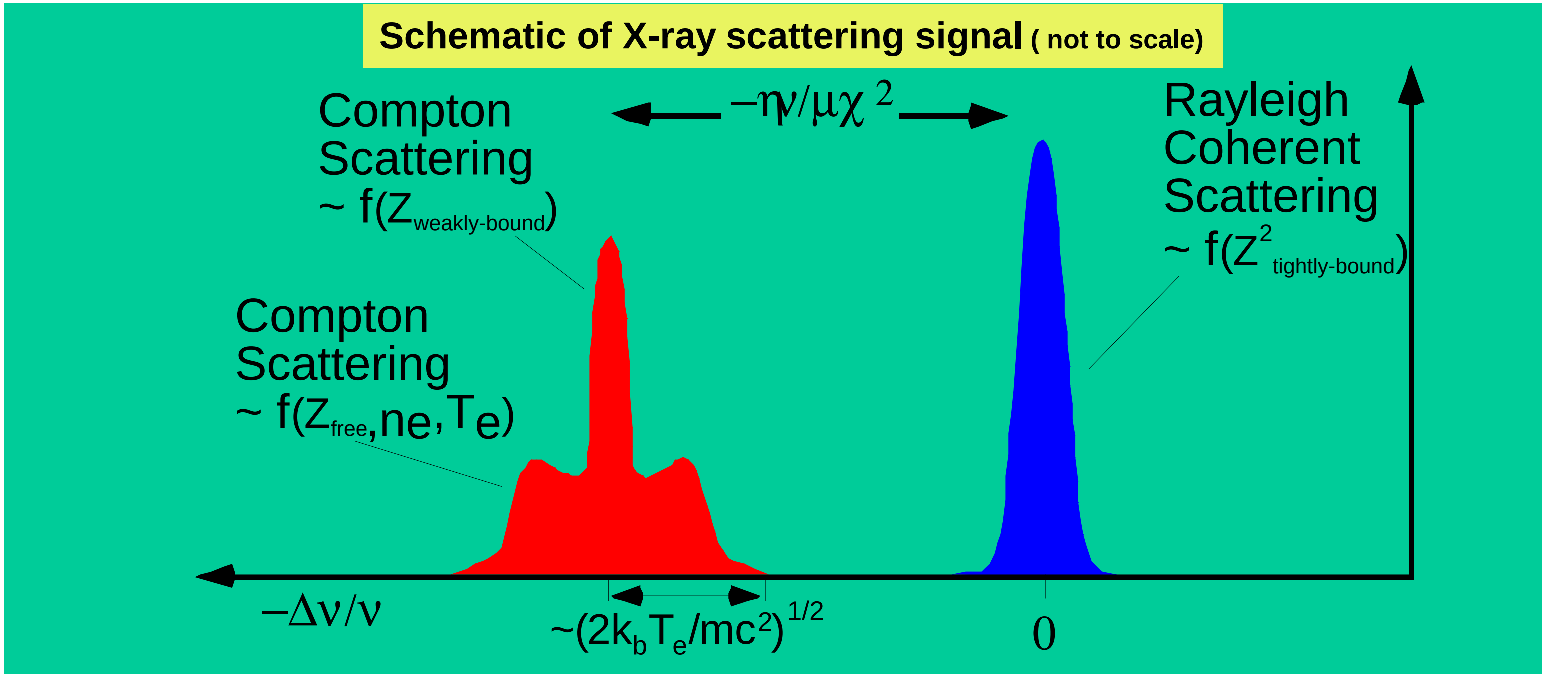


- Scattering from free electrons provides a measure of the T_e, n_e, f(v), and plasma damping
- Due to absorption, refraction and reflection neither visible nor laboratory x-ray lasers can probe high density
- FEL scattering signals will be well above noise for all HED matter



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

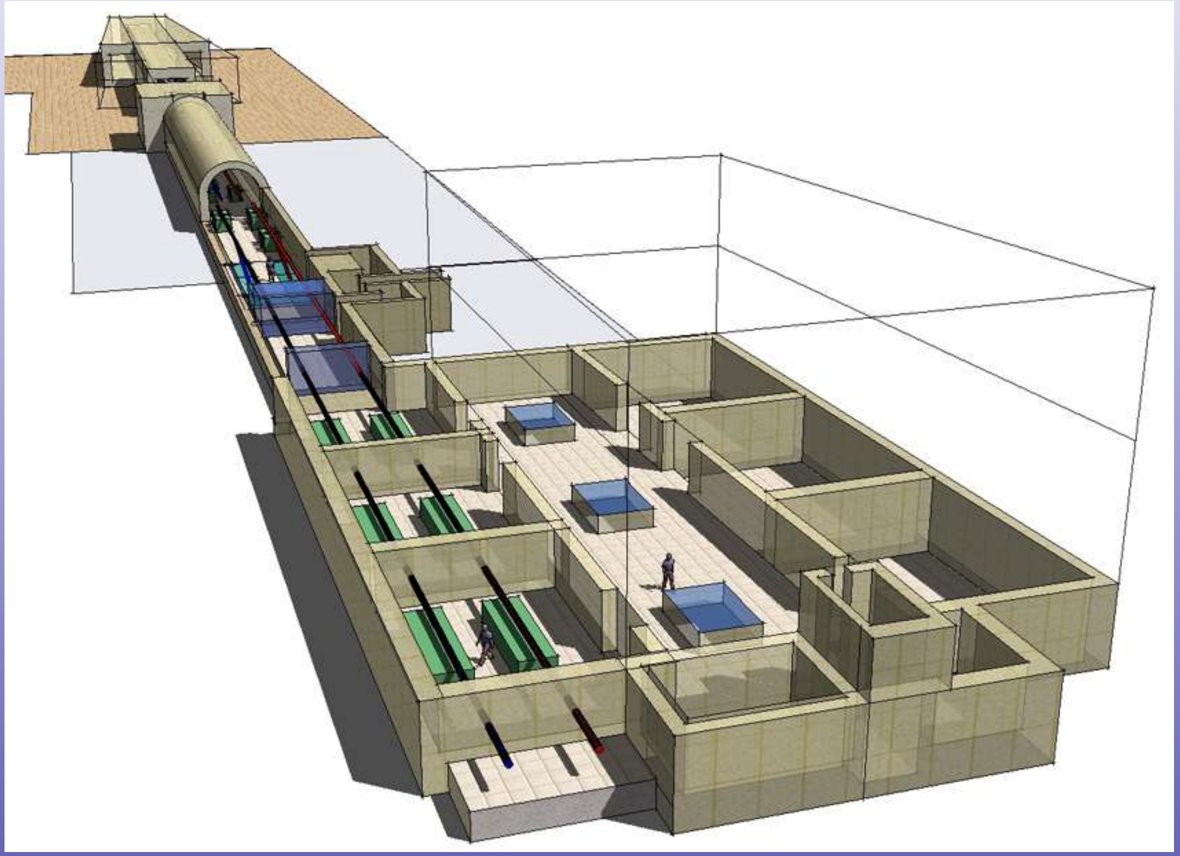
- Weakly-bound and tightly-bound electrons depend on their binding energy relative to the Compton energy shift



- For a 25 eV, $4\text{x}10^{23}$ cm^{-3} plasma the XFEL produces10^{4} photons from the free electron scattering

- Can obtain temperatures, densities, mean ionization, velocity distribution from the scattering signal

LCLS Experimental Hall



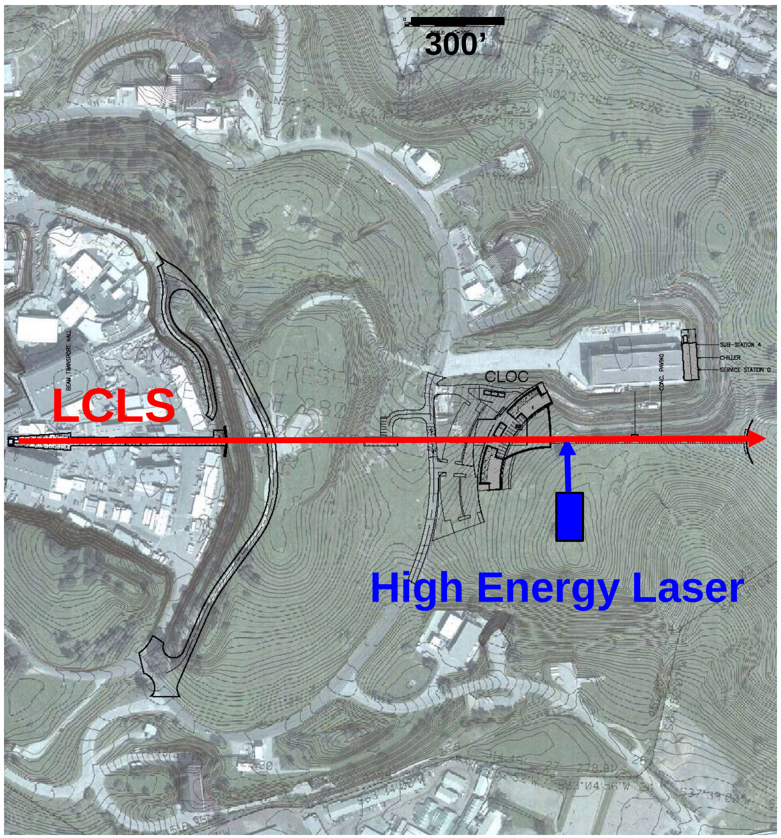
Proposal for HEDS end station endorsed by LCLS Science Advisory Committee

- Meet needs of an international HEDS community
  - e.g., high pressure and plasma physics
  - many facets of HDM/WDM research possible

- Current efforts to solicit support
  - range of users from universities to national labs

Placement of a high-energy laser adjacent to x-ray FEL and target area

Top View of SLAC site

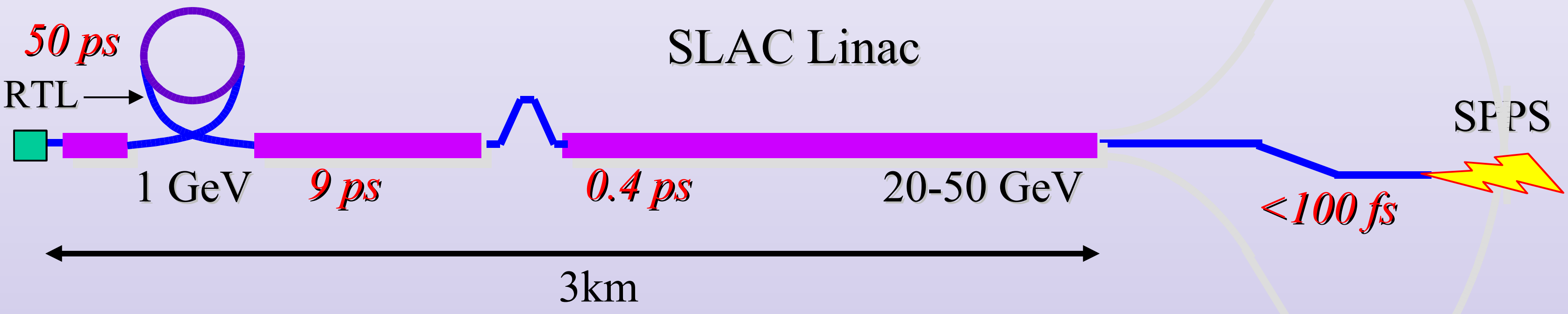


Characteristics of High Energy Laser

LCLS HEDS end station proposal proposal
(courtesy of Dick Lee)

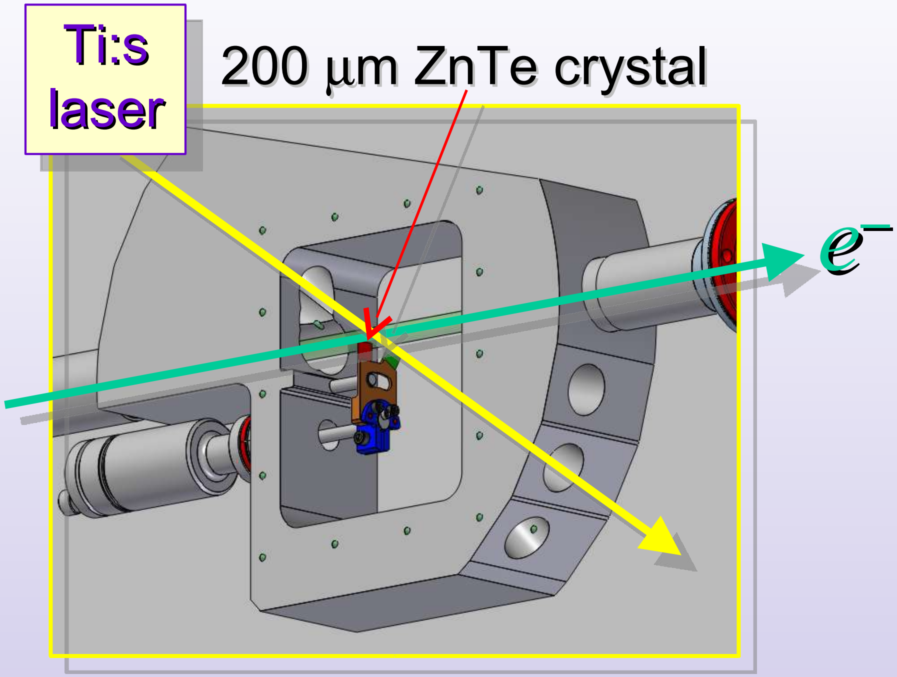
Experiment	Description	PARTICIPANTS
Warm Dense Matter Creation	Using the XFEL to uniformly warm solid density samples	H.-K. Chung, S. Glenzer, G. Gregori, S. Moon, O. Landen. K. Widmann, P. Young, M. Murillo, J. Benage, A. Lindenberg, A. Correa, R. Falcone, W. Nellis, W. Rozmus, A. Ng, T, Ao, J. Wark, J. Sheppard, R. Redmer, D. Schneider, F. Rosmej
Equation of State	Heat / probe a solid with an XFEL to provide material properties	K. Widmann, K. Budil, G. Collins, S. Glenzer, G. Gregori, M. Koenig, A. Bennuzi, A. Nelson, O. Landen, W. Nellis, A. Ng, P. Young, J. Benage, M. Taccetti, S. Rose, D. Schneider
Absorption Spectroscopy	Heat a solid with an optical laser or XFEL and XFEL to probe	P. Heimann, S. Johnson, R. Lee, S. Tzortzakis, S.Bastiani-Ceccoti, C. Chenais, P. Audebert, F. Rosmej, R. Falcone, R. Schuch, A. Lindenberg, D. Chambers, J. Wark, S. Rose
Shock Phenomena	Create shocks with a high-energy lasers and probe with the XFEL	G. Collins, H. Lorenzana, J. Belak, P. Celliers, C.-S. Yoo, K. Budil, M. Koenig, A. Bennuzi, S. Clark, P. Heimann, R. Jeanloz, P. Alivisatos, R. Falcone, W. Nellis, A. Ng, T. Ao
Surface Studies	Probe ablation/damage process to study structural changes and disintegration	A. Nelson, J. Kuba, A. Andrejczuk, J. B. Pelka, J. Krzywinski, R. Sobierajski, K. Sokolowski-Tinten, L. Juha, M. Bittner, J. Krasna, T. E. Glover
XFEL / Gas Interaction	Create exotic, long-lived highly perturbed electron distribution functions in dense plasmas	R. London, S. Hau-Riege, P. Young, H. K. Chung, W. Rozmus, R. Fedosejev, H. Baldis, V. N. Shlyaptsev, T. Ditmire, H. Fiedorowicz, M. Fajardo, A. Bartnik, F. Dorchies, J.-C. Gauthier, P. Audebert, V. Bychenkov, D. van der Spoel, C. Caleman, T. Möller, T. Tschentscher, H. Merdji
XFEL / Solid Interaction	Use XFEL directly to create extreme states of matter at high temperature and density	S. Glenzer, K. Budil, H.K. Chung, J, Dunn, S. Hau-Riege, R. London, K. Sokolowski-Tinten, J. Krzywinski, H. Fiedorowicz, A. Bartnik, V. Letal, K. Rohlena, K. Eidmann, D. Chambers, N. Woolsey, A. Andrejczuk, F. Dorchies, J. Gauthier, M. Fajardo, J. Dias, N. Lopes, G. Figueira, M. Bergh, T. Tschentscher
Plasma Spectroscopy	Use XFEL as a pump/probe for excited bound state populations	R. W. Lee, M. Foord, H.K. Chung, D. Riley, F. Y. Khattak, E. Förster, F. Dorchies, J.-C. Gauthier, S. Tzortzakis, S.Bastiani-Ceccoti, C. Chenais-Popovics, P. Audebert, S. Rose, J. Wark, N. Woolsey, R. Schuch, K. Eidmann, F. Rosmej, S. Ferri
Diagnostic Development	Develop Thomson scattering, SAXS, interferometry, and radiographic imaging	S. Glenzer, G. Gregori, R. Bionta, H. Baldis, P. Heimann, H. Padmore, U. Bergmann, H. Merdji, P. Zeitoun, J. Seely, E. Förster

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

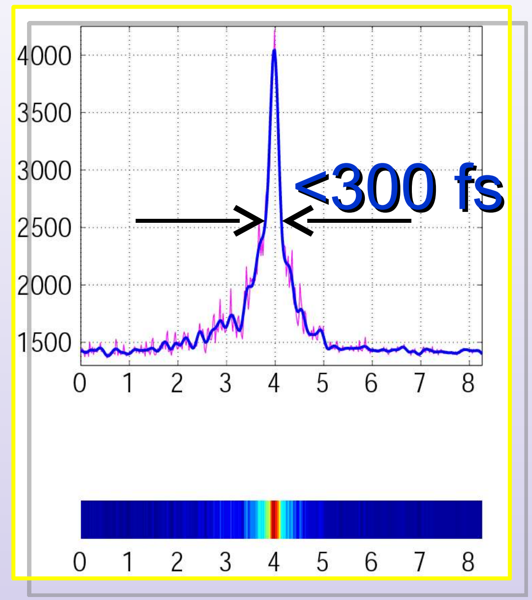


- **Electron bunches generated now at SPPS are 80 fs in duration, comparable to the bunches that will drive future x-ray FELs such as LCLS**
- **A 2m undulator delivers 80 fs duration hard x-ray pulses**

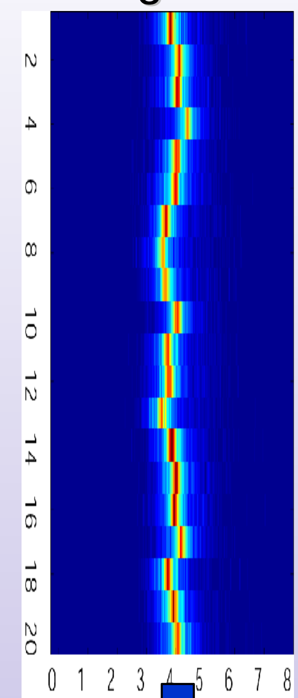
Electro-Optical Sampling for timing at the SPPS



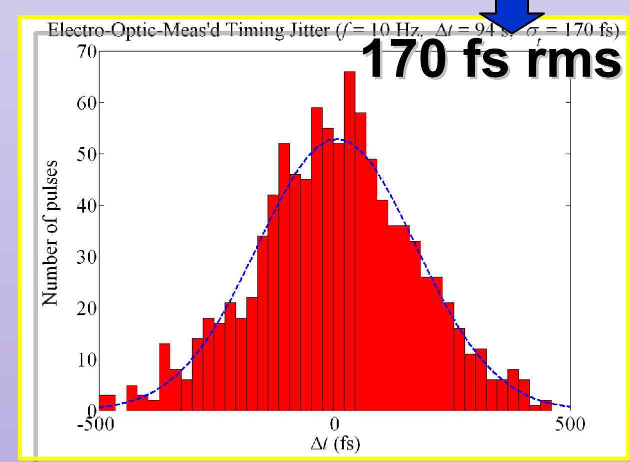
Single-Shot



Timing Jitter



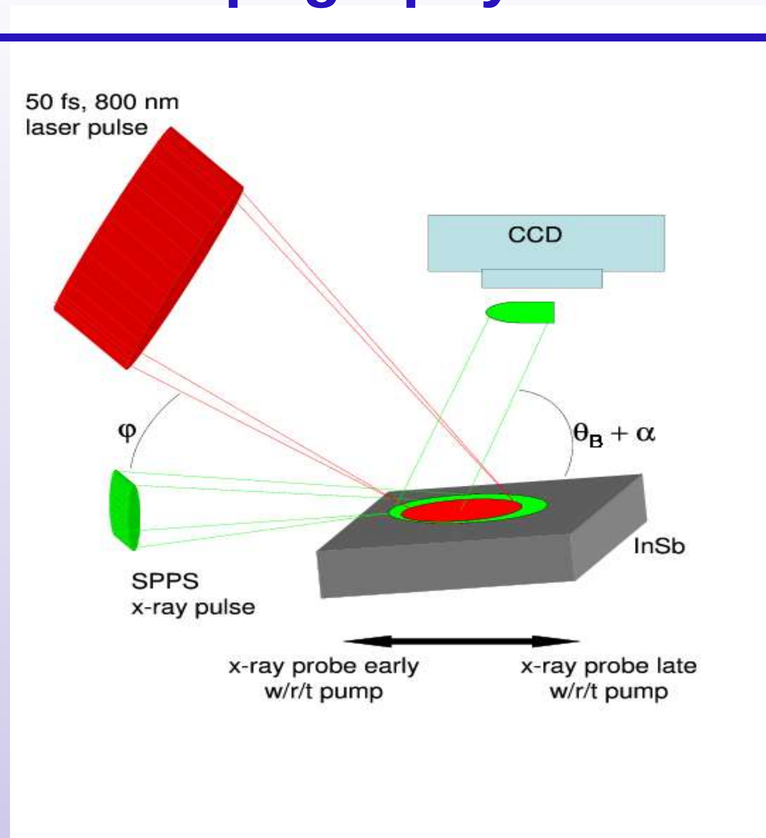
e^- temporal information is encoded on transverse profile of laser beam

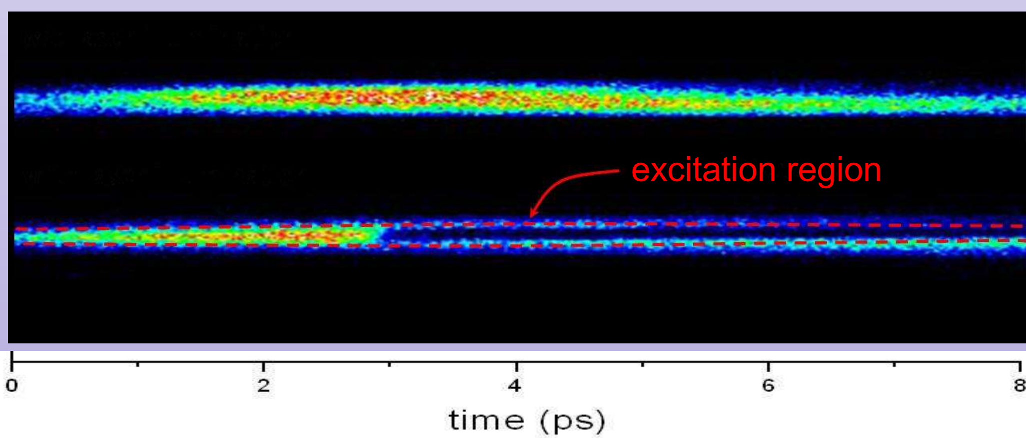


Adrian Cavalieri et al., *U. Mich.*

Sub-Picosecond Pulse Source

Crossed-Beam Topography for timing at the SPPS

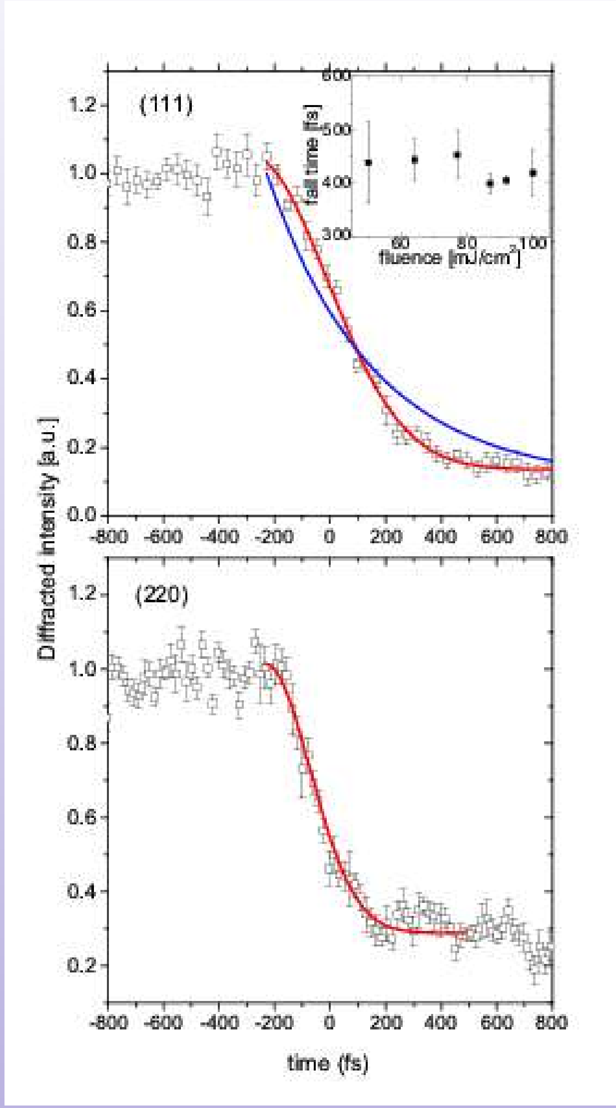




- Crossed-beam technique transforms temporal information into spatial information.
- Measures complete time history in a single shot.
- Position of edge indicates x-ray/laser timing

Sub-Picosecond Pulse Source

InSb melting: (111) vs. (220) Reflection



•X-ray probe depth is the same for the two reflections (~50 nm) Measurements conducted under identical laser excitation conditions.

$$\frac{\tau_{(111)}}{\tau_{(220)}} = 1.6 \pm 0.2 = \frac{G_{(220)}}{G_{(111)}}$$

$$\sqrt{2^2+2^2+0^2}/\sqrt{1^2+1^2+1^2} = \sqrt{\frac{8}{3}}$$

Jitter between SPPS X-Ray Source and Ultrafast Laser

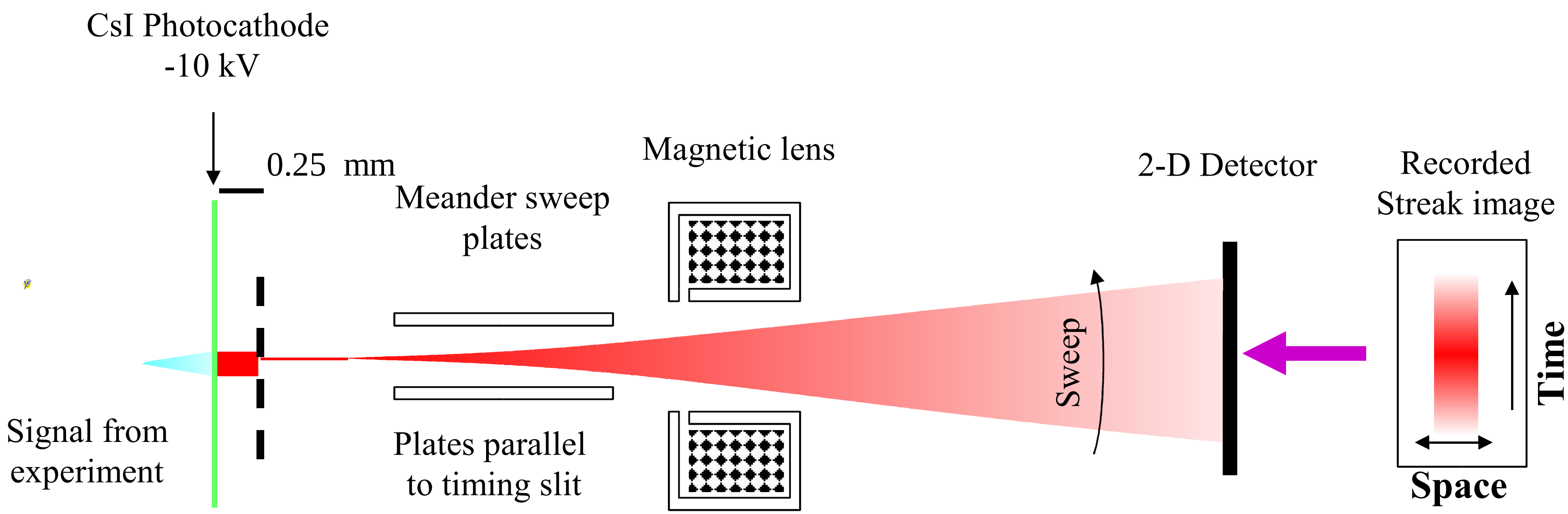
As Measured by Streak Camera

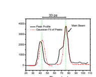
Laser fiducial: dual pulses separated by 6 ps

SPPS x-ray pulse

time

Ultrafast x-ray streak camera detectors can also enable time-resolved x-ray science

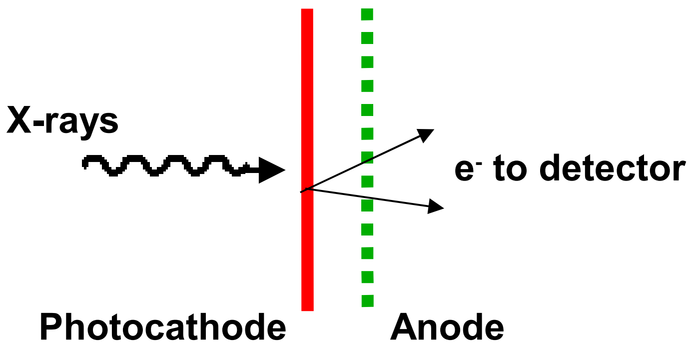


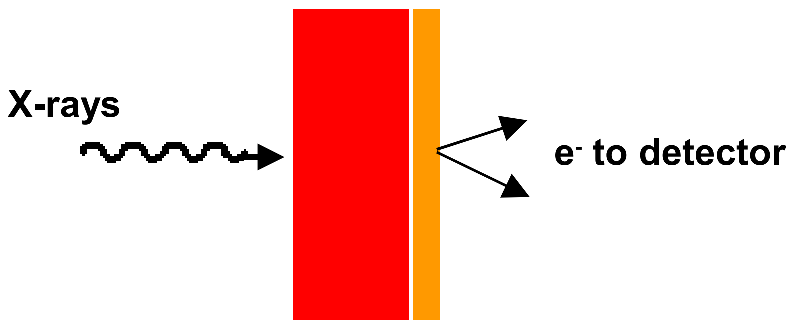


typical detection quantum efficiency
is low
and timing jitter limits temporal resolution
to a few ps

QE in Normal Incidence vs. Grazing Incidence

Transmission Photocathode

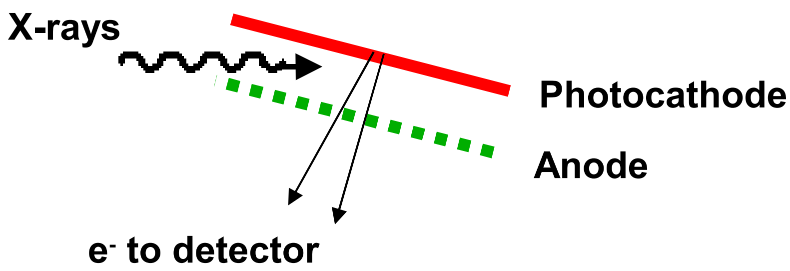


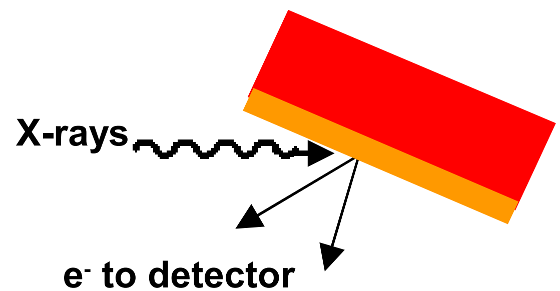


Thickness optimization

QE ~ 0.1 - 10 %

Reflection Photocathode

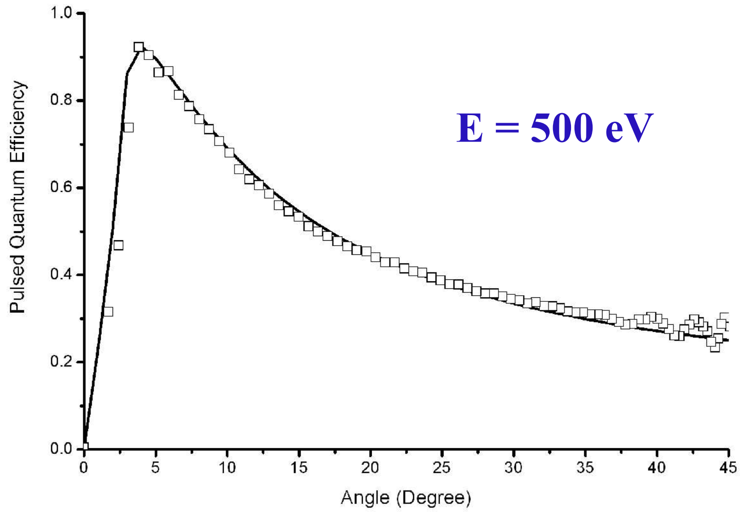


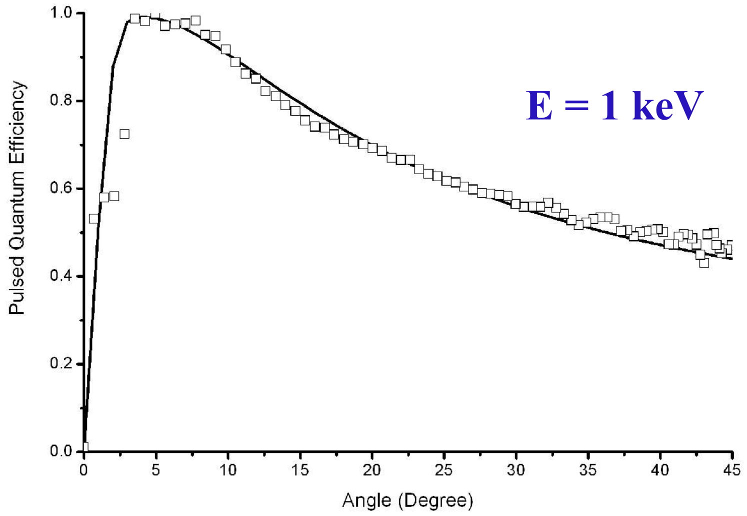


Match X-ray penetration and secondary electron escape depths

QE ~ 100 %

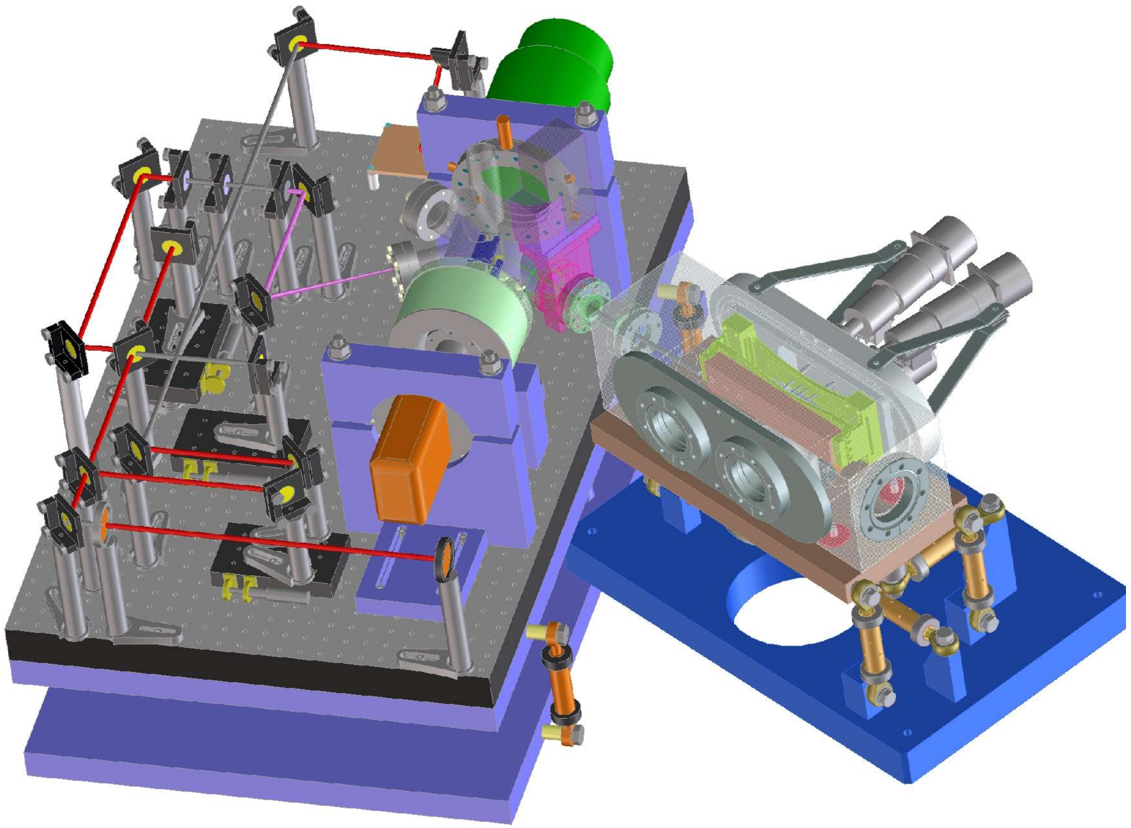
Reflection photocathodes demonstrated to have high quantum efficiency at grazing incidence





- 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.

Ultrafast Streak Camera Detectors



Can we achieve:

- **100 fs temporal resolution in the x-ray spectrum?**
- **100 % quantum efficiency / single photon detection?**

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

- Storage rings provide ~ 100-ps duration pulses (perhaps few ps 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
  (photons/pulse/BW = **10^{12}** w/FEL vs. **10^{7}** w/spontaneous vs. **10^{3}** w/slicing at 100 fs)
  - 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

ALS BL 6 Collaboration

R. Abela
T.K. Allison
A. Belkacem
C. Bressler
A. Cavalleri
M. Chergui
A. Correa
R.W. Falcone
T.E. Glover
C.M. Greaves
P.A. Heimann
M. Hertlein
S.L. Johnson
M. Kaiser
J. Larsson
R.W. Lee
A.M. Lindenberg
D. Lowney
A.G. MacPhee
T. Mathews
F. van Mourik
M. Saes
R.W. Schoenlein
H.A. Padmore
J.S. Wark
D. Weinstein
A.A. Zholents
M.S. Zolotorev

SPPS Collaboration

LCLS Collaboration

Five science teams