

An Overview of High Energy Density Science on Free-electron Lasers

P. Audebert, J. Benage, M. Bergh, C. Caleman, R. Cauble, P. Celliers, M.H. Chen, H.K. Chung, G. Collins, M. Fajardo, R. W. Falcone, R. Fedosejevs, E. Foerster, J. Gauthier, S. Glenzer, E. Glover, G. Gregori, J. Hajdu, P. Heimann, S. L. Johnson, L. Juha, F. Y. Khattak, J. Krzywinski, H.-J. Lee, R. W. Lee, S. Moon, T. Möller, W.L. Morgan, M. Murillo, B. Nagler, A. Nelson, A. Ng, Y. Ralchenko, R. Redmer, D. Riley, F. Rogers, S. J. Rose, F. Rosmej, W. Rozmus, R. Schuch, H. A. Scott, T. Schenkel, D. Schneider, J. R. Seely, R. Shepherd, R. Sobierajski, K. Sokolowski-Tinten, T. Stoelker, S. Toleikis, T. Tschentscher, H. Wabnitz, J. S. Wark., S. Vinko, K. Widmann

LULI, UC Davis, LANL, Uppsala, LLNL, IST-GoLP, UC Berkeley, Jena, CELIA, LBNL, RAL, Stanford, PSI/SLS, Czech Academy, QU Belfast, Polish Academy, SLAC, MPI, TU Berlin, Kinema, NIST, Stockholm, Rostock, AWE, Marseille, Alberta, Warsaw, Essen, GSI, DESY, Oxford, LIXAM...

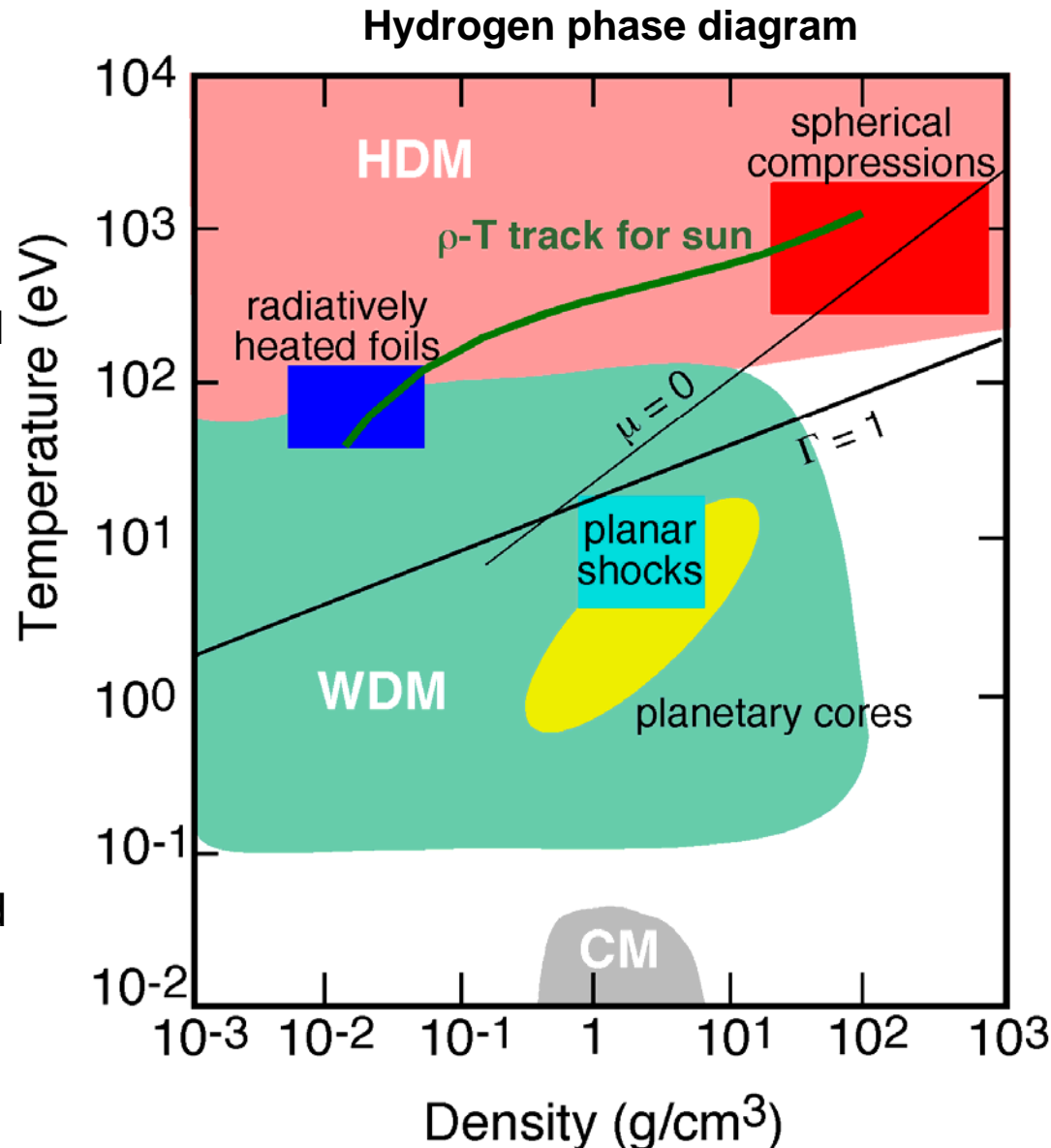
High Energy Density matter is interesting because it occurs widely

• Hot Dense Matter (HDM) occurs in:

- Supernova, stellar interiors, accretion disks
- Plasma devices: laser produced plasmas, Z-pinch
- Directly and indirectly driven inertial fusion experiments

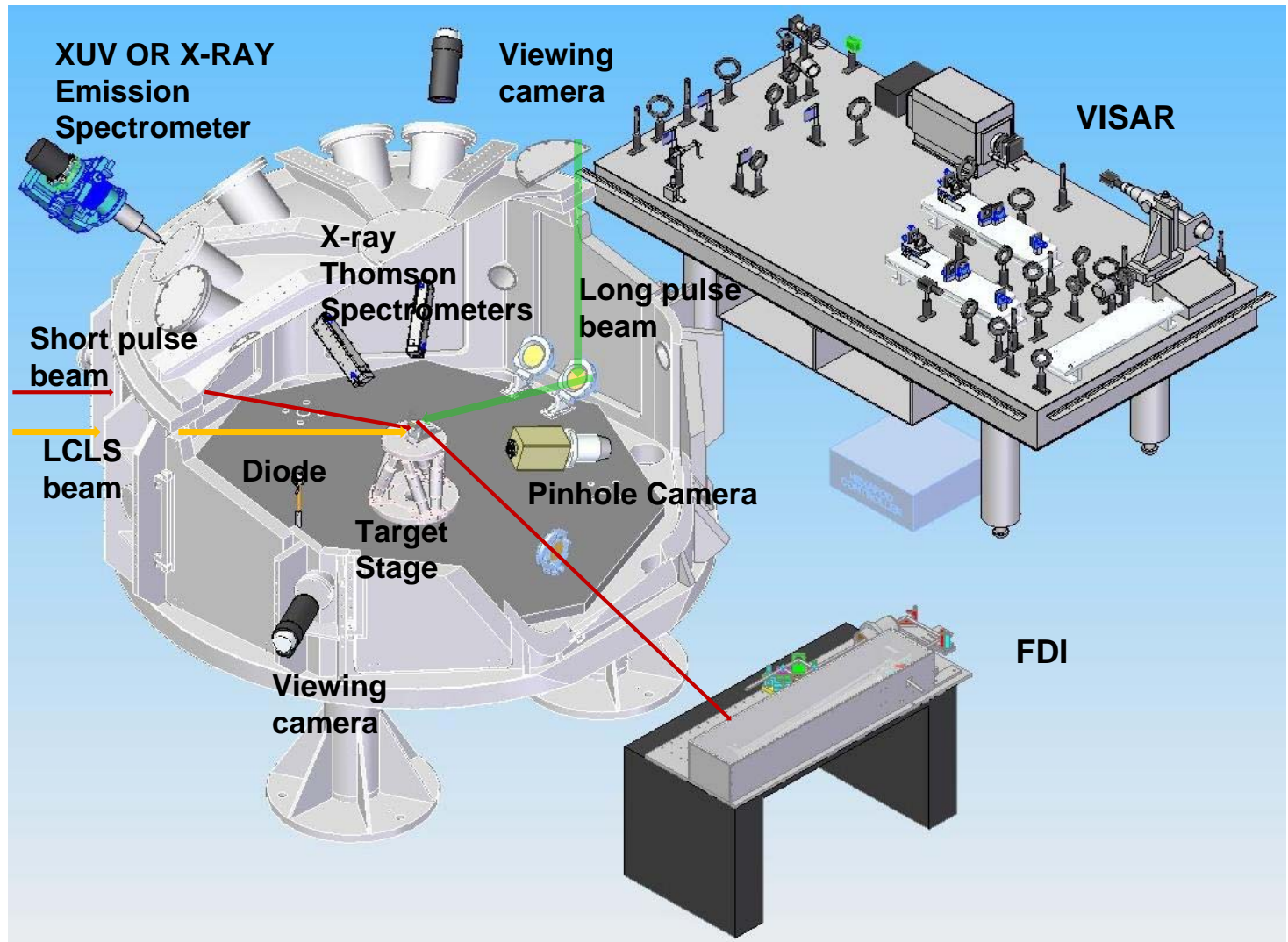
• Warm Dense Matter (WDM) occurs in:

- Cores of large planets
- Systems that start solid and end as a plasma
- X-ray driven inertial fusion experiments



Target diagnostics with target chamber: provides a full suite of instruments

- Fourier Domain Interferometer (FDI)
- VISAR system
- Thomson Scattering spectrometers
- XUV and X-ray spectrometers
- X-ray Streak camera
- Sample stage
- X-ray beam diagnostics and focusing
- Alignment system
- Transmitted beam diagnostics



• **Concept is to encourage single investigators to participate in the HED research**

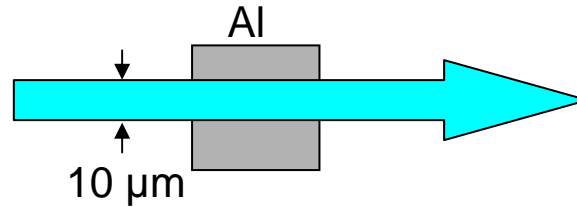
A HEDS experimental station should cover broad range of applications

Experiment	Description
Warm Dense Matter Creation	Using the XFEL to uniformly warm solid density samples
Equation of State	Heat / probe solids with XFEL to obtain material properties
Absorption Spectroscopy	Heat solids with optical laser or XFEL / use XFEL to probe
High Pressure Phenomena	Create high pressure with high-energy laser, probe with the XFEL
Surface Studies	Probe ablation/damage processes
XFEL / Gas Interaction	Create exotic, long-lived highly perturbed electron distribution functions in dense plasmas
XFEL / Solid Interaction	XFEL directly creates extreme states of matter
Plasma Spectroscopy	XFEL pump/probe for atomic state
Diagnostic Development	Develop Thomson scattering, SAXS, interferometry, and radiography

Warm Dense Matter

WDM created by isochoric heating will isentropically expand sampling phase space

- Concept is straightforward

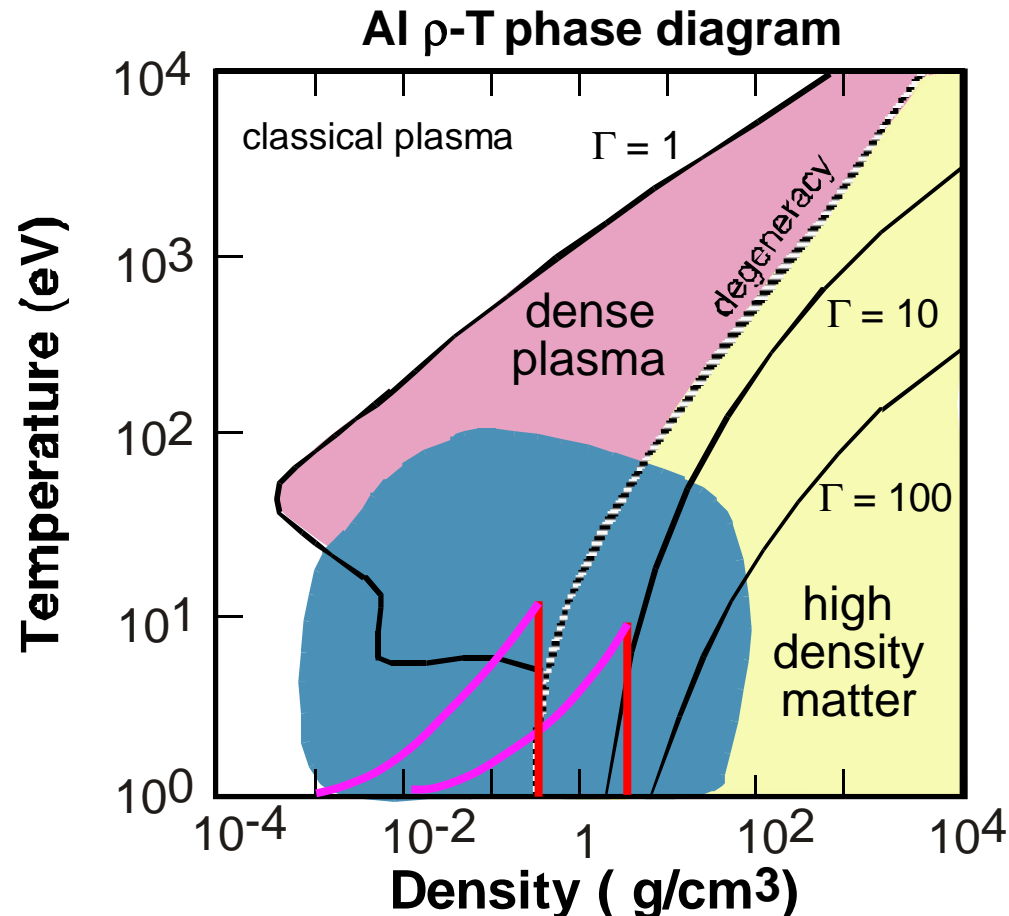


- XFEL can heat matter rapidly and uniformly to create:

- Isochores (constant ρ)
- Isentropes (constant entropy)

- Using underdense foams allows more complete sampling

- Isochores (constant ρ)
- Isentropes (constant entropy)





WDM Studies on VUV FELs

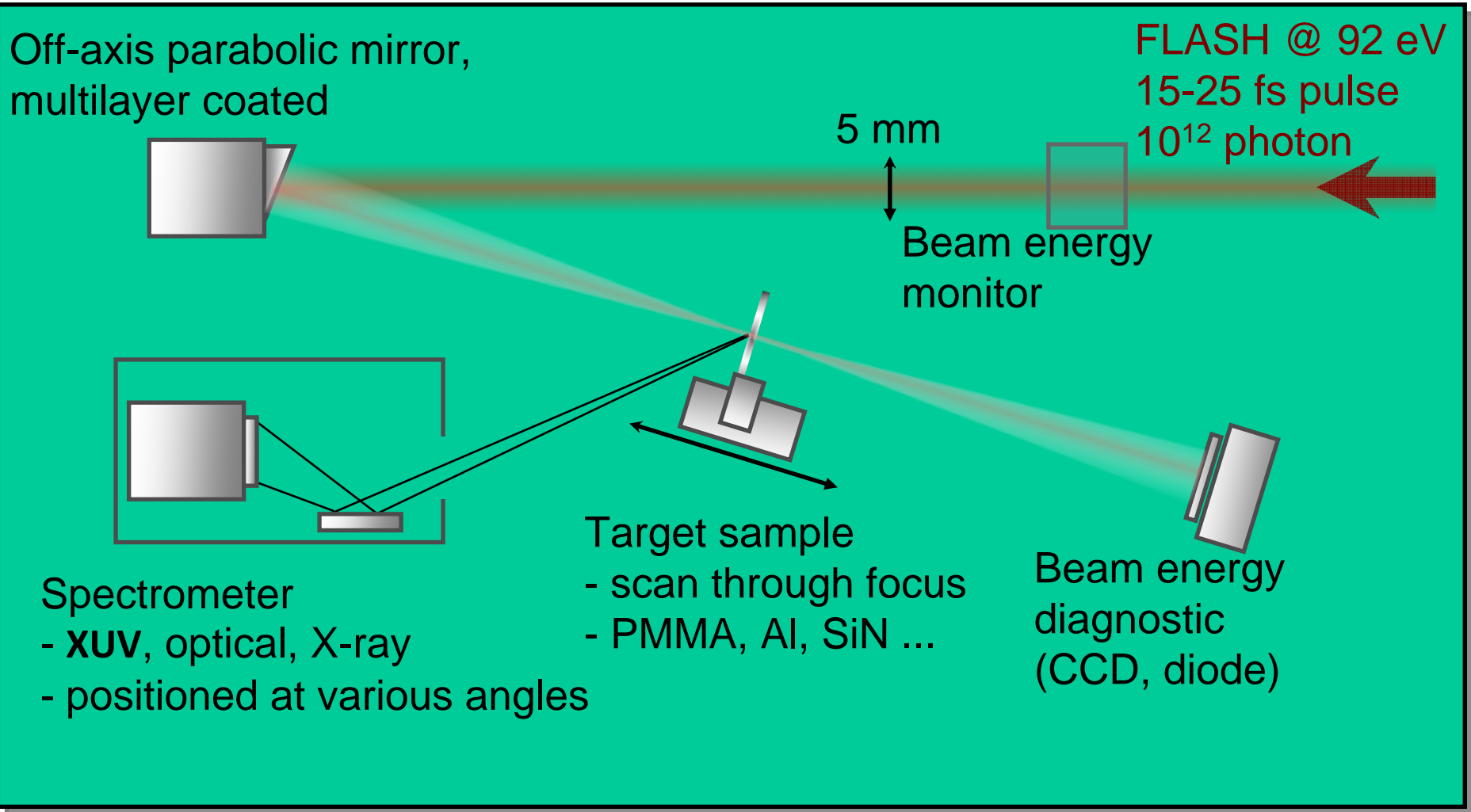
Peak Brightness Collaboration:

S.Bajt, L.F. Cao, J. Chalupsky, H. Chapman, J. Cihelka, H.-K. Chung, T. Döppner, S. Dusterer, T. Dzelzainis, M. Fajardo, C. Fortmann, E. Galtier, S. Glenzer, G. Gregori, P. Hajkova, P. Heimann, L. Juha, M. Jurek, R. Faüstlin, F. Khatkhat, M. Kozlova, J. Krzywinski, T. Laarmann, H.J. Lee, R. W. Lee, R. Levesque, P. Mercere, B. Nagler, A. Nelson, A. Przystawik, P. Radcliffe, H. Reinholz, D. Riley, G. Ropke, F. Rosmej, J. K. Seksl, R. Sobierajski, R. Thiele, T. Tiggesbaumker, S. Toleikis, N.X. Truong, T. Tschentscher, I. Uschmann, S. Vinko, J. Wark, T. Whitcher, A. Wierling, E. Forster, R. Redmer, U. Zastra

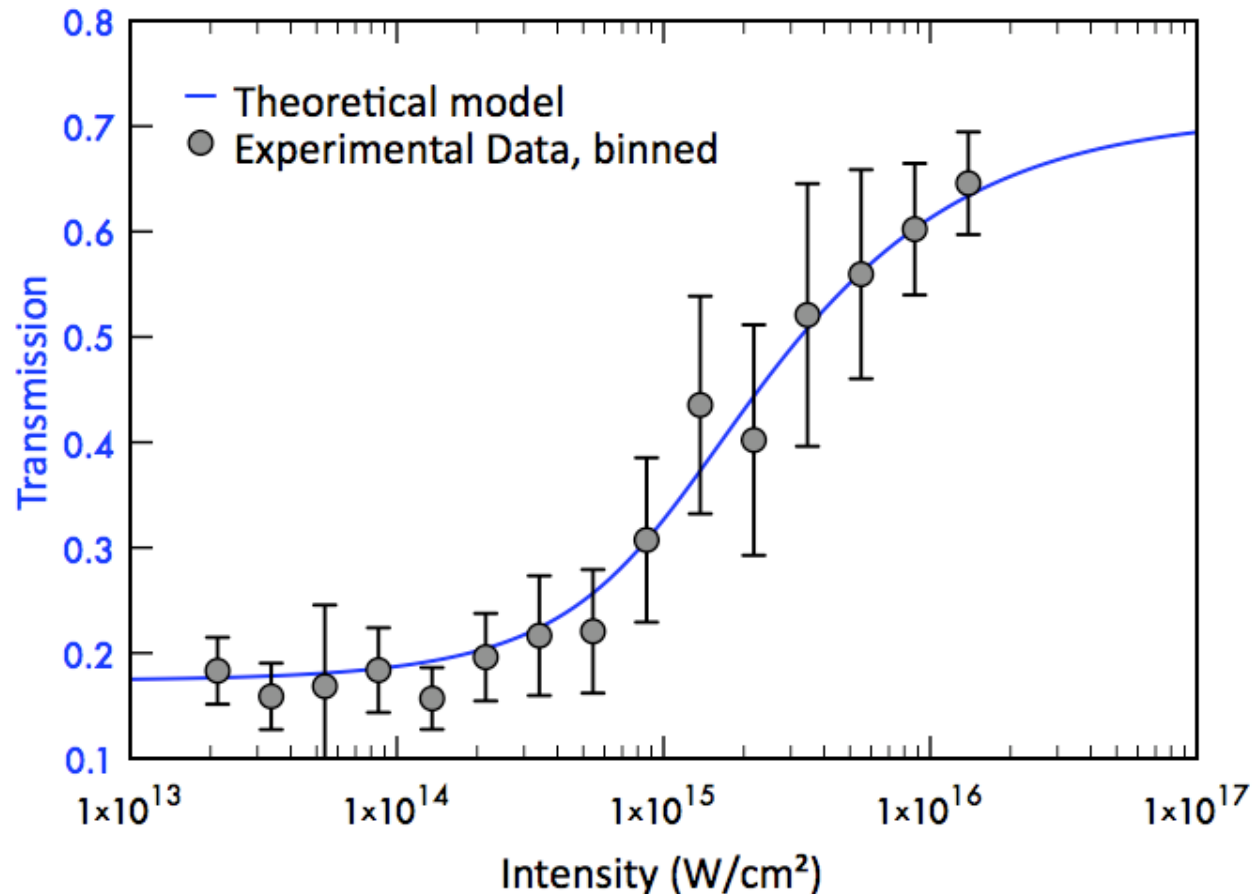
MEC High Energy Density Science Endstation at LCLS:

P. Audebert, R. Boyce, K.S. Budil, R.C. Cauble, J. Dunn, R.W. Falcone, S.H. Glenzer, J.B. Hastings, P. A. Heimann, J. Krzywinski, R.W. Lee, H.J. Lee, D. Montgomery, B. Nagler, R. Shepherd, T. Tschentscher, J.S. Wark, W. E. White, K. Widmann

1st FLASH WDM experiment use 92eV beam measure transmission and spectra

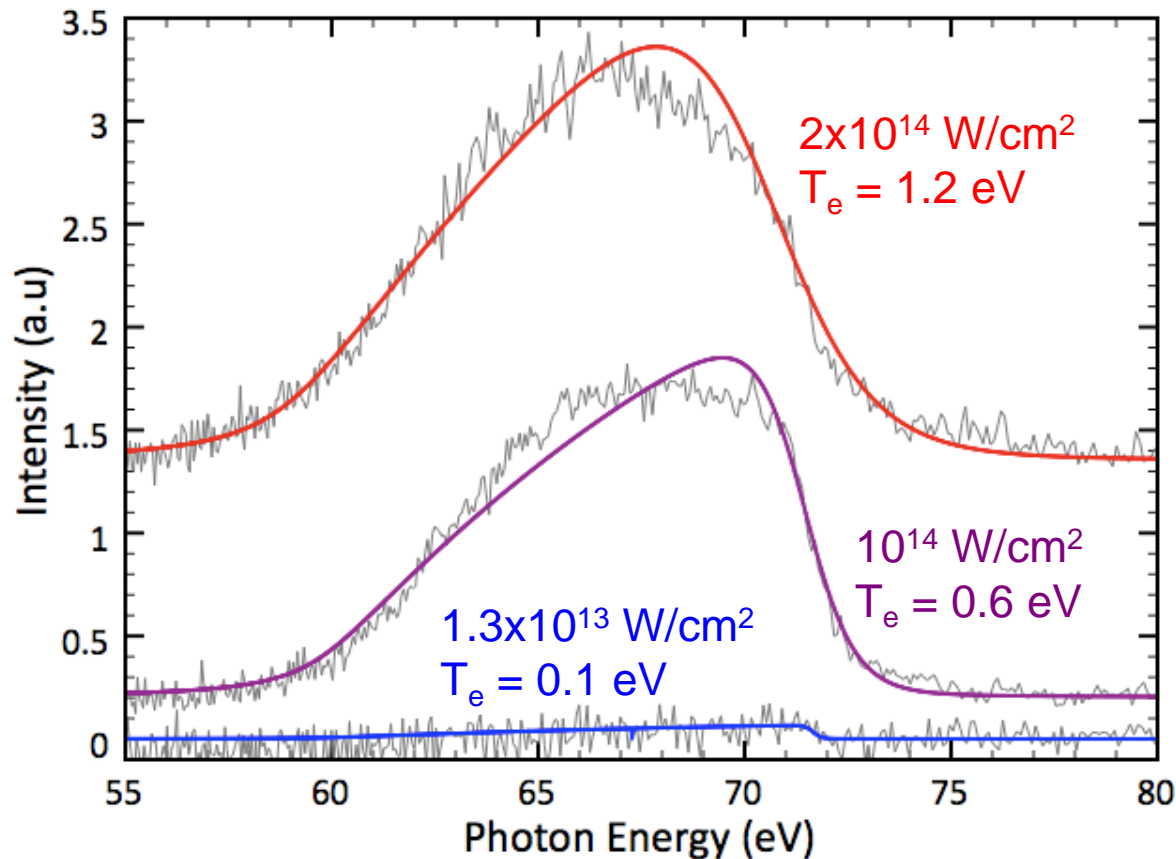


Creating transparent aluminum



- 52 nm Al foil (+20 nm oxide layer) transmitting 92 eV photons
- Best focus $\sim 1\mu\text{m}$, $>10^{16}$ W/cm² intensity
- Effect takes place on 15-25 fs time scale - no ion movement!
- Nagler et al., Nature Physics **5**, 693 (2009)

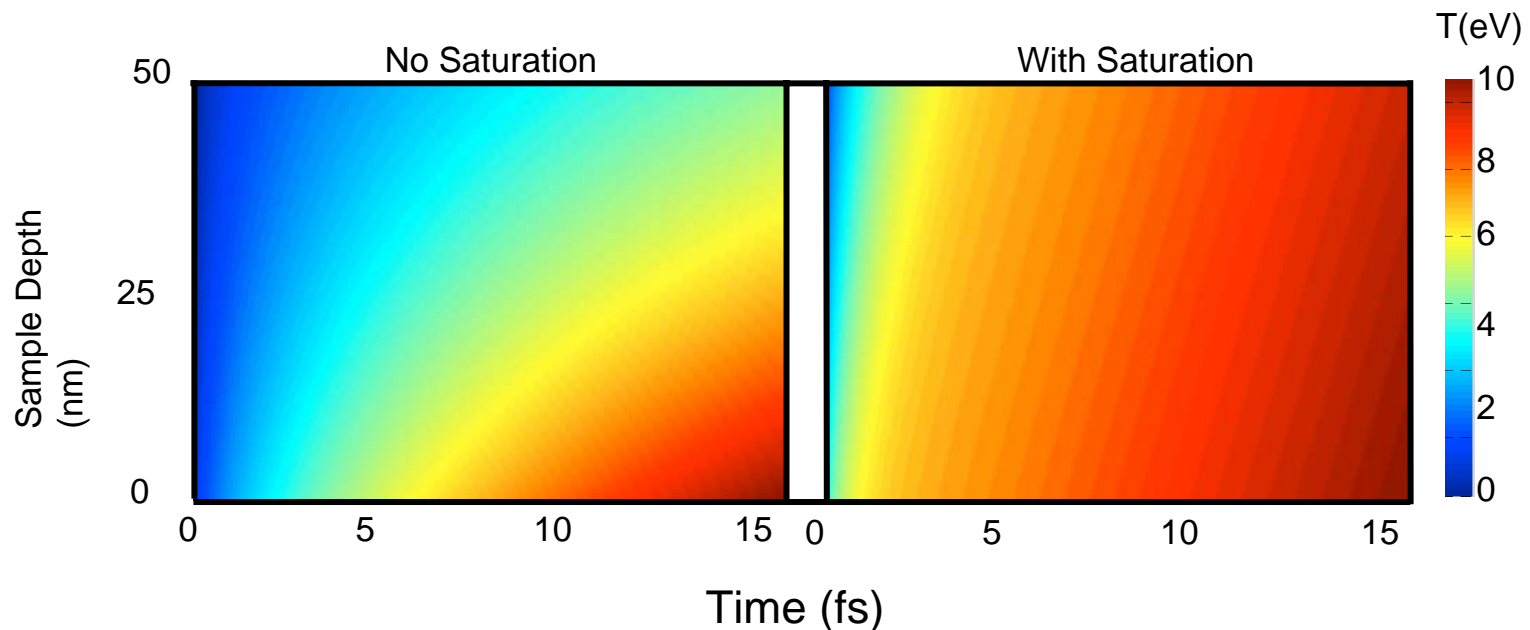
Soft x-ray emission spectroscopy



- Experimental data taken at $20 \mu\text{m}$ spot (low intensities)
- Model represents valence band density of states at a given temperature
- Temperatures broadly agree with spectroscopy

Efficient homogeneous WDM creation

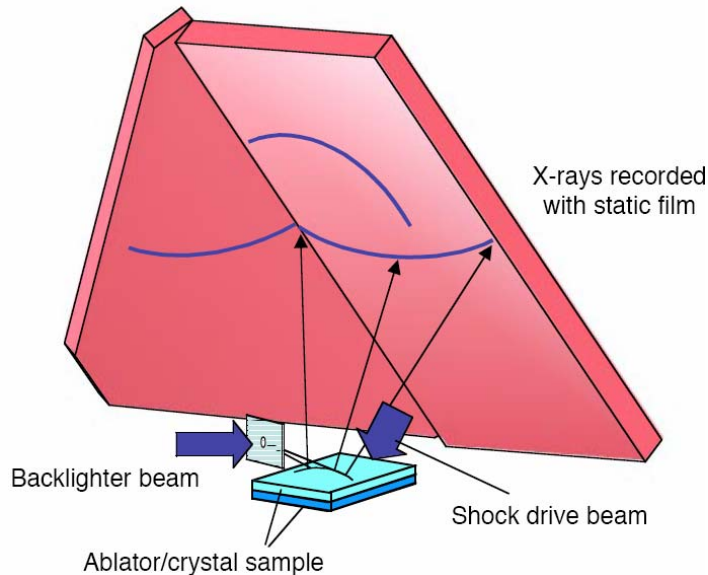
- Essential to create WDM in a well-defined state
 - very fast & homogeneous heating imperative to obtain near constant (T, ρ)
- Saturation provides an **order of magnitude more efficient** production of homogeneity



High Pressure States

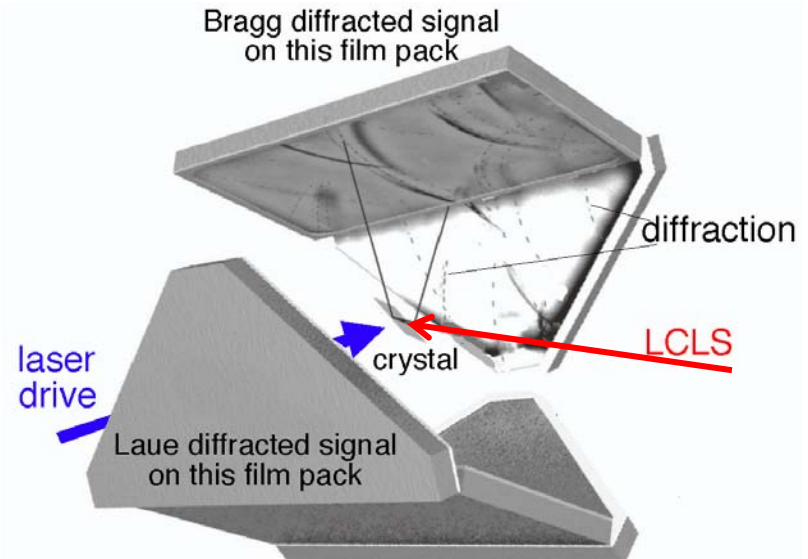
Lasers provide shocks *and* high divergence probe - LCLS provides low divergence probe

- Schematic of High Energy Laser shock experiment



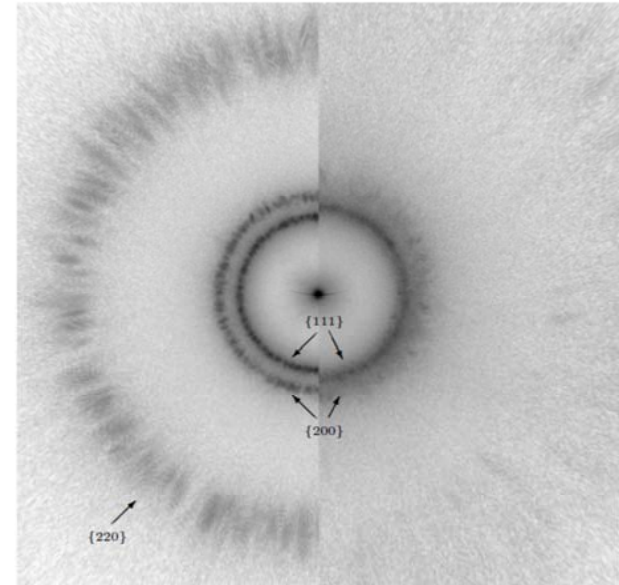
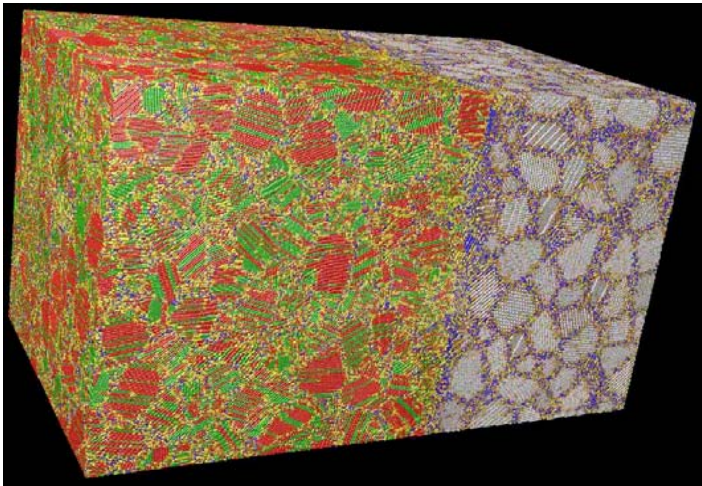
- Laser creates a shock in a **single-crystal** sample
- Delayed beams create ns-scale highly divergent x-ray source
- Angular spread of the x-ray source samples many crystal planes
- **Technique provides critical data on dynamics at high pressure**

- Schematic of **LCLS** XFEL shock experiment



- Laser creates a shock in a **polycrystalline** sample
- XFEL creates fs-scale non-divergent monochromatic source
- Grains in the polycrystal diffract the beam
- **Low Divergence \Rightarrow nm-scale fs diffraction of real solids**

1st planned LCLS shock experiment will use polycrystalline material (LLNL/Oxford)



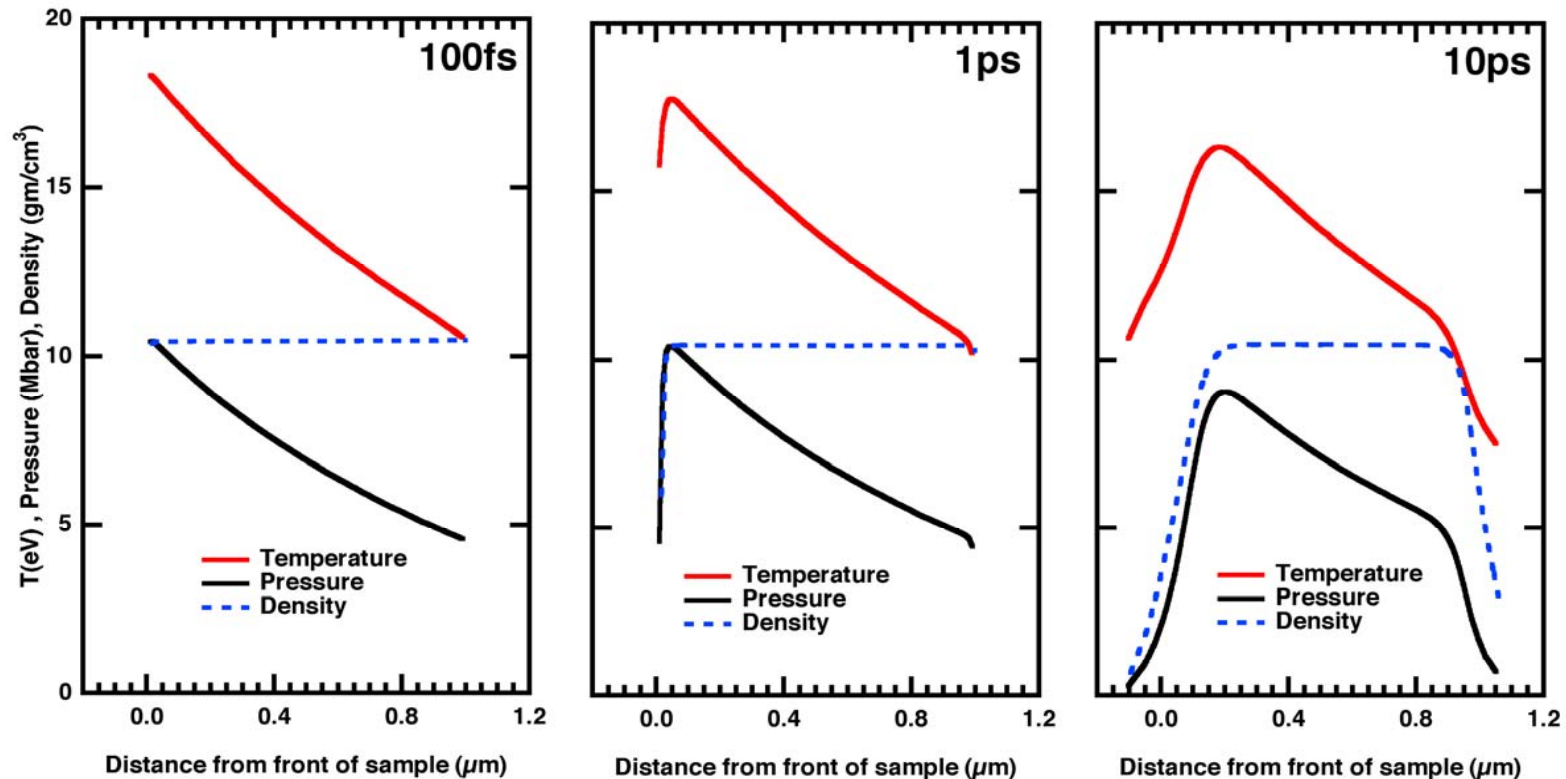
Unshocked

Shocked

- Collimated, monochromatic X-ray beam
- Polycrystalline target
- VISAR will provide additional diagnostic

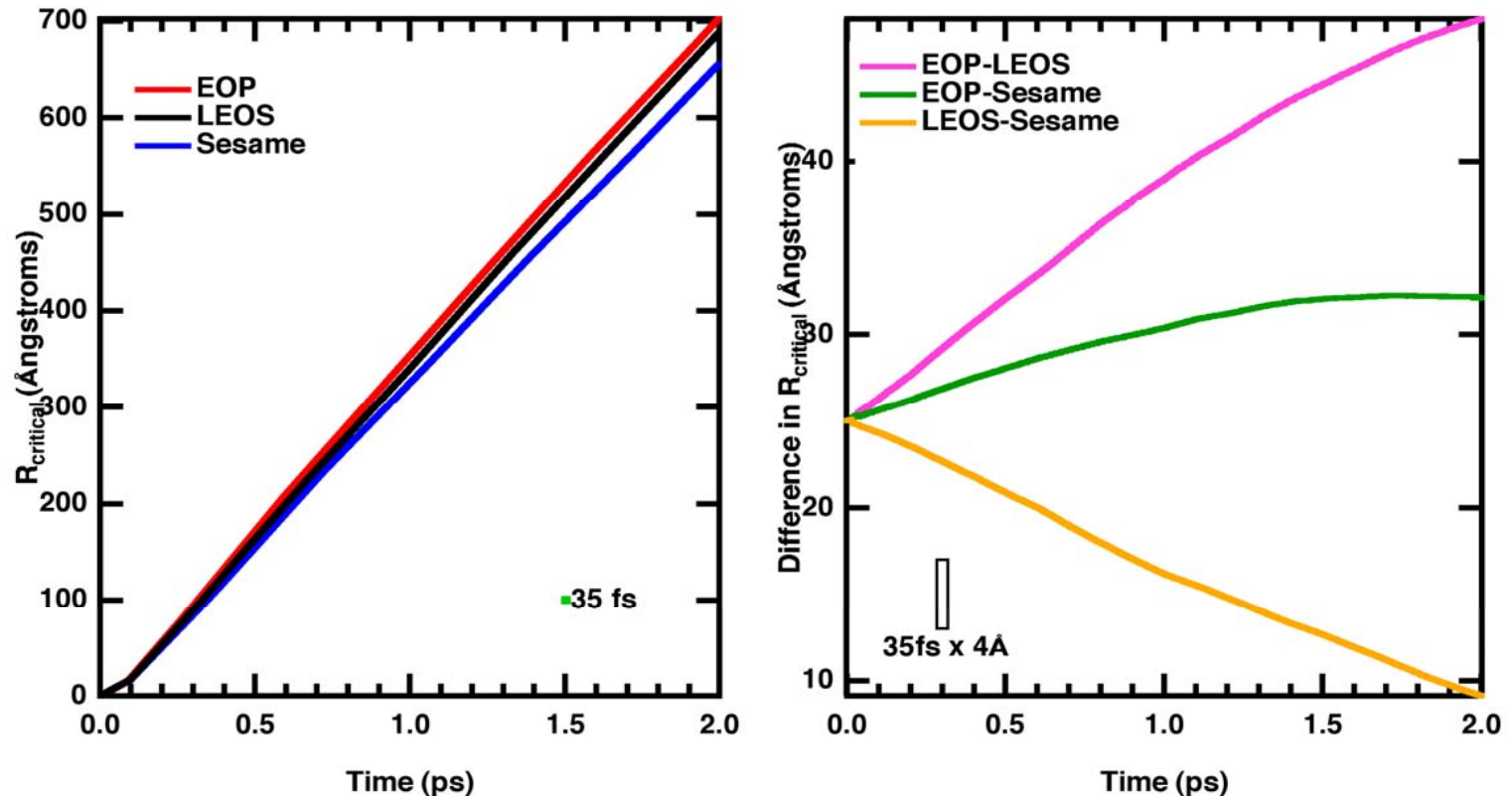
**LCLS
creates
high pressure
states**

With an initial condition the simulation predicts the 'long' time behavior



- Simulations indicate that the gradient leads to a distinct expansion on front and rear surfaces.
- Heating is isochoric; but, **not** isothermal so the system is NLTE
- Important to measure the expansion to determine if we understand global front/rear differences.

Diagnosis of Ag sample may differentiate EOS models off the principle Hugoniot



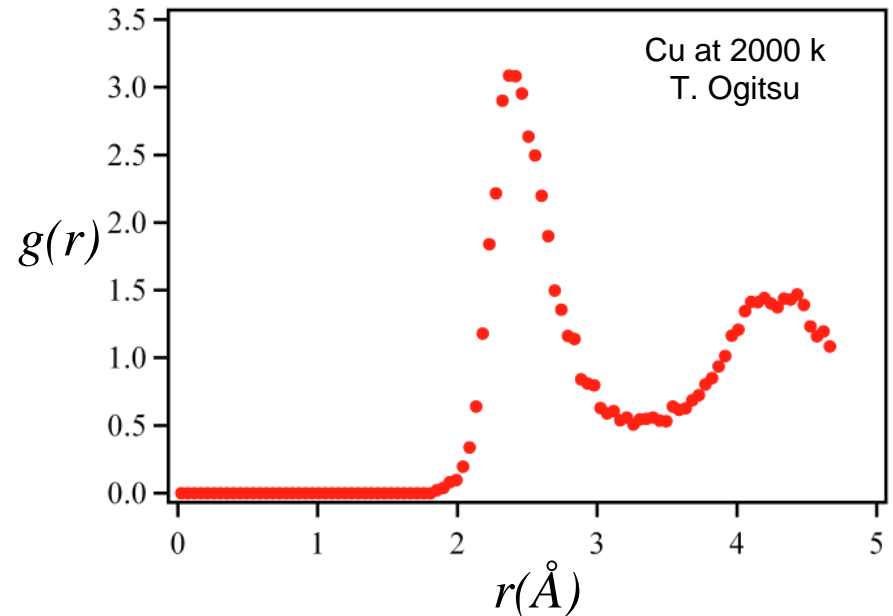
- Simulations require the EOS's which are in question
- FDI data front and rear will be sufficient to provide information on the validity of the modeling
- FDI front versus back will indicate if the degree of thermalization is consistent with simulations.

Planned LCLS experiments on WDM will
probe shocks and create high pressure state

*Next set will
extend capability*

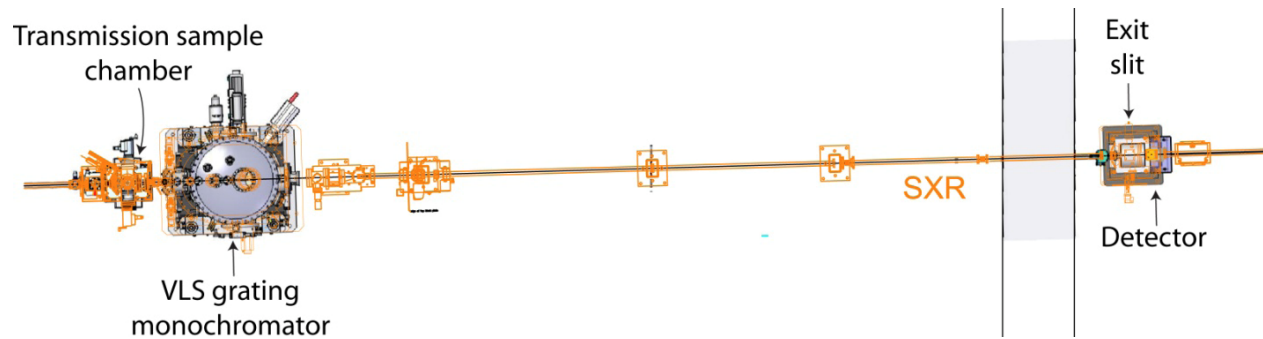
Future (1): other source creates WDM and use LCLS to measure diffuse scattering

$$g(r) = \frac{1}{2\pi^2 \rho_0 r} \int_0^\infty \frac{I(q)}{\langle F^2 \rangle} q \sin(q) r dq$$

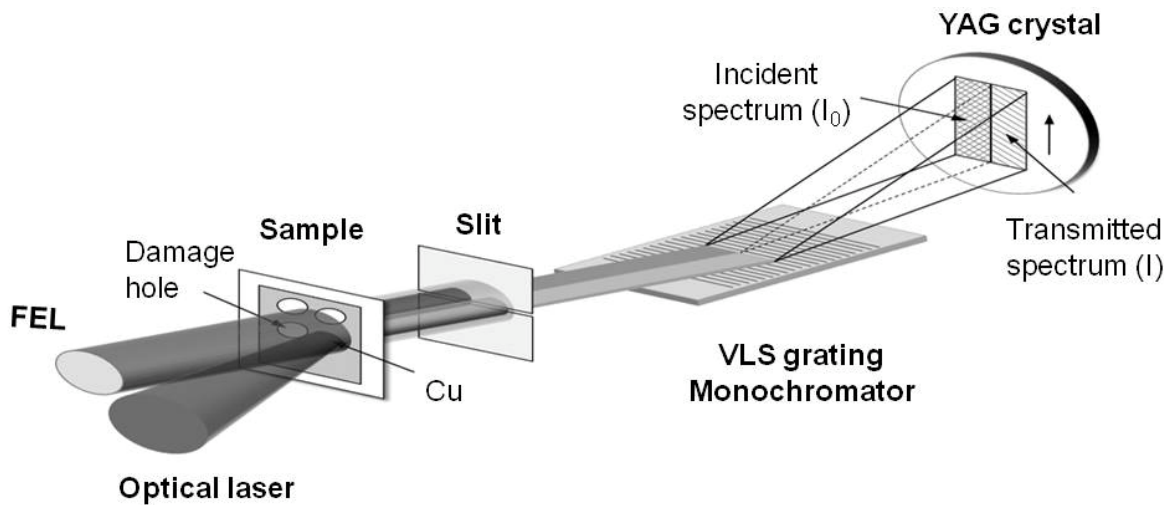


- At 10-100fs timescale LCLS can measure the diffuse scattering, which can be directly compare to the $g(r)$
- Short pulse laser warms sample and the delayed LCLS probes to measure the $g(r,t)$
- Delay set within 200fs with more precision available post mortem

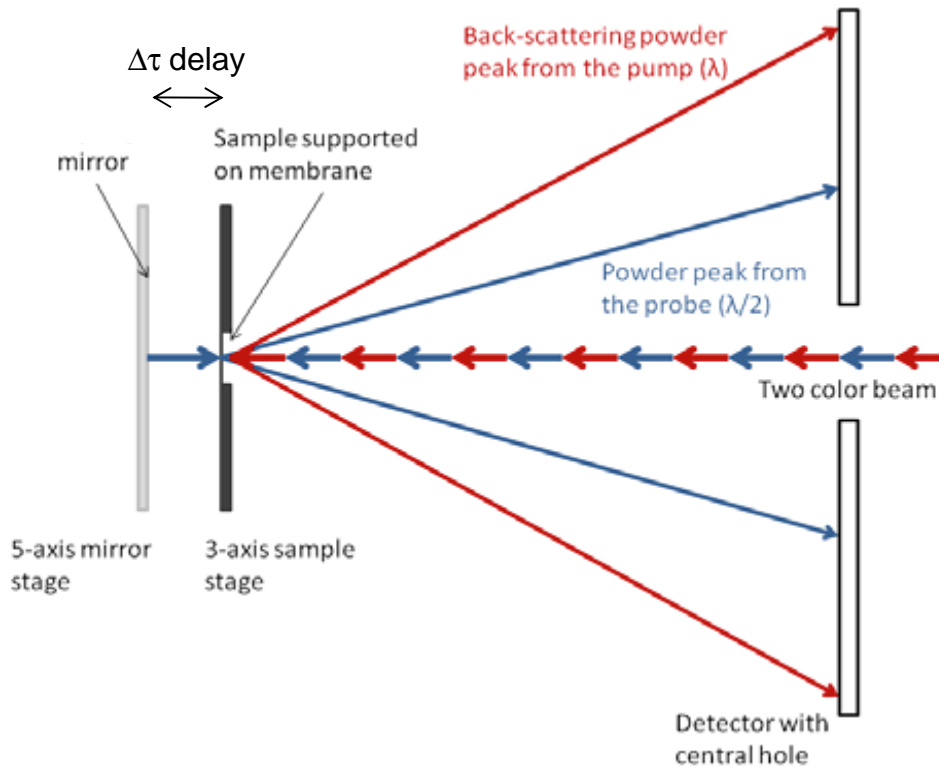
Future (2): other source creates WDM and use LCLS as a absorption probe



- LCLS provides a fs-scale absorption source
- Short pulse laser heats the sample, probed by delayed LCLS
- I_0 and $I_{\text{transmitted}}$ measured on each shot
- Provides the DOS measurement: unoccupied WDM states as the T_e becomes , occurring



Future (3): LCLS will soon propagate 1st and 2nd harmonic with intensity ratio of 10 to 1



$\Delta\tau$ varies from 5fs to 10ps

$\Delta\tau$ jitter is of order 3 fs

- Two-color beam passes through 2D detector before hitting sample
- 2nd harmonic ($\lambda/2$) is reflected back onto sample
- 2D detector allows measurement of:
 - back-scattered λ powder ring from the pump pulse and
 - $\lambda/2$ powder ring from the probe pulse time delay of $\Delta\tau$

Future (4): Measure the full $S(k, \omega)$ in carbon using Thomson Scattering

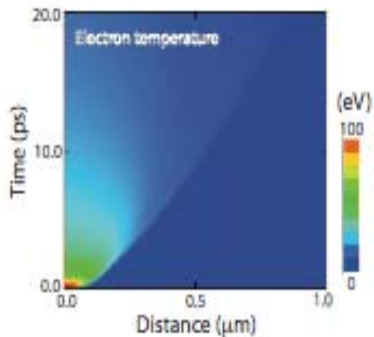
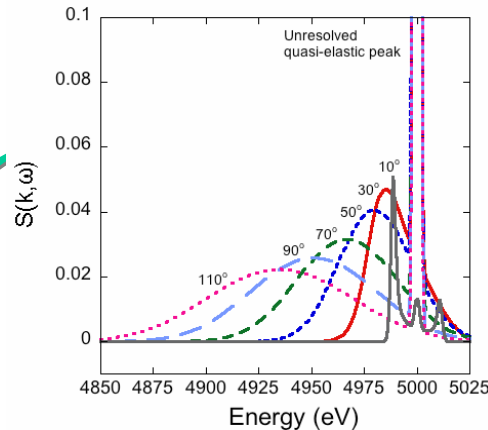
HOPG crystal Scattering

angles: $20^\circ, 35^\circ, 50^\circ, 130^\circ, 145^\circ, 160^\circ$

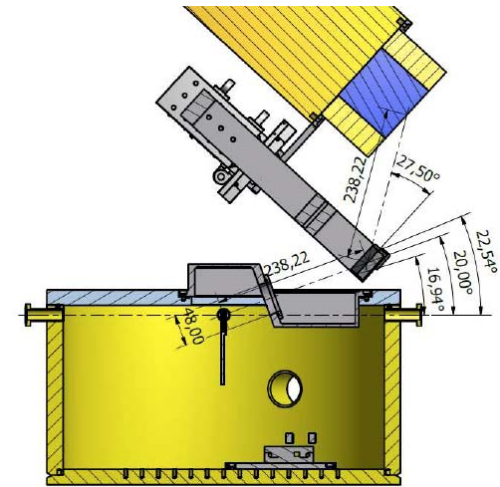
Optical laser pump

C, B₄C foils

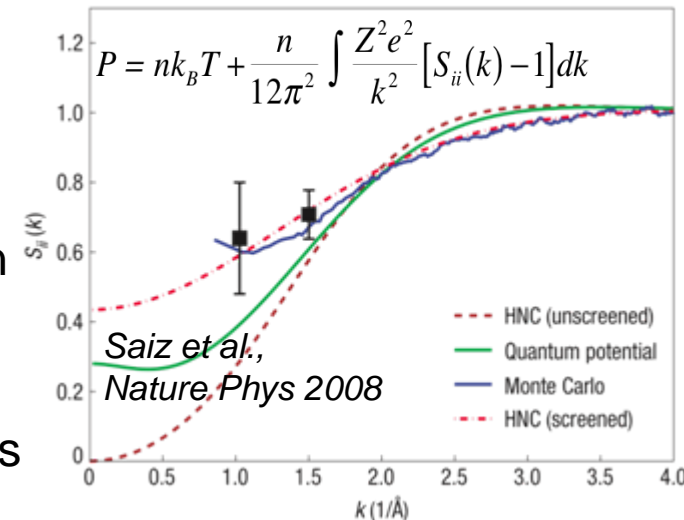
5-8 keV
FEL



- Experimental aimed at directly measure thermodynamic properties and dynamic structure factors of laser driven carbon, as it irreversibly transitions from a cold solid into a warm dense matter (WDM) state

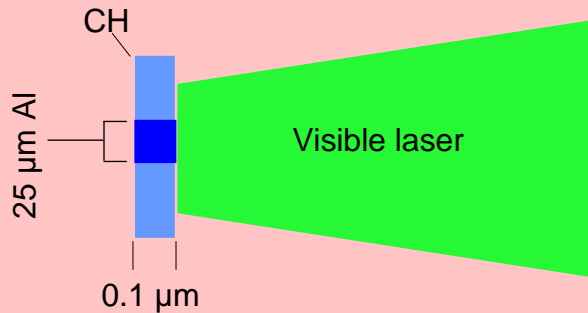


- Use optical laser to induce melting of carbon/boron-carbide samples and FEL probe at several scattering angles and pump delays
- Full dynamic structure factor is a direct representation of the many-body correlations (both e-e and i-i correlations)
- Thermodynamic properties are obtained from integrals of the structure factors

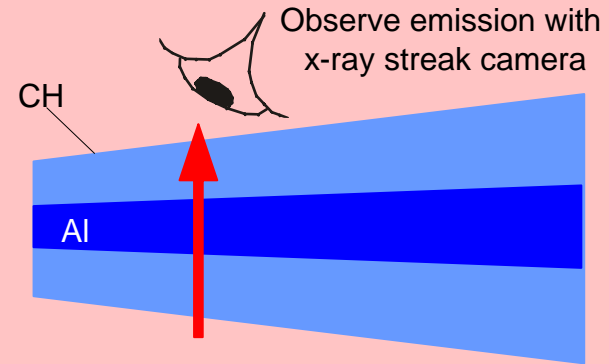


(5) In Hot Dense Matter regime will provide critical data on the population kinetics

• Schematic experiment



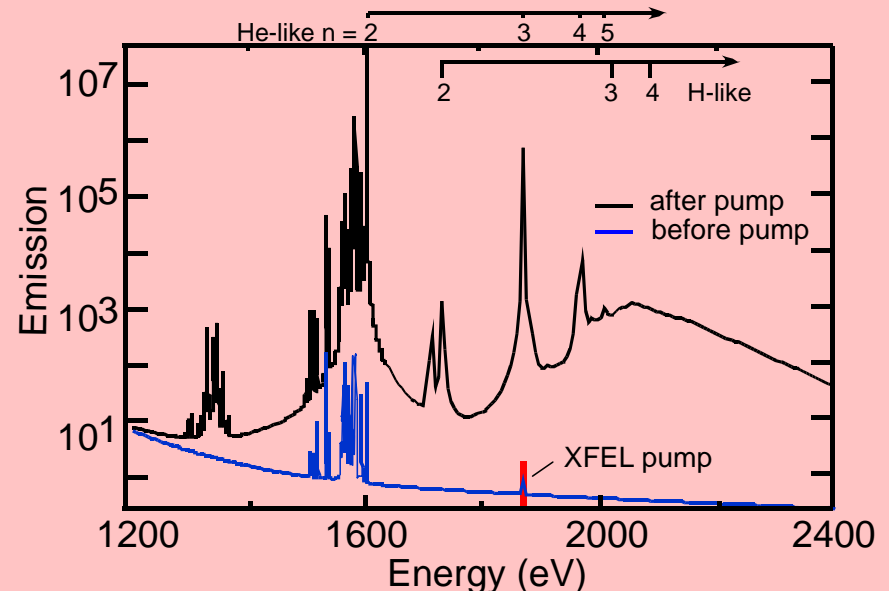
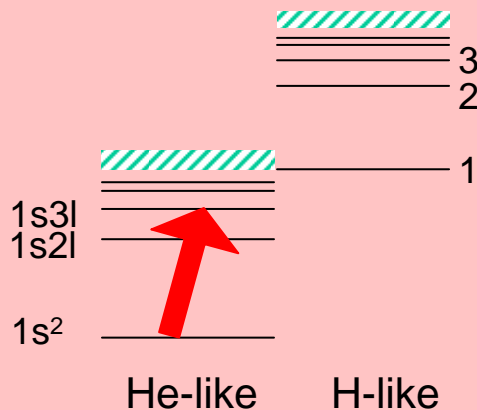
• $t = 0$ laser irradiates Al dot



FEL tuned to 1869 eV

• $t = 100 \text{ ps}$ FEL irradiates plasma

• Simulation

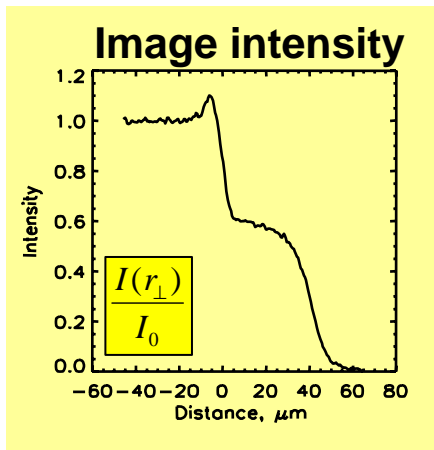
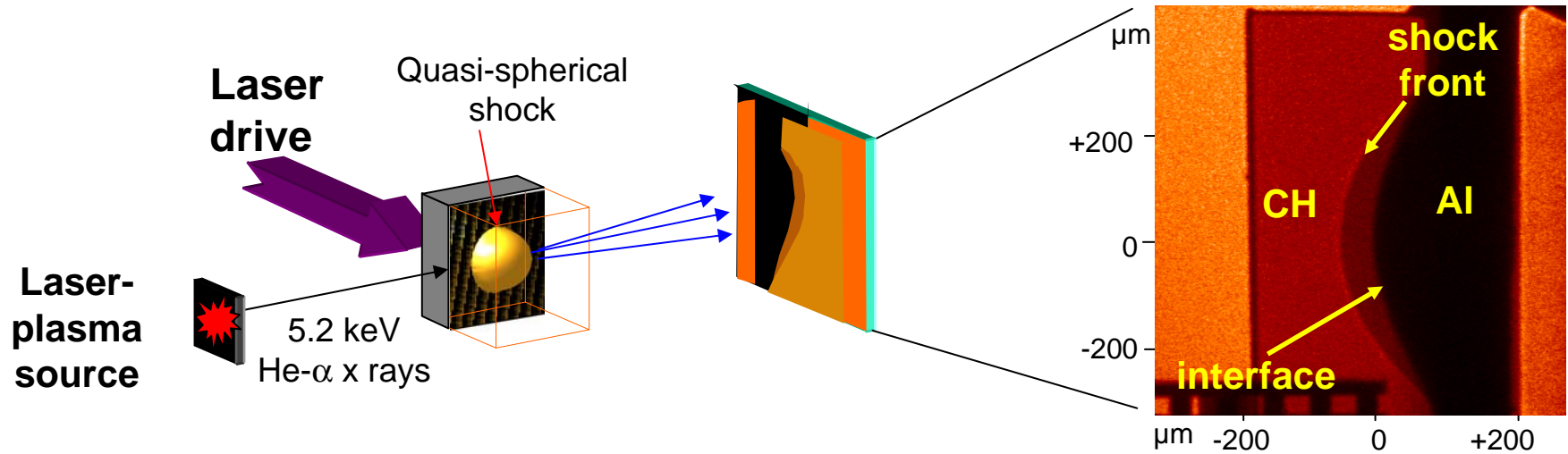


‘Unproposed’

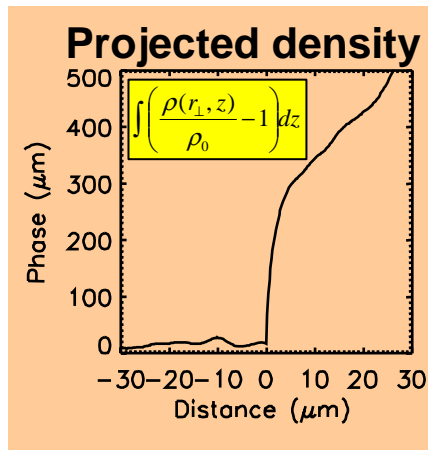
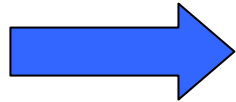
XFEL

uses

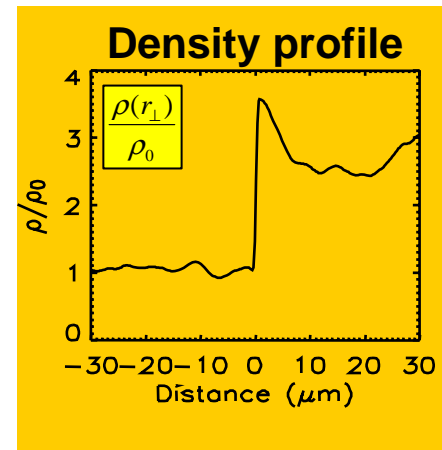
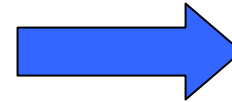
Current x-ray *phase-contrast imaging* at ~ 5 μm resolution uses laser-plasma sources



Iterative
Phase
Retrieval

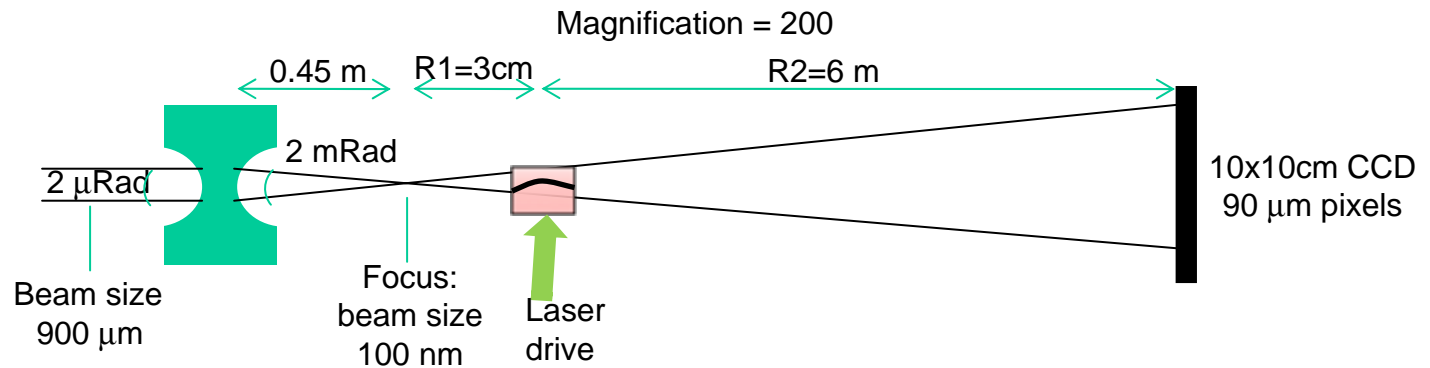


Tomo-
graphic
inversion



Current techniques are limited by spatial coherence & flux of laser-plasma x-ray source [D. G. Hicks 2006]

Phase contrast imaging of a laser-driven sample: Requirements summary



Requirements:

- Focal spot size: 100 nm
- Field of view (spot size @ target): $\sim 100\text{ }\mu\text{m}$
- Photons/pulse: $\leq 10^9$
- Pulse duration: ~ 10 to 100 fs
- Resolution:

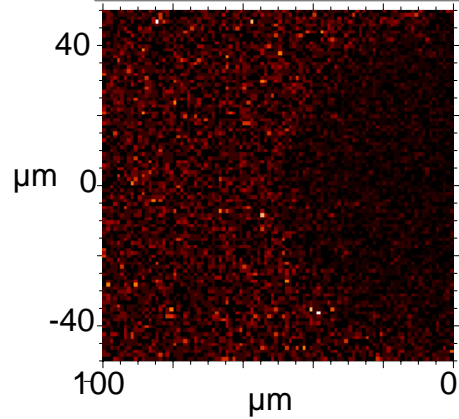
Max – 0.1 μm (this would need $< 90\text{ }\mu\text{m}$ CCD pixels with current magnification)

Min – 1 μm

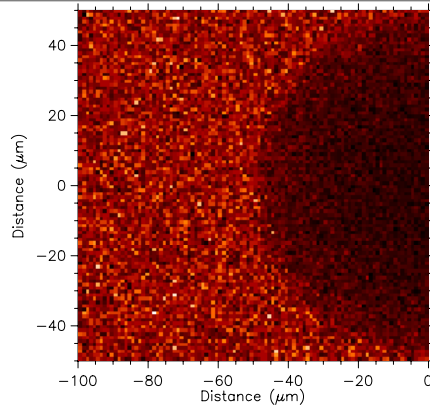
- Energy: $\sim 8\text{ keV}$ (5-15 keV would be useful to explore different materials)
- Bandwidth: $< 10\%$ for PCI only (probably $< 0.1\%$ for CDI)
- Capability for both side-on and face-on radiography
- R1 and R2 can be traded off to increase resolution at the expense of field of view
- High repetition-rate target positioner would enable accumulation of multiple

2-D imaging: Photon requirements

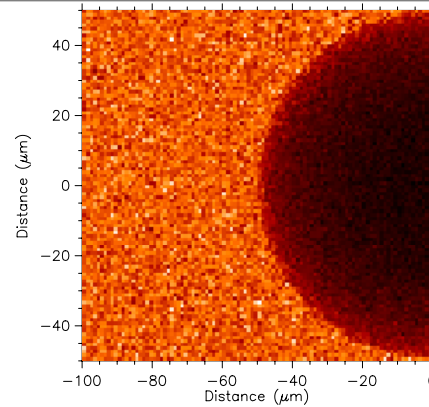
10^4 photons
1 μm resolution



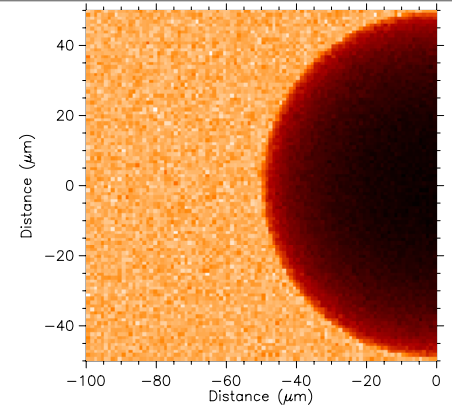
10^5 photons
1 μm resolution



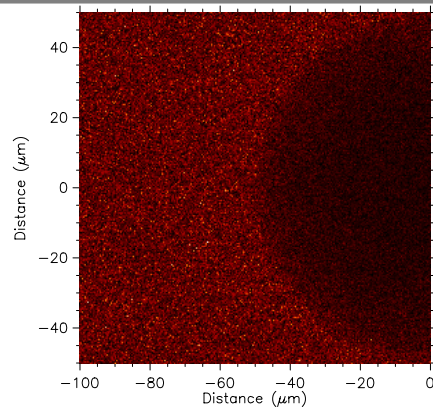
10^6 photons
1 μm resolution



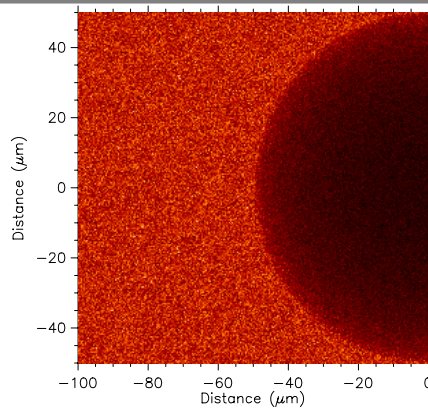
10^7 photons
1 μm resolution



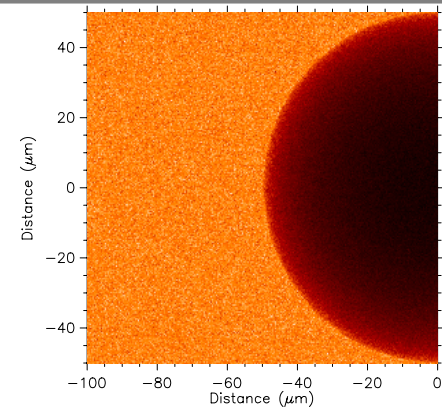
10^6 photons
0.2 μm resolution



10^7 photons
0.2 μm resolution

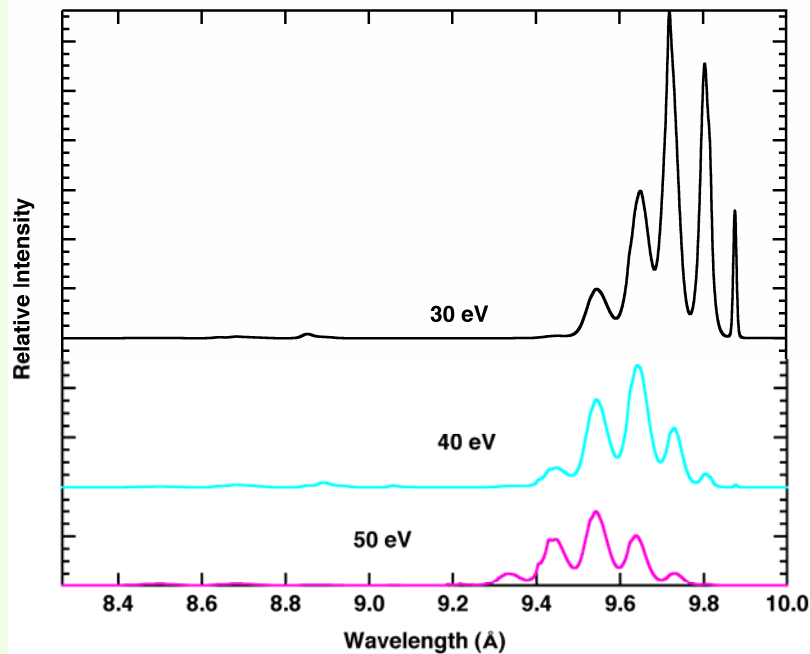


10^8 photons
0.2 μm resolution

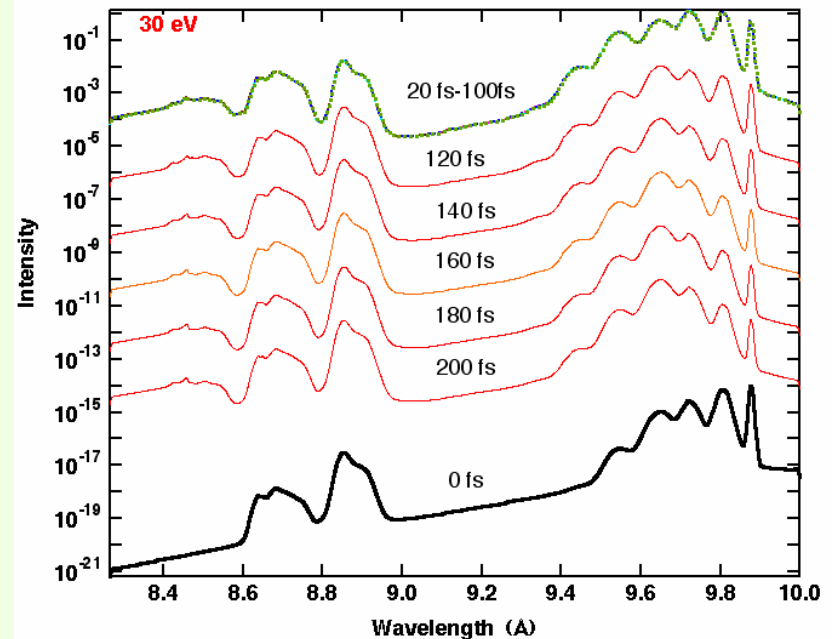


In Warm Dense Matter regime the hollow ions provide time-resolved diagnostic information

- XFEL can provide *in situ* diagnostics with 100 fs resolution
 - 5×10^{10} 1.85 keV photons in 30 μm spot into a $n_e = 10^{23} \text{ cm}^{-2}$ plasma
 - Strong coupling parameter, $\Gamma_{ii} = \text{Potential/Kinetic Energy} \sim 10$



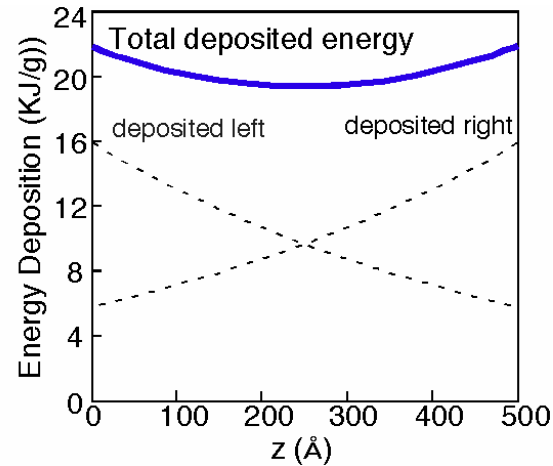
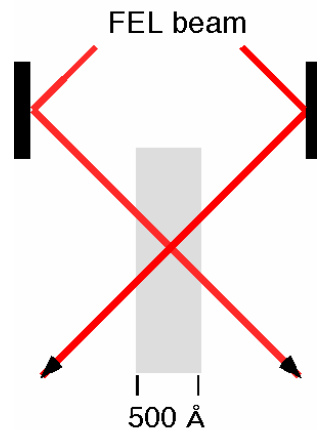
- Spectra vary measurably with T_e



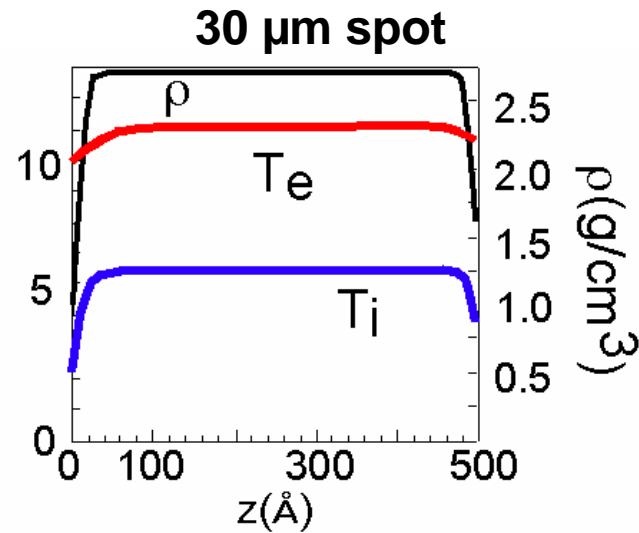
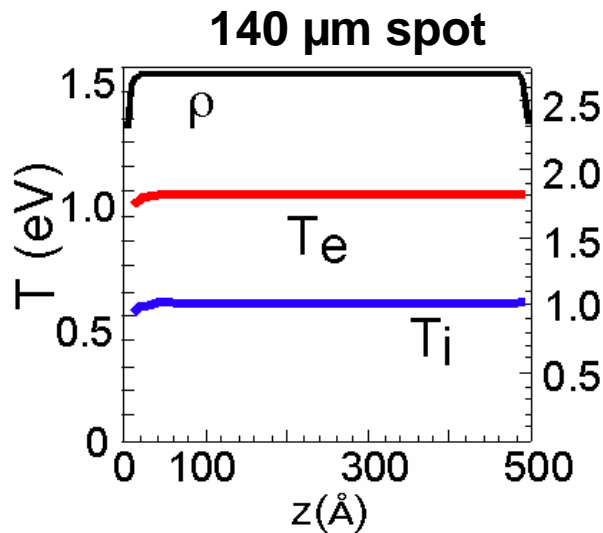
- At high n_e emission lasts ~ 100 fs

Simulations of two-sided illumination provide estimates for creating uniform WDM

- 500 Å Al irradiated by split FEL 10 fs beam

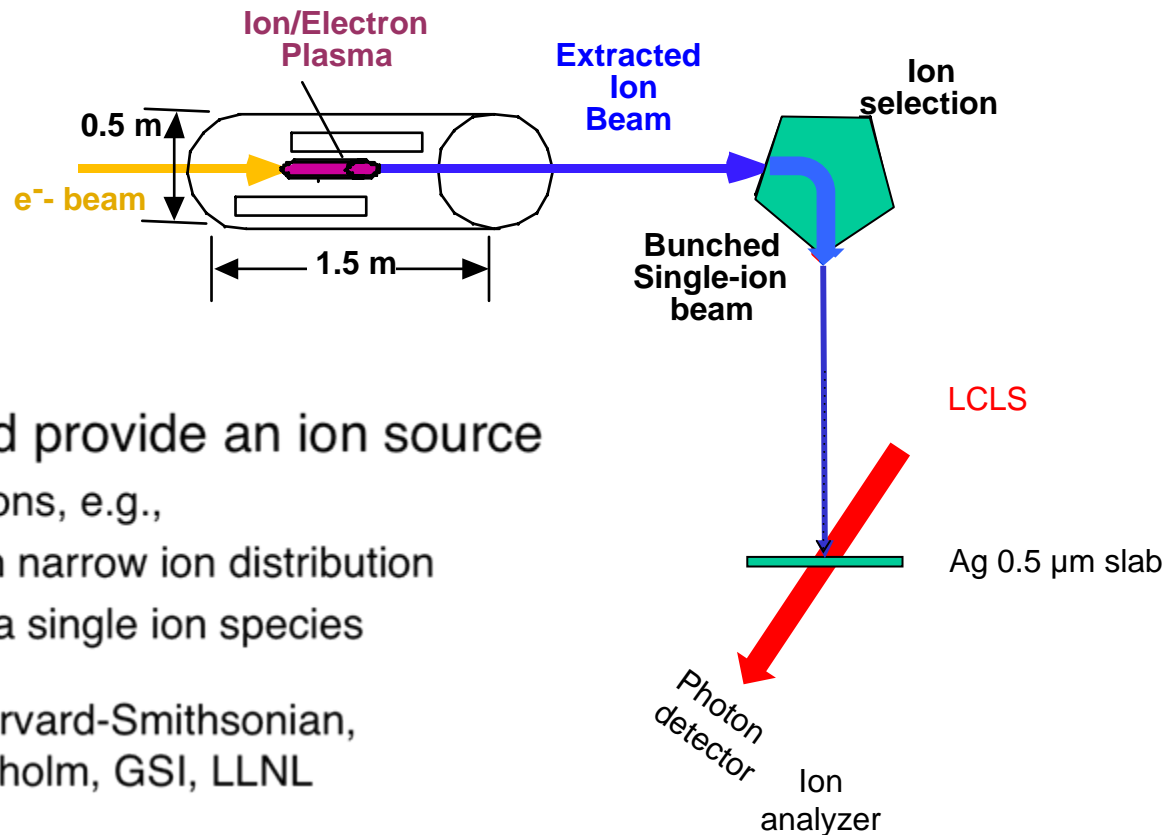


- Temperature and density at 100 fs after the FEL pulse



An LCLS created isochoric, isothermal slab should be ideal for WDM experiments

- Use LCLS beam with the split and delay to warm a slab from both sides of sample
 - Assume the $0.5\ \mu\text{m}$ Ag case with 4500 eV irradiation
 - Spatial variation in energy density is 3% - and similar for T and P
 - $T = 15\ \text{eV}$ and $P = 8.5\ \text{Mbar}$

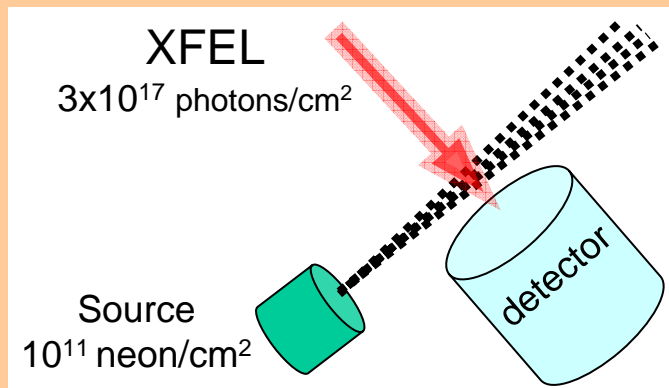


- Use of as EBIT would provide an ion source
 - Extracted beam of ions, e.g.,
 - $\sim 10^8$ /pulse with narrow ion distribution
 - $\sim 10^6$ /pulse for a single ion species
 - Collaboration: Harvard-Smithsonian, NIST, U. of Stockholm, GSI, LLNL

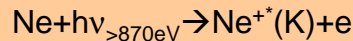
XFEL provides opportunity for AMO and HED plasma spectroscopy to join efforts

• AMO atomic physics case:

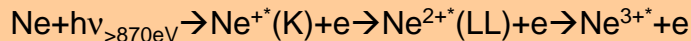
- Source for hollow ion experiment prepared as an atomic beam



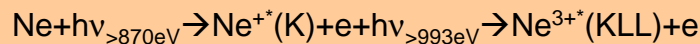
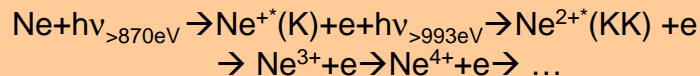
• Photoionization:



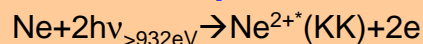
• Auger Decay:



• Sequential multiphoton ionization:

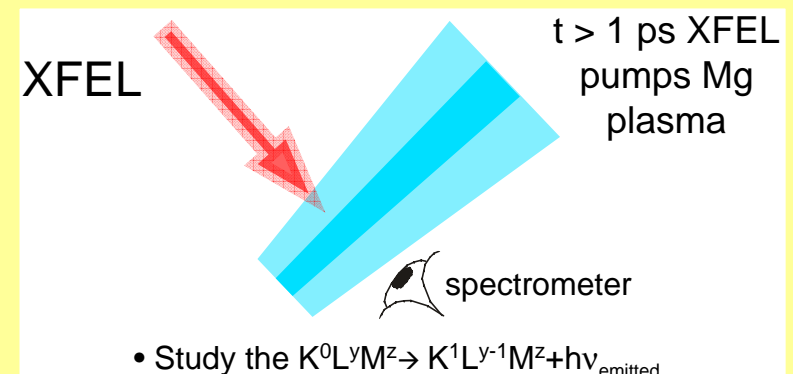


• Direct multiphoton ionization:

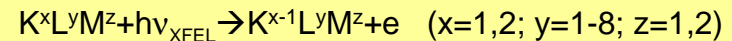


• HED 'atomic physics' case:

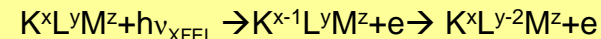
- Source for hollow ion experiment prepared by high energy laser



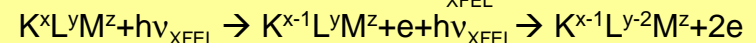
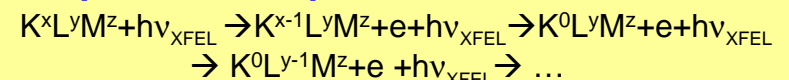
• Photoionization of multiple ion species:



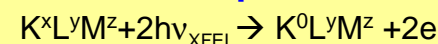
• Auger Decay of multiple ion species:



• Sequential multiphoton ionization:



• Direct multiphoton ionization:

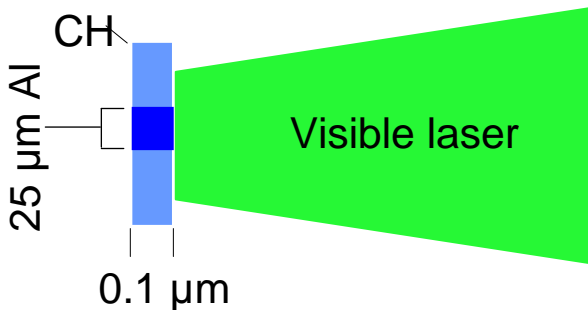


XFEL only

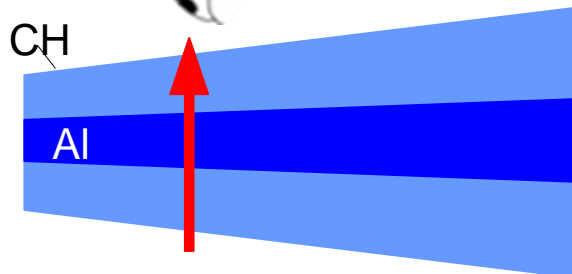
An XFEL can photopump a transition: provides critical tests of plasma processes

- Experiment

- $t = 0$ laser irradiates Al dot

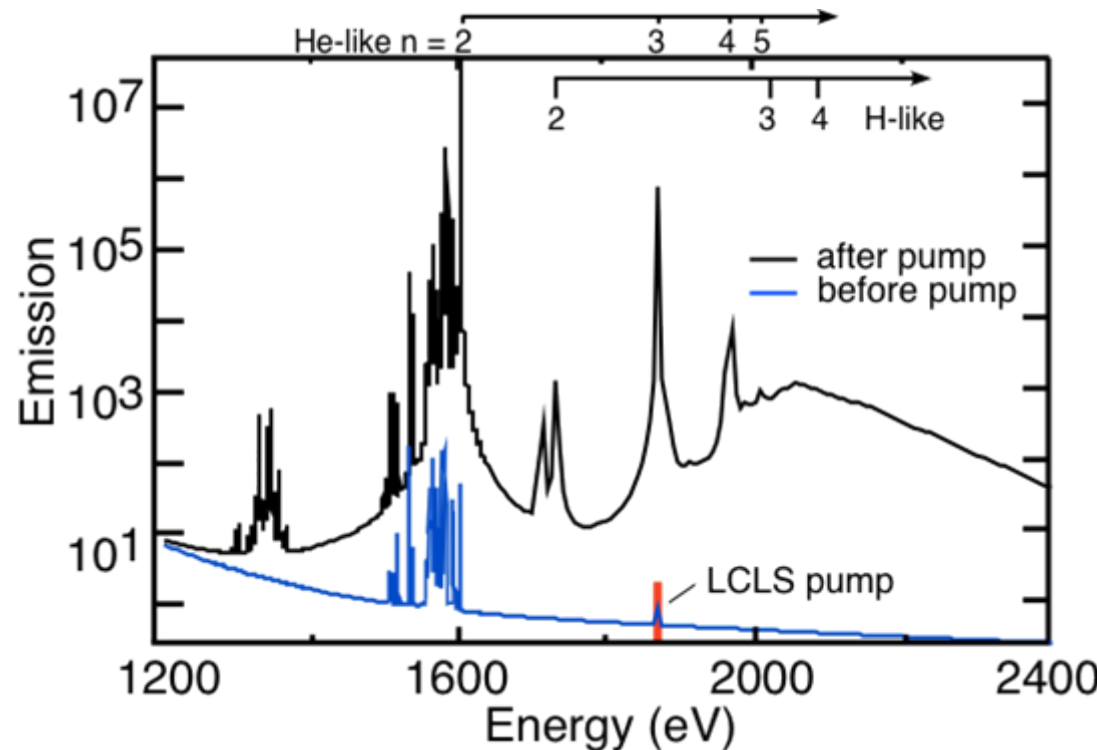


- $t = 100$ ps FEL pumps plasma
Observe emission with x-ray streak camera



X-rays pump tuned to 1869 eV

- Simulation



Line intensity, line position, and line shape *may* be effected by strong coupling

- Simple form for emission illustrates the observable aspects

$$I(\omega) = N_{UL} A_{UL} \hbar \omega_{UL} \phi(\omega)$$

level populations *line shape*

$$\phi(\omega) = \int d\varepsilon P(\varepsilon) J(\omega, \varepsilon) \quad \text{where } P(\varepsilon) \text{ is the ion microfield}$$

$$J(\omega, \varepsilon) \sim \frac{\text{Im}}{\pi} (\omega_{UL}(\varepsilon) + \gamma(\omega) + i\gamma(\omega))^{-1}$$

- Investigate $\phi(\omega)$ and $\gamma(\omega)$ to look at effects on shape
- Investigate $\delta(\omega)$ to look at line position (shift)
- Investigate kinetics for effects on populations, n_i

Ultimate test is the study of the radiation redistribution function $R(\omega_L, \omega_S)$

- I is the power spectrum of the radiation emitted at ω_S by a system pumped at ω_L

$$I(\omega_S, \omega_L) \propto \lim_{\eta \rightarrow 0} \text{Im} \sum_{i,f} p_i \left(\langle \langle \mathbf{V}_S | \mathbf{G}_W(i\eta) | \mathbf{V}_L \rho_o \rangle \rangle \right)_{i,f}$$

V_S = interaction for emission
 V_L = interaction with pump
 G = resolvent of the evolution

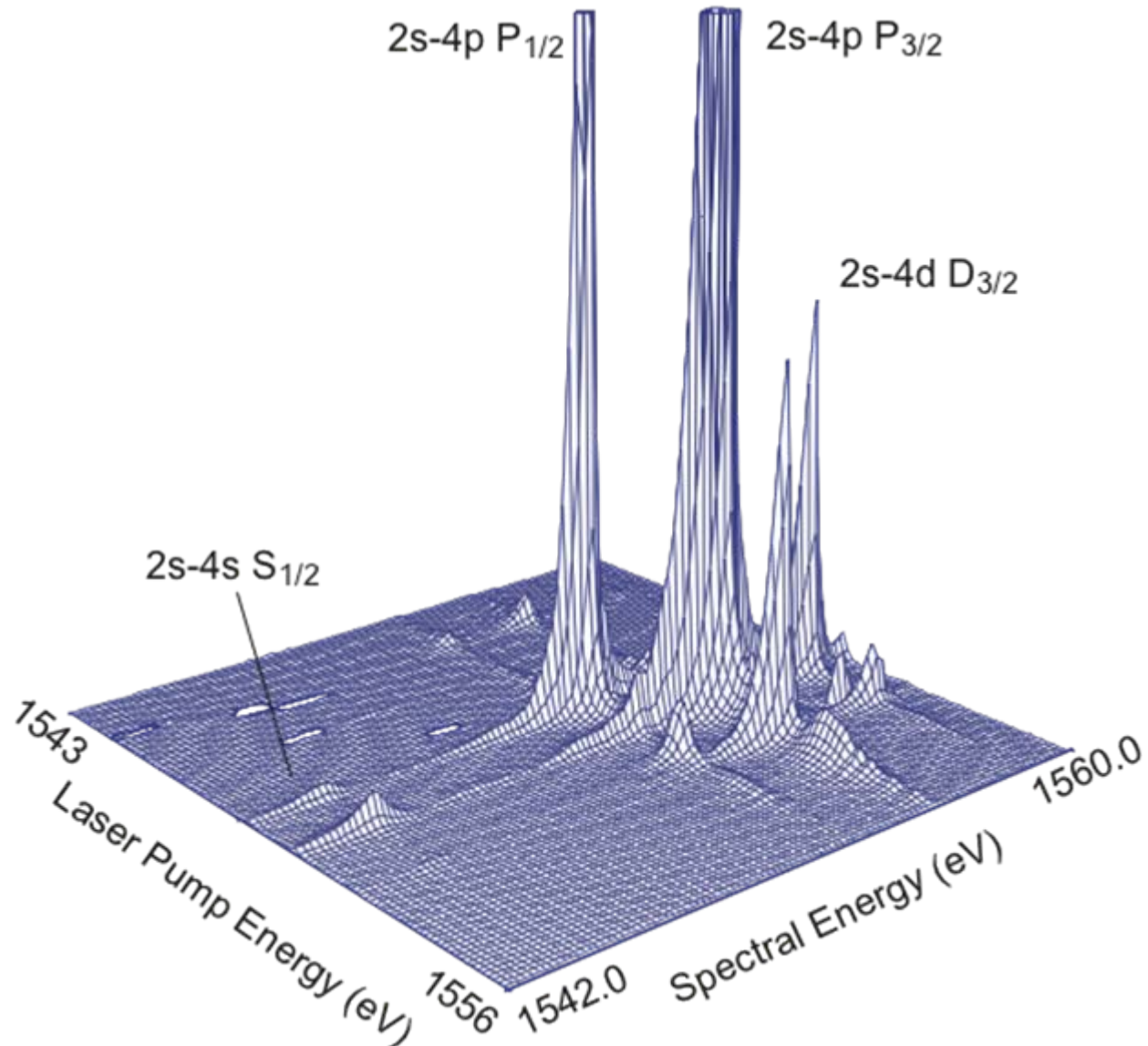
- $R(\omega_L, \omega_S)$ is the redistribution function

$$R(\omega_L, \omega_S) = \frac{I(\omega_L, \omega_S)}{\int \int I(\omega_L, \omega_S) d\omega_L d\omega_S}$$

- Investigate the redistribution using the XFEL

With bandwidth control and tuning, can pump within line to provide plasma rate data

- Example:
pumping Li-like
Fe $1s^22l - 1s^24l$
- Collision rates
and plasma field
fluctuations can
be measured
- Bandwidth of
 $\sim 10^{-4}$ is easily
obtained by use
of a crystal

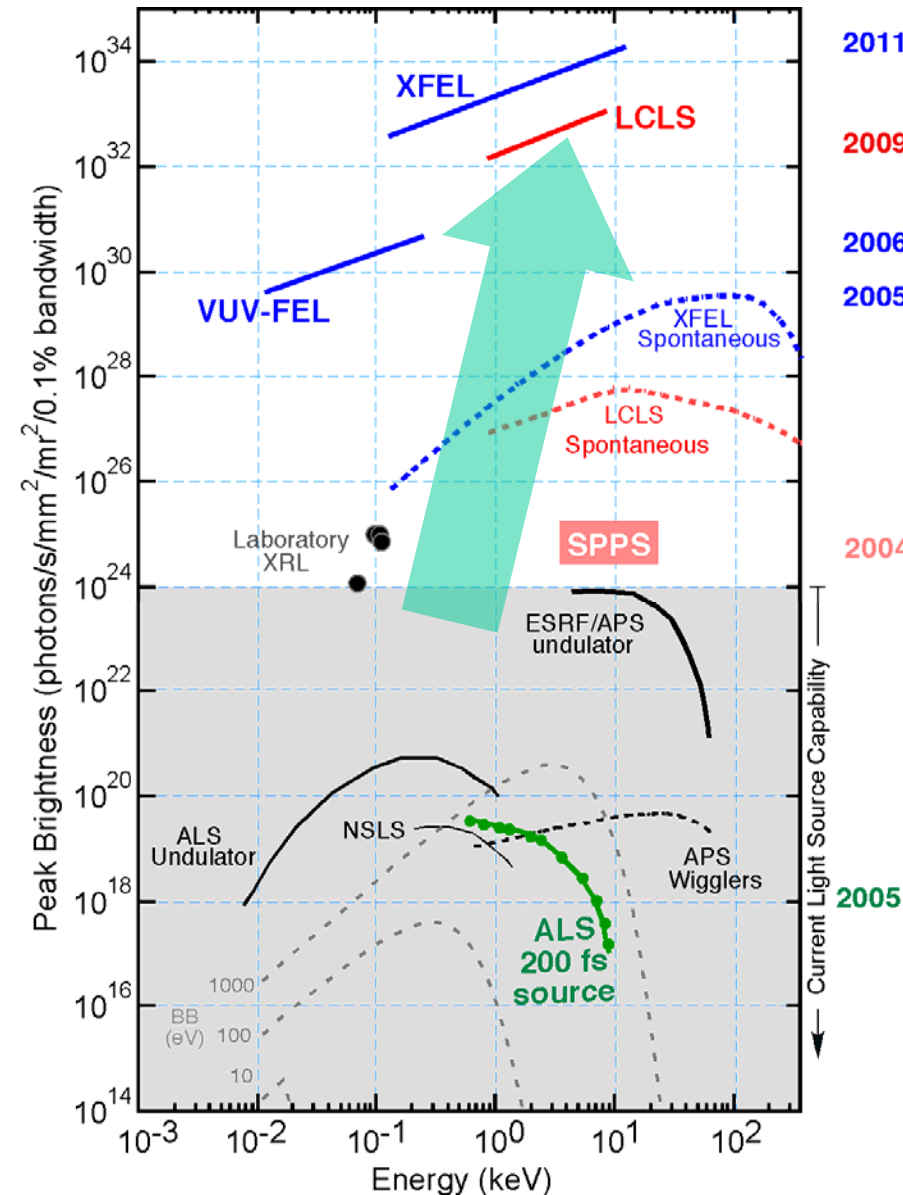


The End

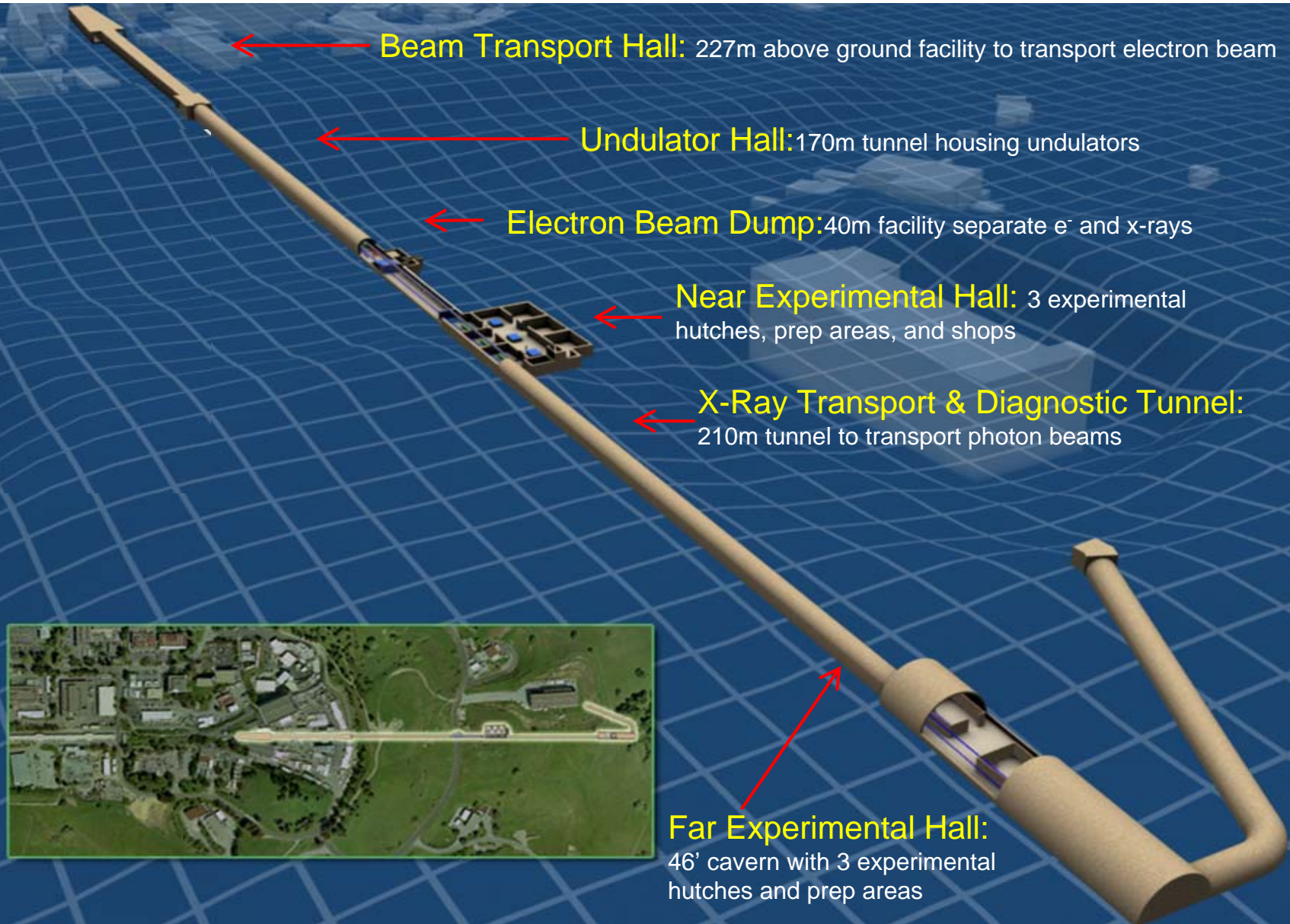
LCLS is the first x-ray FEL providing more than 10^{10} increase in peak x-ray brightness

FEL Specifications

- Short bunch duration
(~10 - 100 fs)
- Full transverse coherence
- High repetition rate
(~120 Hz)
- Tunable from 600 to 8500 eV
- High # of photons per bunch
> 10^{12}
- *Possibilities for HED studies*

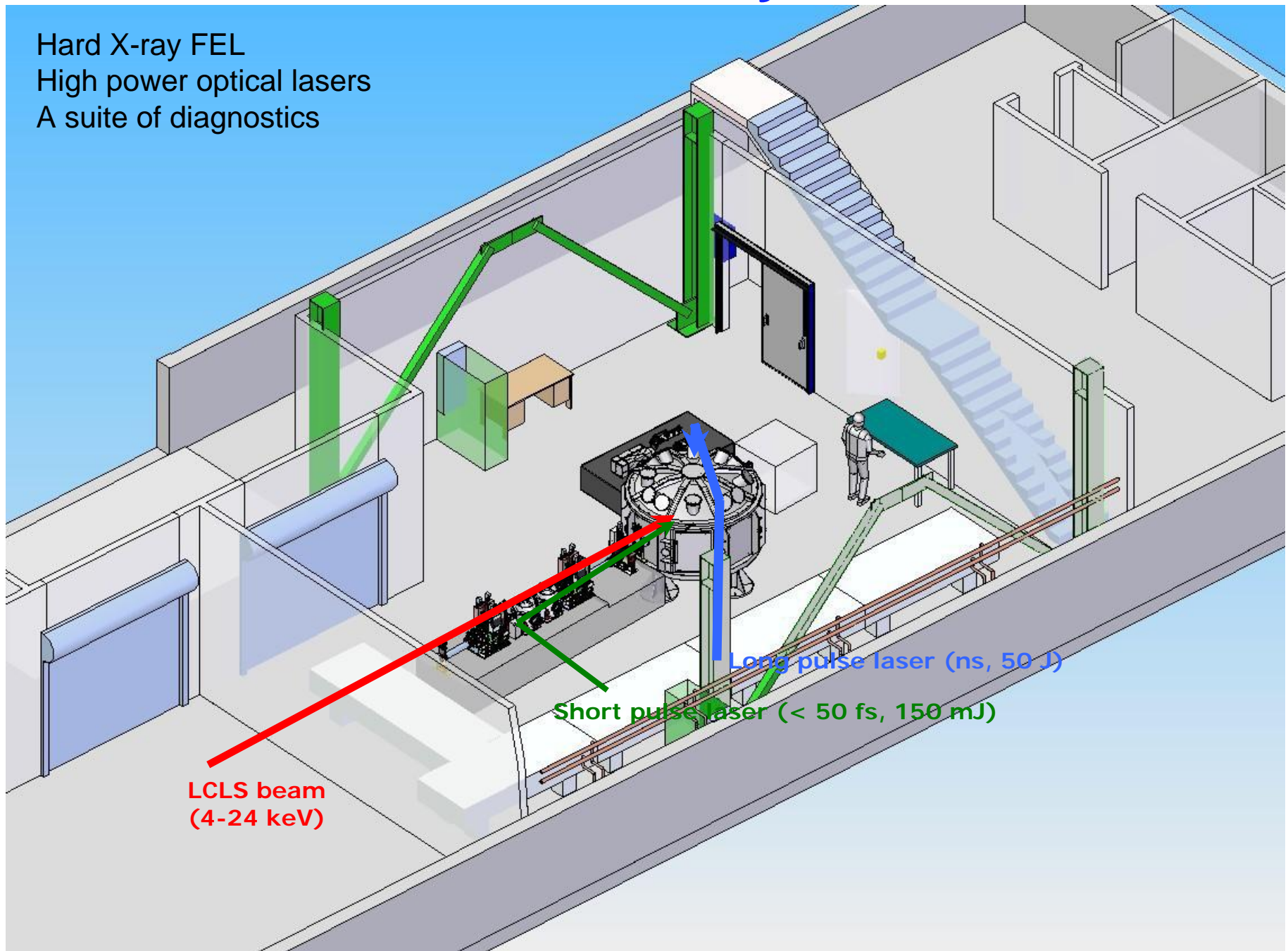


Overview of LCLS: Linac to X-Ray Halls



MEC instrument layout

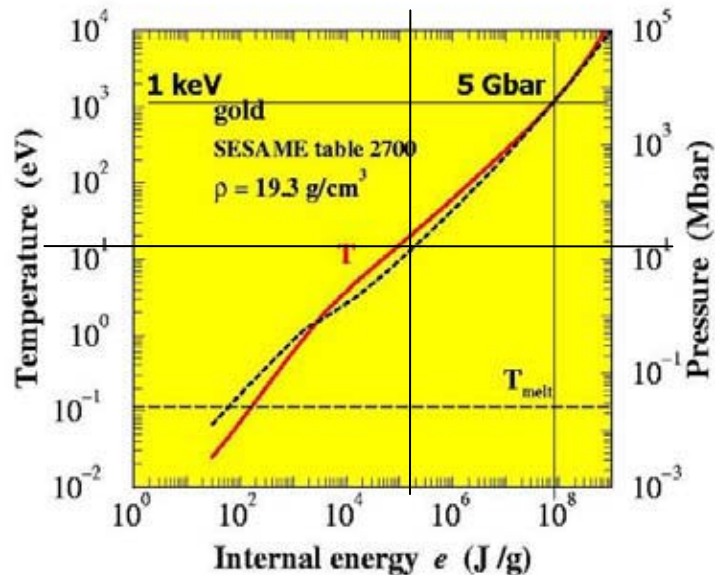
Hard X-ray FEL
High power optical lasers
A suite of diagnostics



LCLS X-ray laser can create high pressures without generating a shock

- Phase-Space Plot*

Temperature and Pressure
Versus Internal Energy

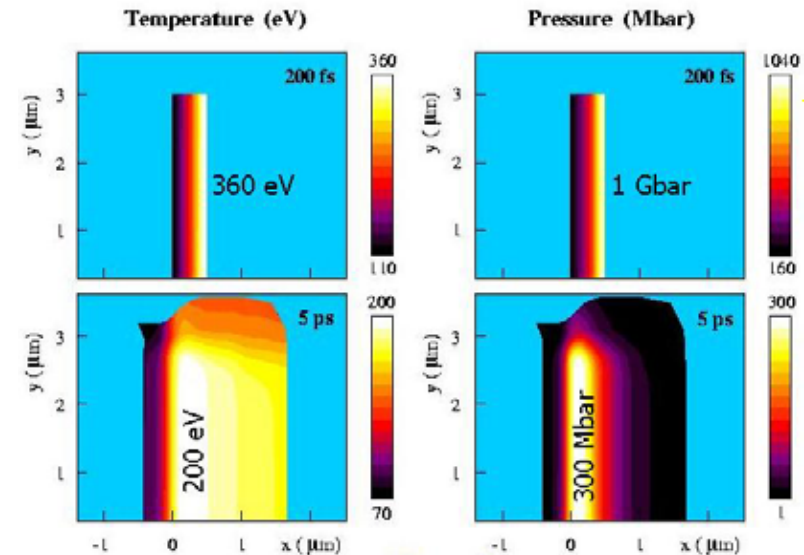


- Temperature (—), Pressure (---) vs J/g
- (— →) melting T reached with ~ 100 J/g
- Solid horizontal line corresponds to 10^8 J/g deposition, generating solid plasma of 1 keV, 5 Gbar

* Sesame / LANL

- Simulations*

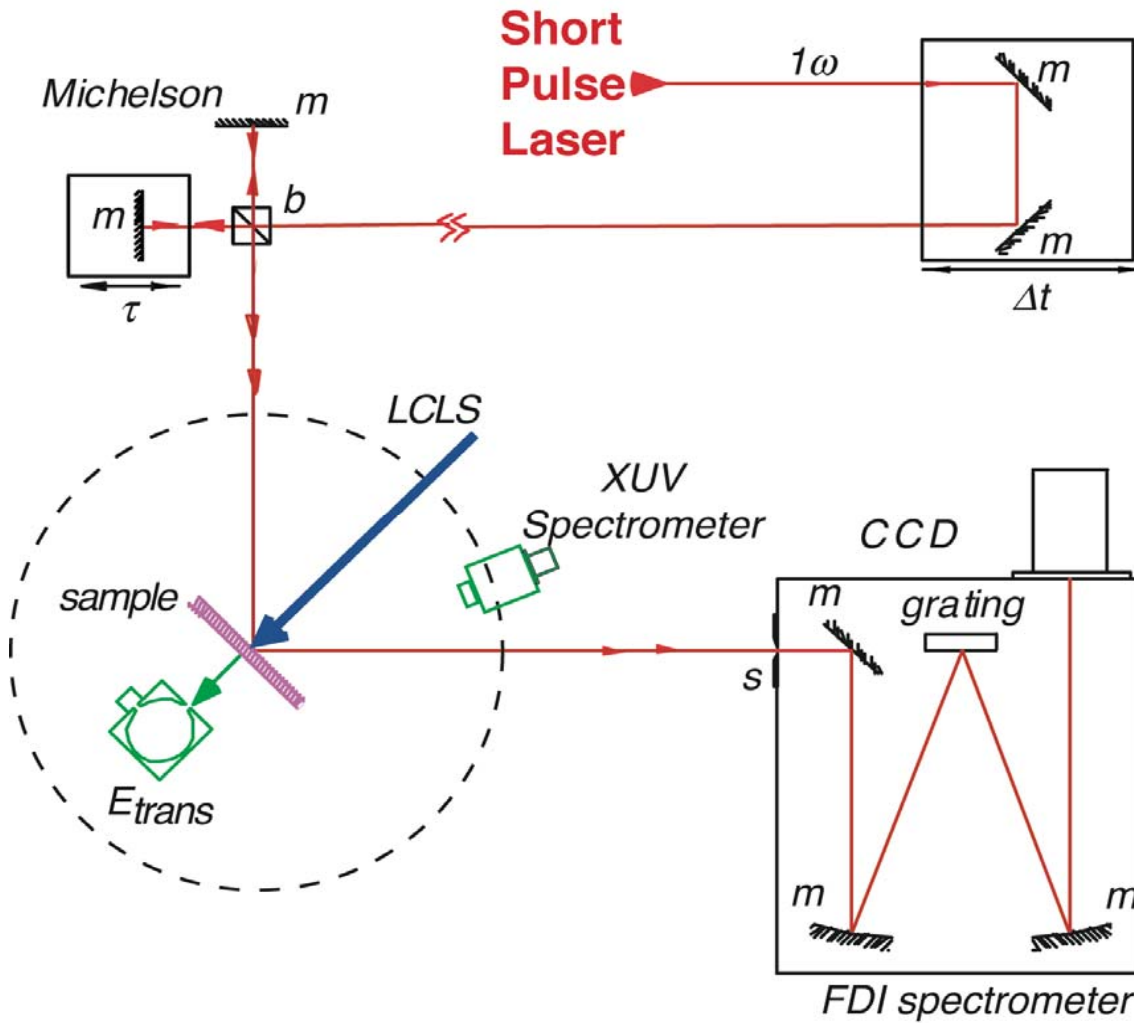
Temperature and Pressure
Versus Time and Space



- Simulation: $0.5 \mu\text{m}$ Au
- Thickness is two absorption lengths
- XFEL: 100 fs, 10^{17} W/cm^2 pulse, 3.1 keV comes from right

* 2-D Multi-code / MPI

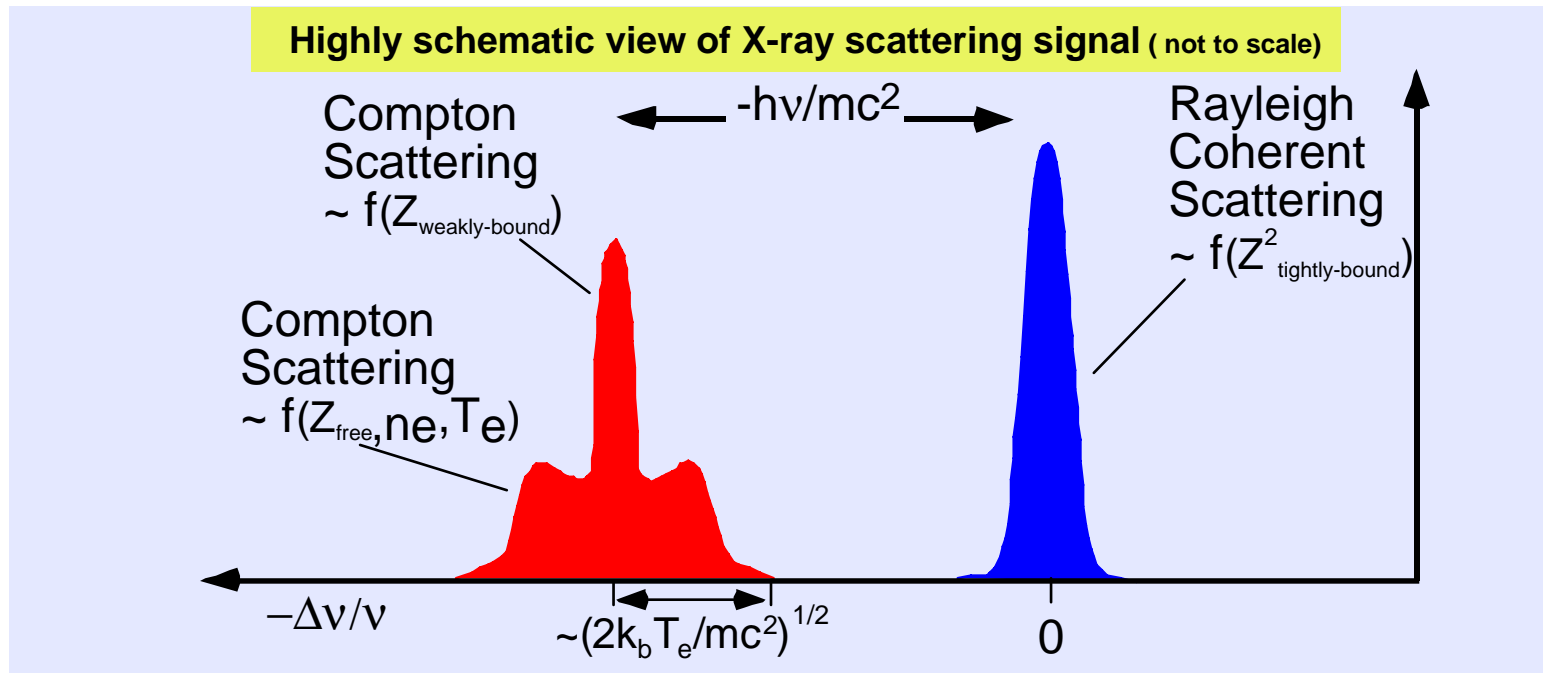
Experiment will monitor the expansion on sub-ps time scales using FDI



- Experiment will measure incident/transmitted energy
- **Front** and **rear** surface expansion using FDI (*only front is shown*)
- XUV emission from the 18eV front surface
- Resolution in time is $\sim 35\text{fs}$
- Resolution in space is $\sim 4\text{\AA}$
- Data is sufficient to test various EOS models

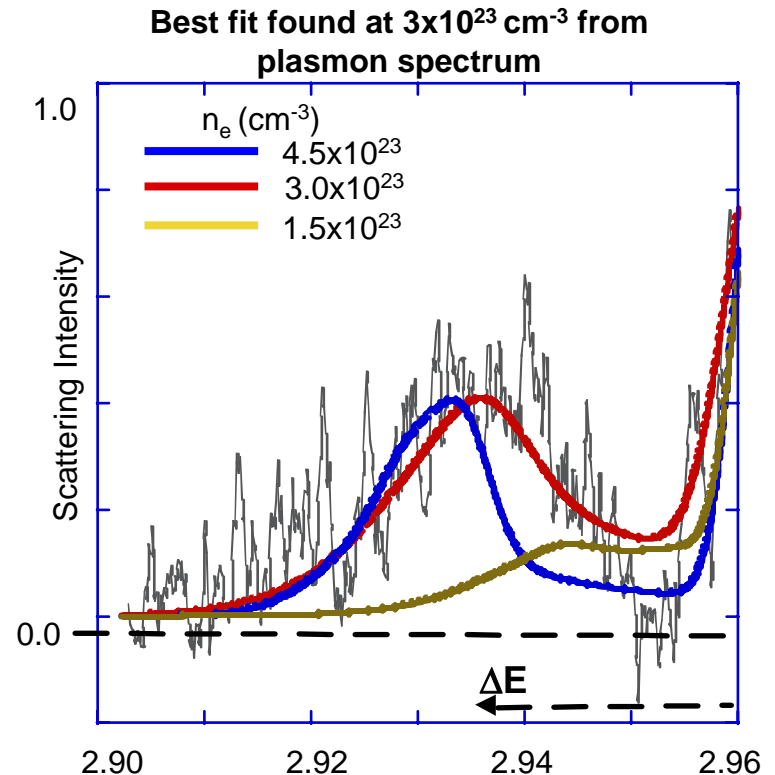
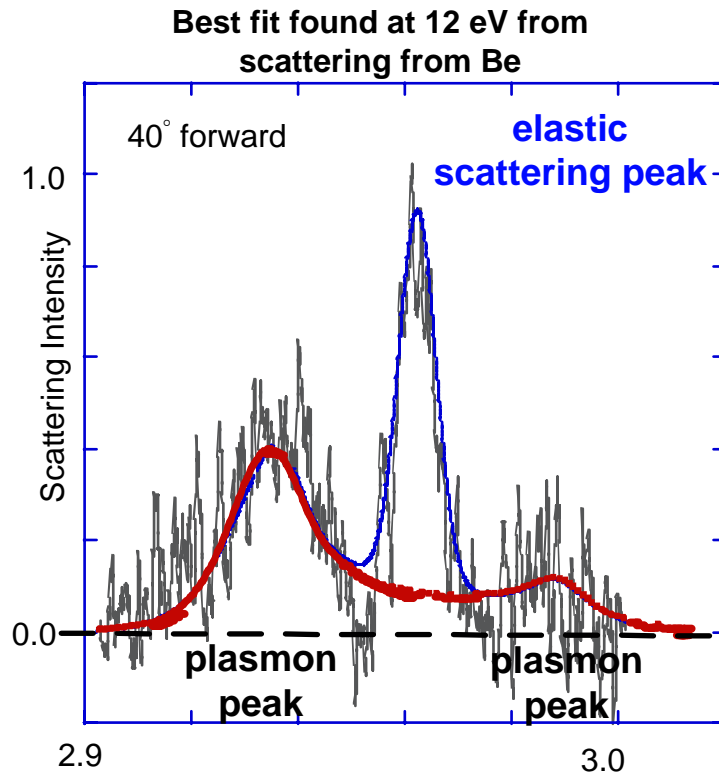
Scattering of the XFEL will provide data on free, tightly-, and weakly-bound electrons

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



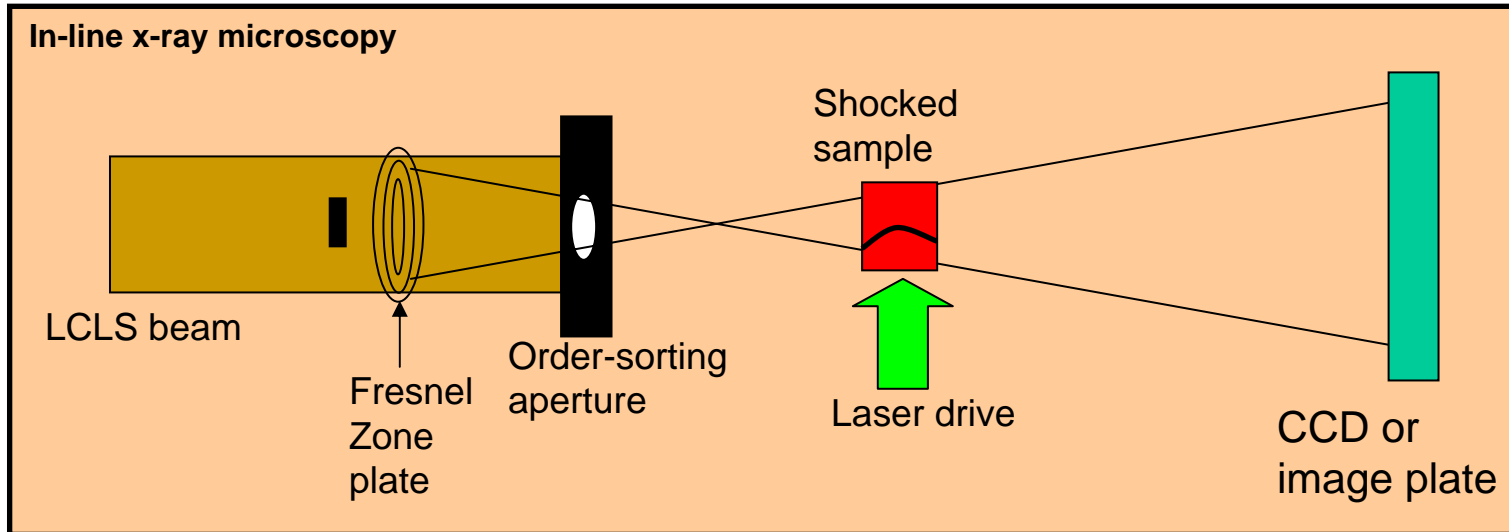
- For a 25 eV, $4 \times 10^{23} \text{ cm}^{-3}$ plasma the XFEL produces 10^4 photons from the free electron scattering
- Can obtain temperatures, densities, mean ionization, velocity distribution from the scattering signal

Thomson forward scattering provides data from collective regime: plasmons yield information

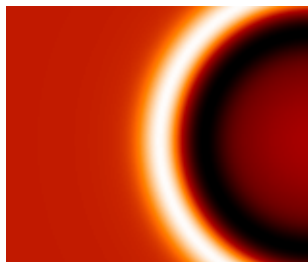


- Plasmon peak intensity related by detailed balance, i.e., $\exp(-2\Delta E/T)$
- Experiments with independent T_e measurement are needed to determine correct approximation for collisions
- ***Experiments have now been performed with photon numbers consistent with LCLS capability.***

In-line x-ray microscopy: Phase contrast and Coherent diffractive imaging



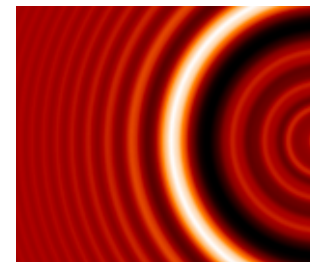
Shock front @ 2 μm resolution



Phase-contrast
imaging (near-field)

**Single-pulse imaging:
 10^5 - 10^7 photons/pulse
(resolution dependent)**

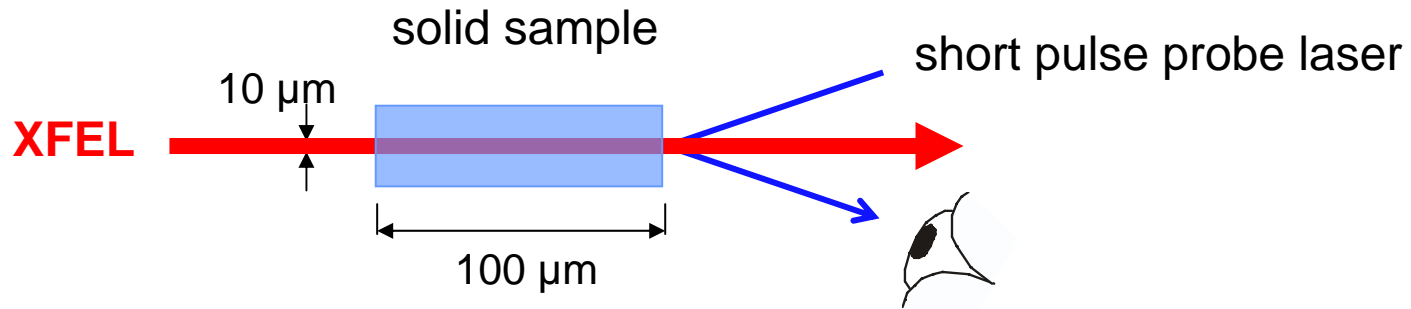
Shock front @ 50 nm resolution



Coherent-diffractive
imaging (far-field)

**With sufficiently high beam coherence, i.e., high spatial resolution,
Phase Contrast imaging \rightarrow Coherent Diffractive imaging**

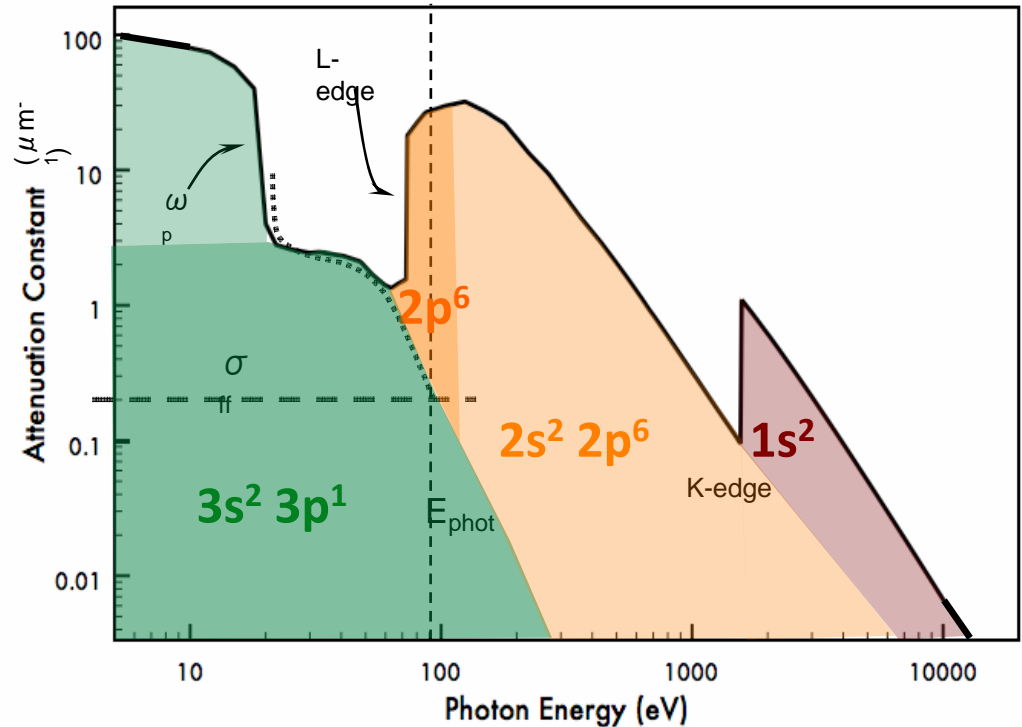
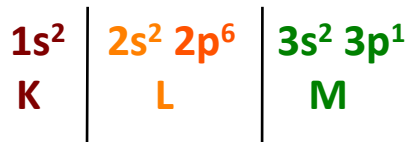
Intense short pulse x-ray sources can create WDM



- For a 10x10x100 μm thick sample of Al
 - Ensure sample uniformity by using only 66% of beam energy
 - Equating absorbed energy to total kinetic and ionization energy
$$\frac{E}{V} = \frac{3}{2}n_e T_e + \sum_i n_i I_p^i \text{ where } I_p^i = \text{ionization potential of stage } i - 1$$
 - Find 10 eV at solid density with $n_e = 2 \times 10^{22} \text{ cm}^{-3}$ and $\langle Z \rangle \sim 0.3$
- State of material on release can be measured with a short pulse laser
- Material, rapidly and uniformly heated, releases isentropically

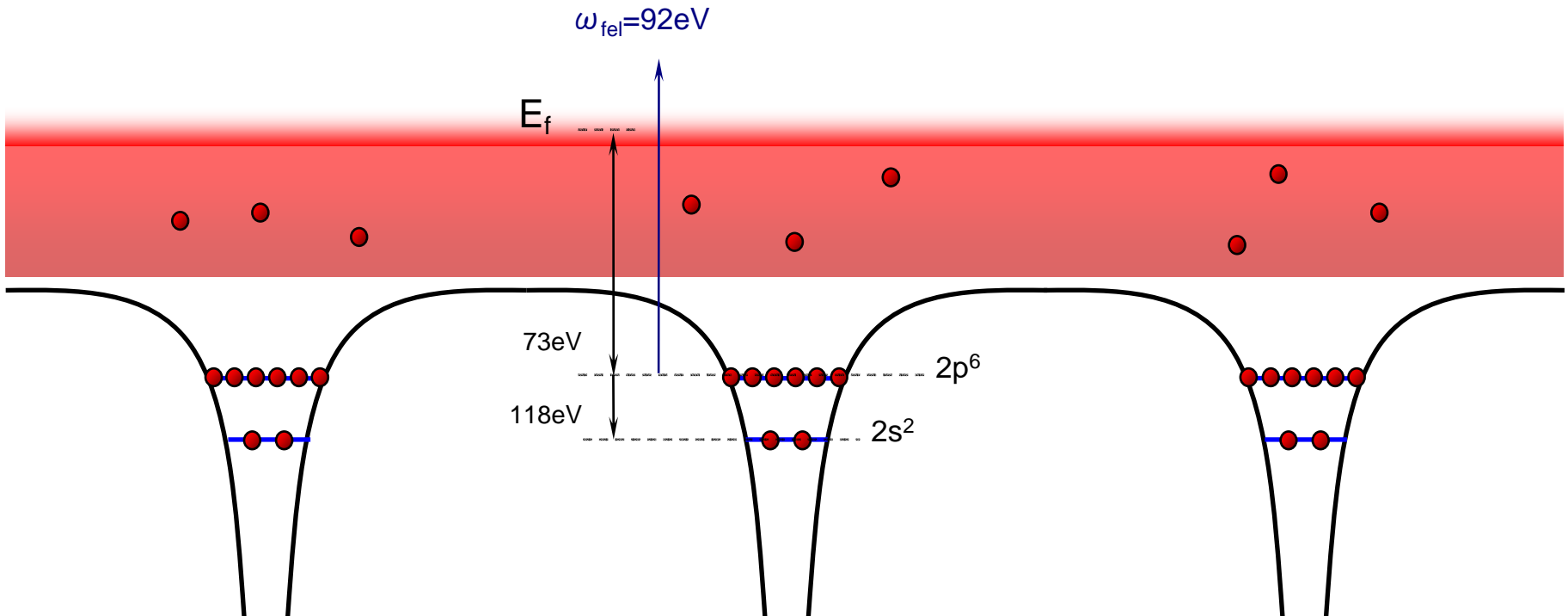
Absorption in solid density aluminium

Electron configuration in atomic aluminium:

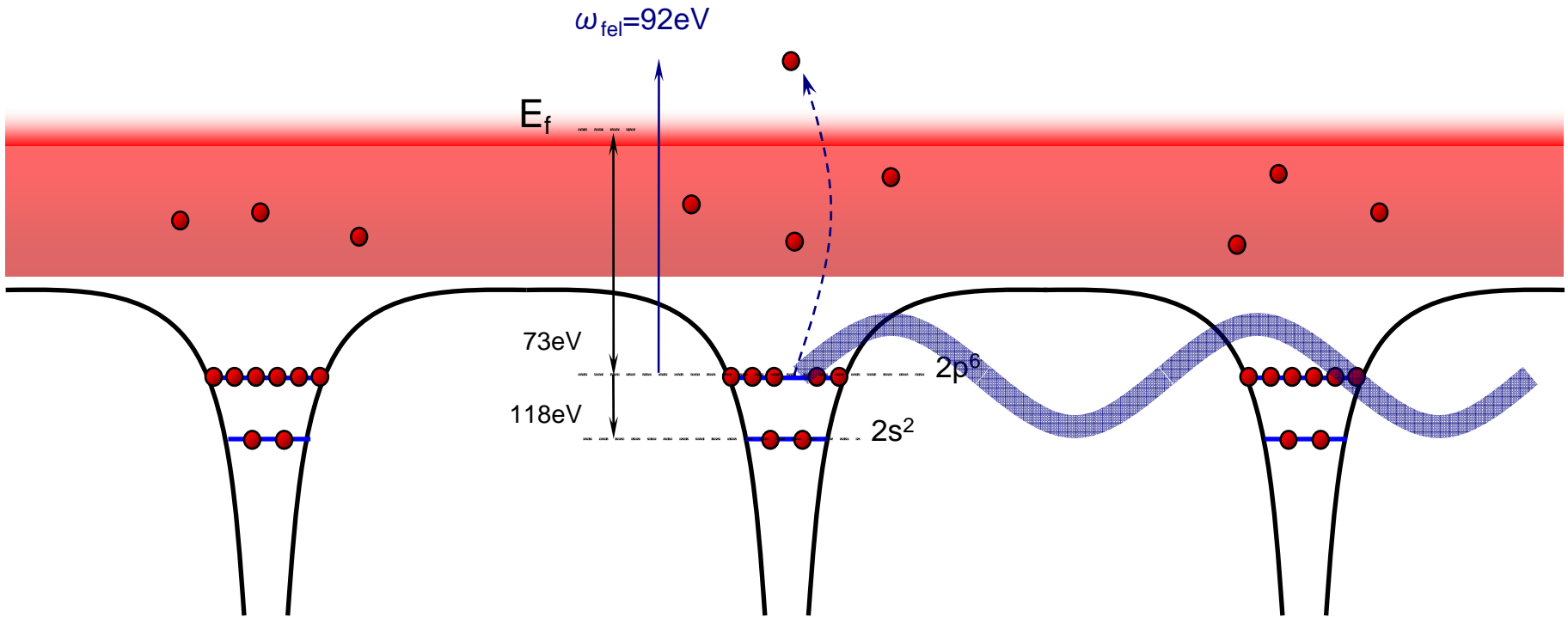


- at 92 eV two absorption channels:
 - bound-free excitation of L-electrons: $\sigma = 27 \mu\text{m}^{-1}$
(CXRO tables online <http://www-cxro.lbl.gov/>)
 - free-free excitation of the valence band: $\sigma = 0.2 \mu\text{m}^{-1}$
(S.M. Vinko et al., HEDP 5, 124 (2009))

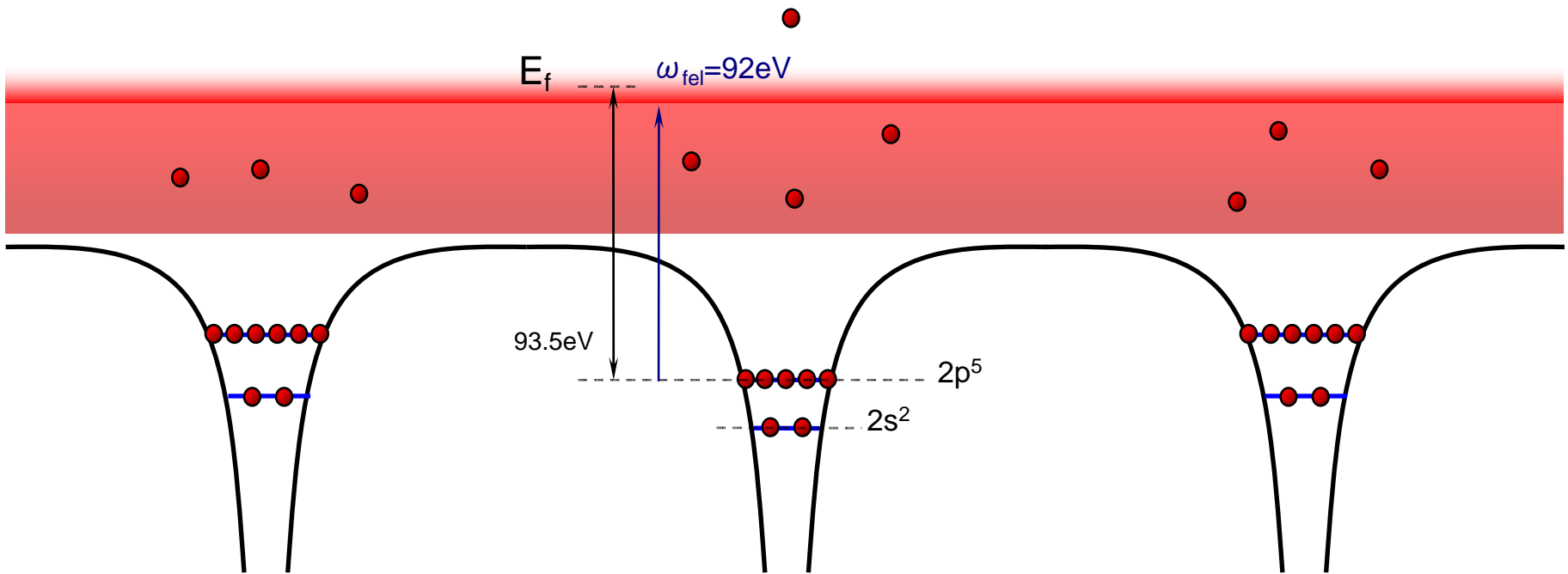
L-shell electron excitation



L-shell electron excitation

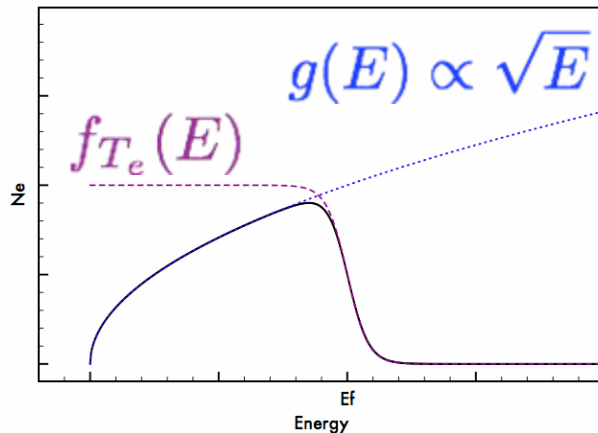
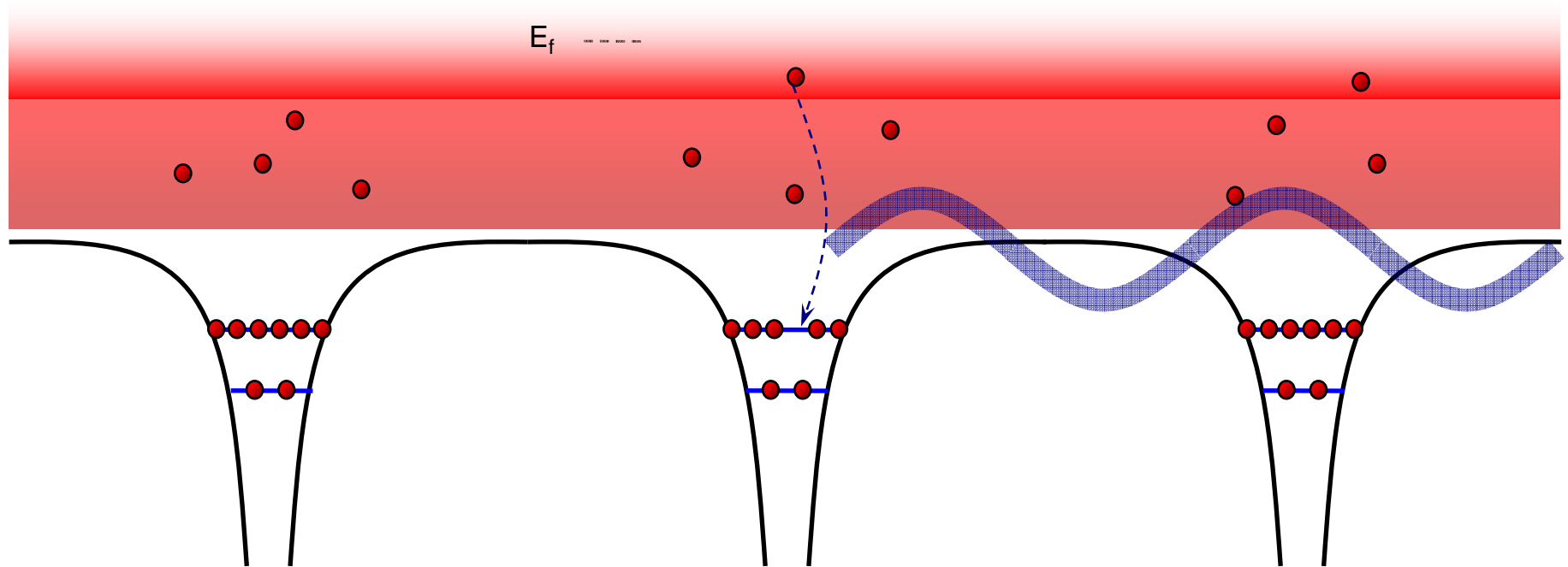


L-shell electron excitation



L-shell hole lifetime in Al ~ 40 fs

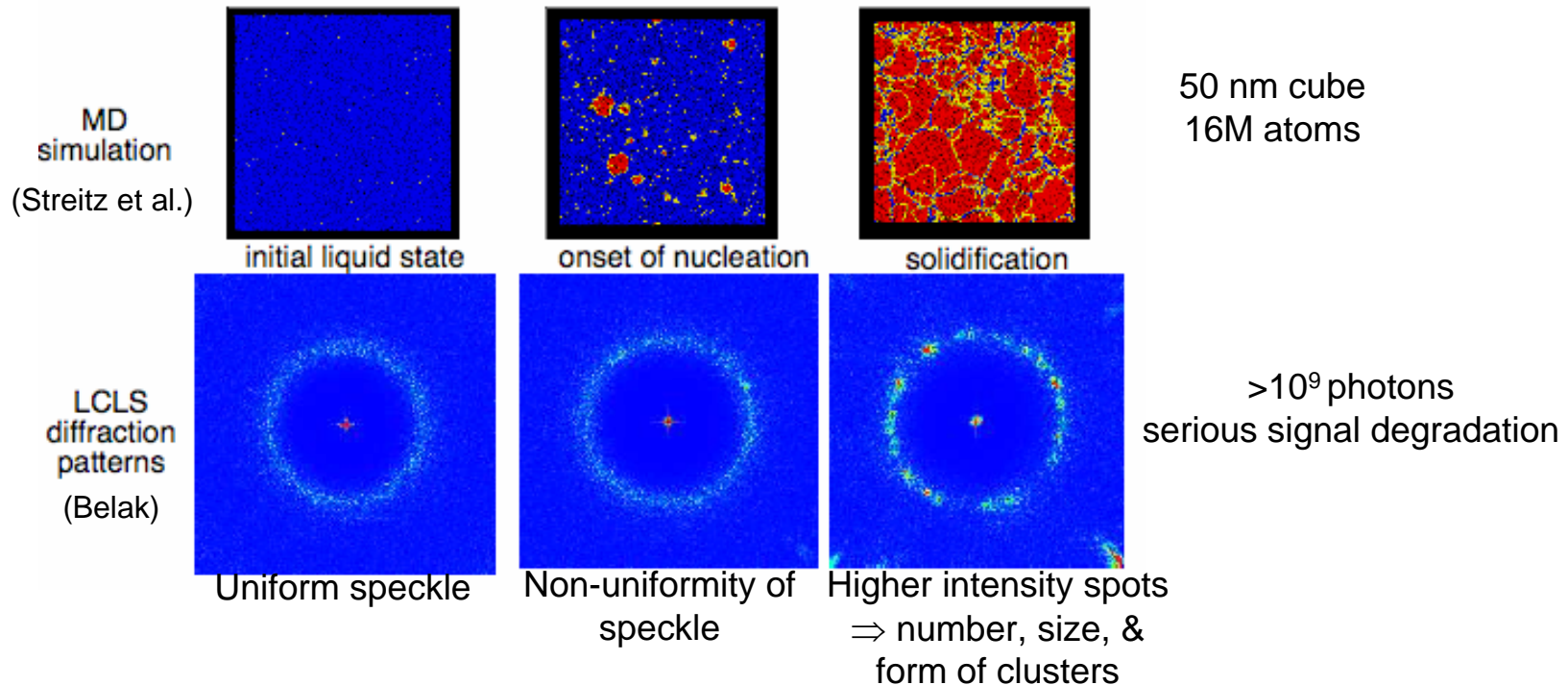
Core hole recombination - soft x-ray emission



Soft x-ray emission 0.2 % of total recombination, rest is Auger

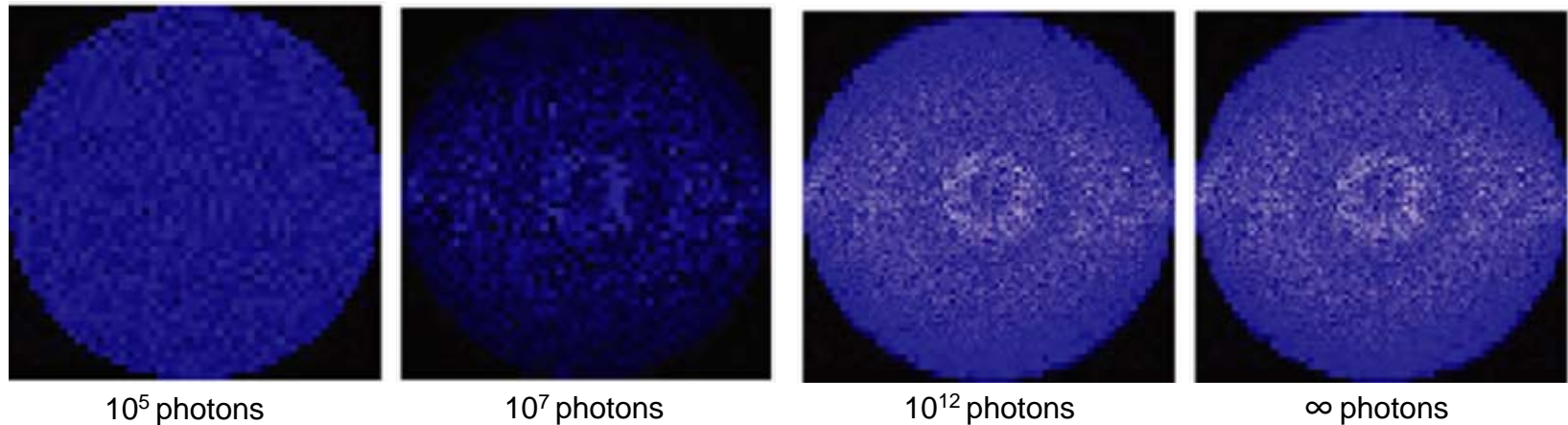
$$I(E) \sim \omega^3 g(E) f_{T_e}(E)$$

MD simulations of Ta show nucleation and solidification the XFEL can probe



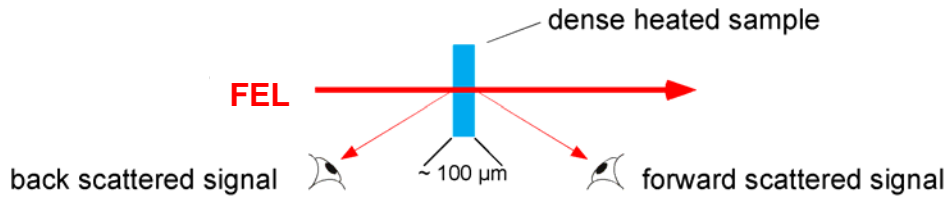
- Goal of *in situ* x-ray diffraction of shocked solids at granular level is to understand the microscopic to inform mesoscopic, and then macroscopic
- Study how individual grains respond elastically and plastically to high pressure as a function of orientation with respect to a given uniaxial shock wave
- XFEL is ideally suited to probe via diffraction polycrystalline high pressure solids because it is an ultra-bright, non-diverging, monochromatic source.

Use the MD simulation to generate the diffractive imaging signal

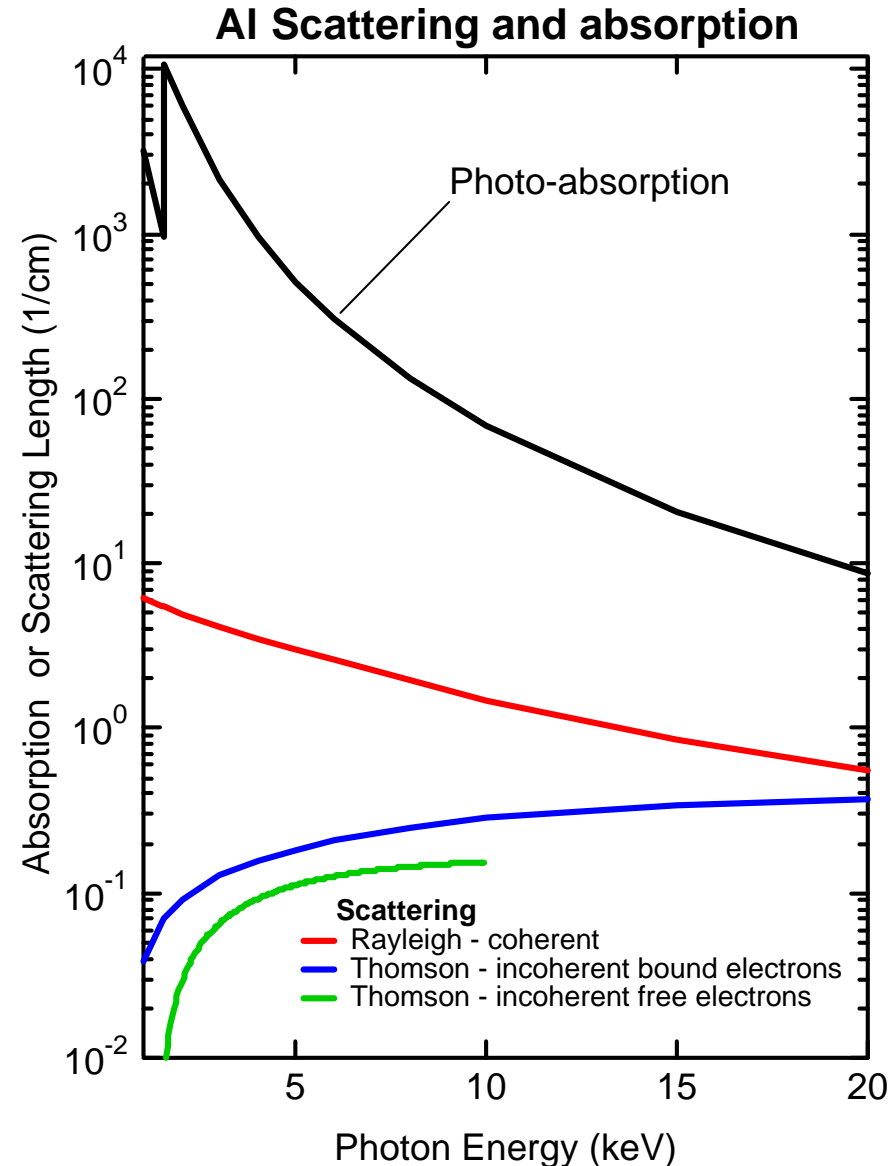


- *MD simulations by Streitz et al., 16M atoms, .5 ns, 100 nm³*
- *X-ray scattering simulations by Stefan Hau-Riege*
- *Somewhere between 10^7 and 10^{12} seems adequate*

X-ray '*Thomson Scattering*' will provide a unique probe for HED matter

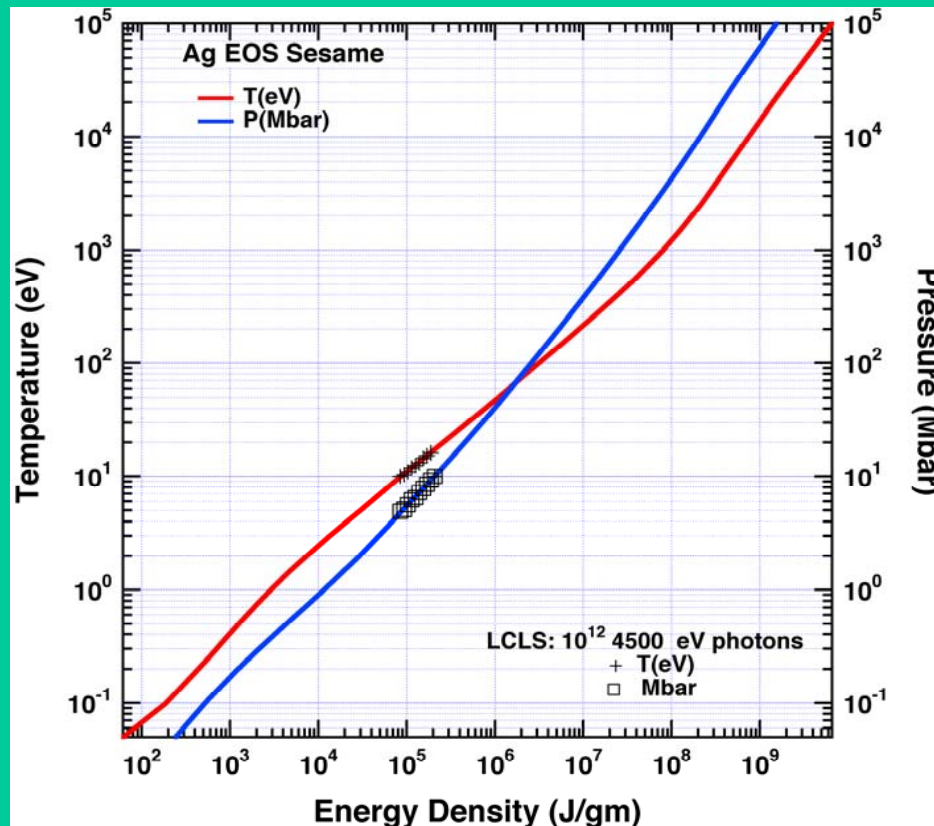


- Due to large disparity of photoabsorption and TS cross-sections, heating while probing is a problem
 - Use photons well above K-edge
 - Probe matter at higher temperatures
- Proof of principle experiment performed with appropriate photon numbers on laser facilities.



1st planned high pressure WDM experiment will study x-ray energy deposition

- LCLS can be focused to 6 μm Au case **but** to provide a diagnosable surface one needs to verify sample response



Further the locus of points for the heating of 1 μm of Ag is indicated for each 0.1 μm section by + and x symbols for the temperature in eV and the pressure in Mbar, respectively, where the highest temperature and pressure are at the irradiated surface.

- The time scale for the deposition is 100fs so there is effectively no hydrodynamic motion during deposition
- As long as the LCLS does not saturate the absorption the energy density deposited can be calculated by using:

$$E_D = n_{\text{photons}} E_{\text{photon}} (1 - e^{-l/\tau})$$

E_D is the energy density of volume of width l and cross-sectional area of the beam, and τ is the absorption length.

- Assuming the system is approximately described by the equilibrium equation of state one can use EOS table to look up the temperature and density

Using an XFEL to pump within a line transition is fundamentally important

- Measuring redistribution within a Stark-broadened bound-bound profile
 - Assumption of complete redistribution within a profile can be invalid
 - ion field fluctuations
 - inelastic collisions
- Measuring the detailed redistribution of population by pumping within a transition can indicate relative plasma rate process

Comment: Obtaining beam time at LCLS

- The LCLS has 6 end stations each designed for a specific scientific area.
- However, for proposals one gets to choose the end station best suited to the experiment
 - e.g., the high pressure study will be performed in the XPP end station
- We encourage you to think of experiments that are important and submit proposals
- ***The selection process is based on the best science possible at the LCLS***