Exploring Materials in Extreme Conditions
(using a variety of x-ray sources)

Roger Falcone

Physics Department
Berkeley

Advanced Light Source
Berkeley Lab

Doing Business

- Facts of LCLS Life
  - LCLS is the most visible project in the Office of Science
  - It is guaranteed we will be carefully scrutinized
    - Inspector General
    - OECM – IER might continue to ∞
    - Safety
  - Be prepared

John N. Galayda
galayda@slac.stanford.edu

Thrust areas:		SSRL/LCLS Contact
(1) Coherent scattering at the nanoscale	G.B. Stephenson* K. Ludwig	S. Brennan
(2) Pump/probe diffraction dynamics	K. Gaffney* D. Reis J. Larsson	A. Lindenberg
(3) High energy densit[illegible]	[illegible] [illegible] Heimann	[illegible]
(4) Nano-particle/single molecule(non-periodic) imaging	J. Hajdu* J. Miao H. Chapman	J. Arthur
(5) Atomic, molecular, and optical science	L. DiMauro* N. Berrah	J. B. Hastings

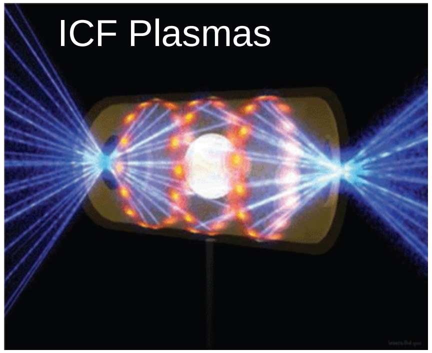
Needed to await American Recovery and Reinvestment Act of 2009

Also added *Soft X-Ray*, per Jo Stohr

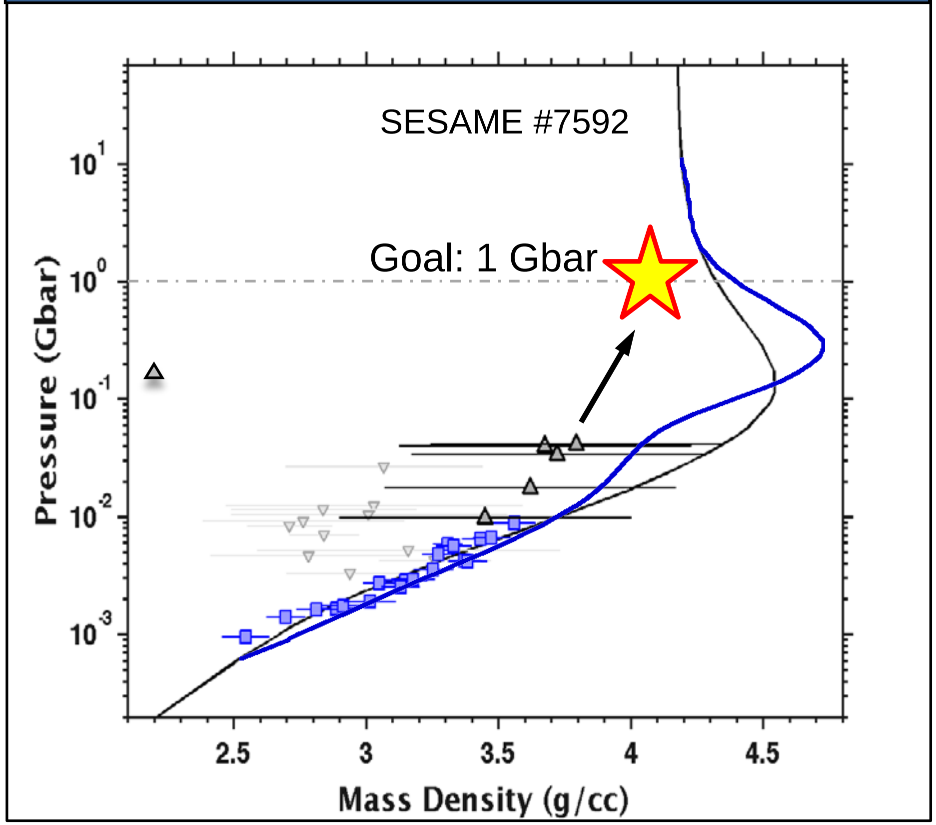
[illegible] Team leader

X-rays enable discoveries and validate models for materials in the most extreme conditions of high energy density (HED)

How pressure, temperature, and ionization are interrelated and affect structure



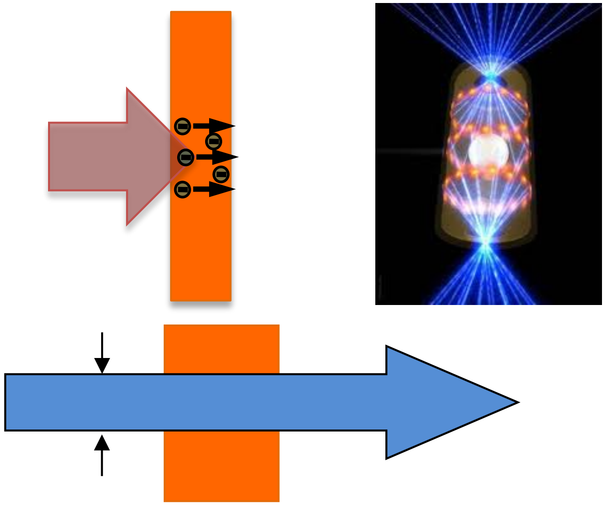
Hugoniot theory and data for CH



Both high energy lasers and x-ray FELS can <u>create</u> and <u>probe</u> materials under extremes of high temperature, pressure and density

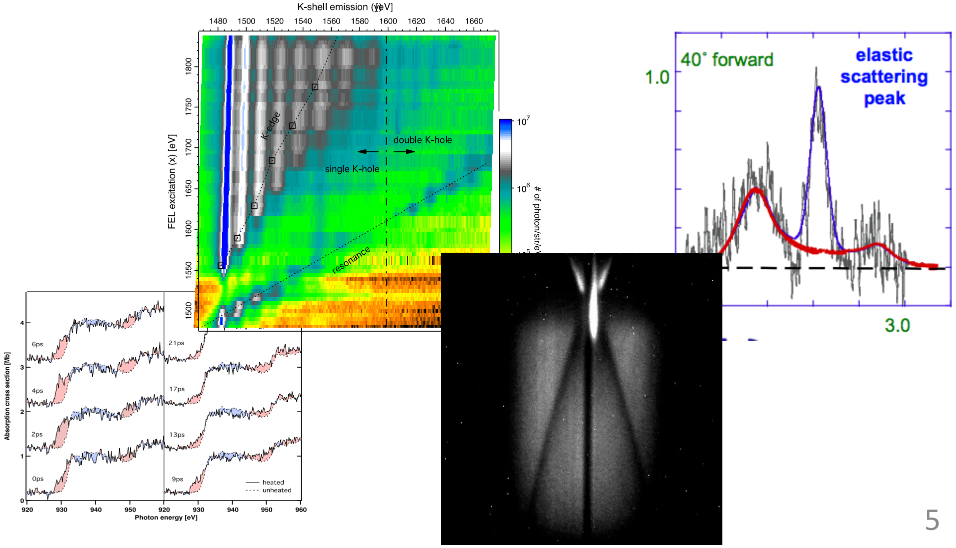
Create by:

- isochoric heating with short pulse laser
  or free-electron x-ray laser
- shock compression with long pulse laser
  or long pulse x-ray heating/ablation



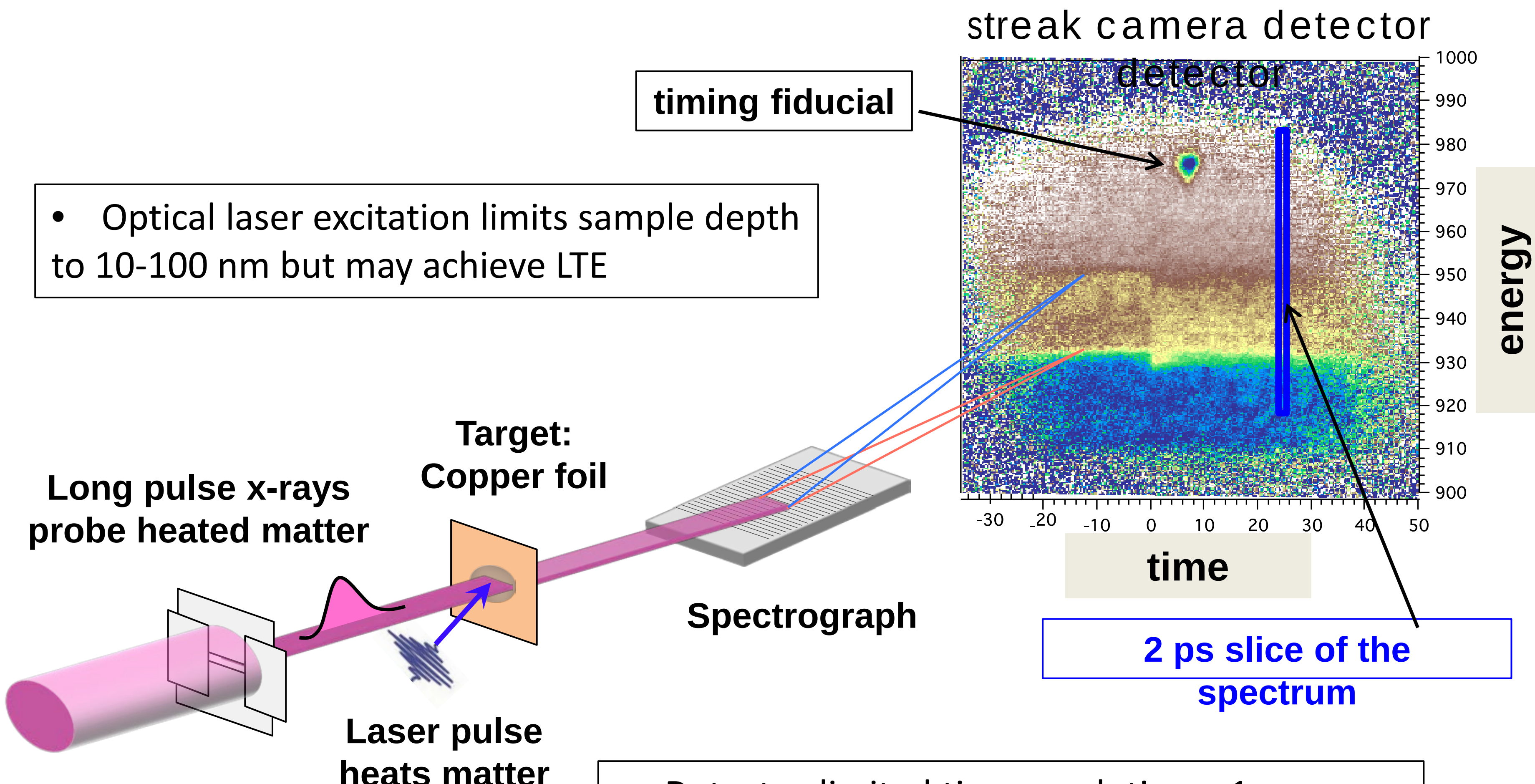
Probe with x-rays:

- near-edge absorption
- emission spectroscopy
- spatial imaging
- inelastic scattering



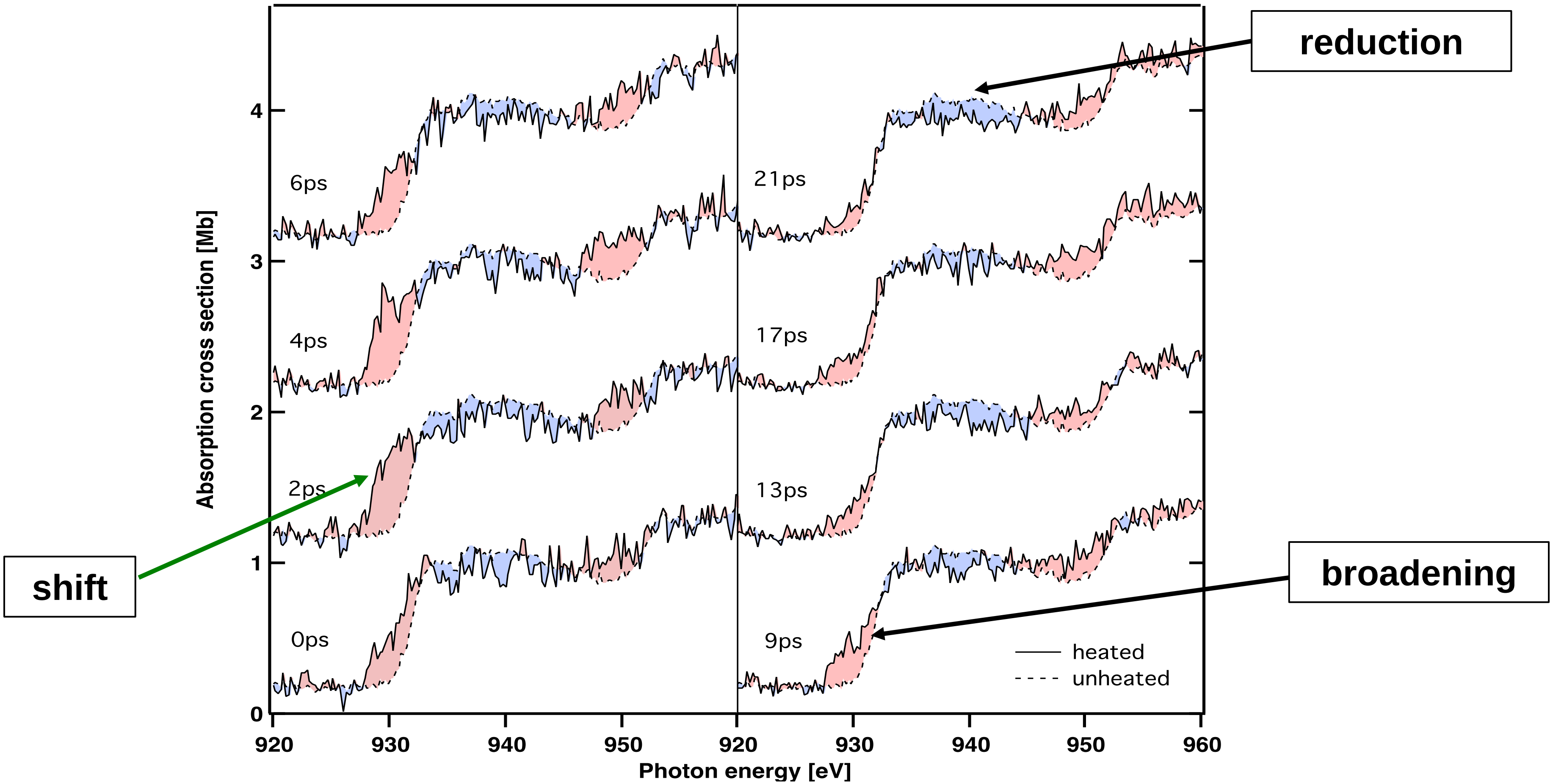
Electronic structure of hot and dense matter can be studied by near edge absorption spectroscopy

- Optical laser excitation limits sample depth to 10-100 nm but may achieve LTE



- Detector-limited time resolution ~ 1 ps
- Broad spectral line-width allows for NEXAFS

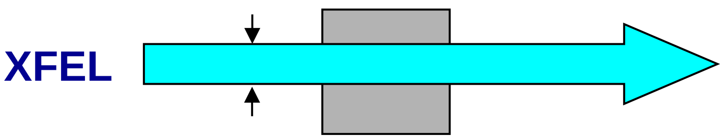
Changes in near edge x-ray absorption in hot and dense matter reveals modified density of states



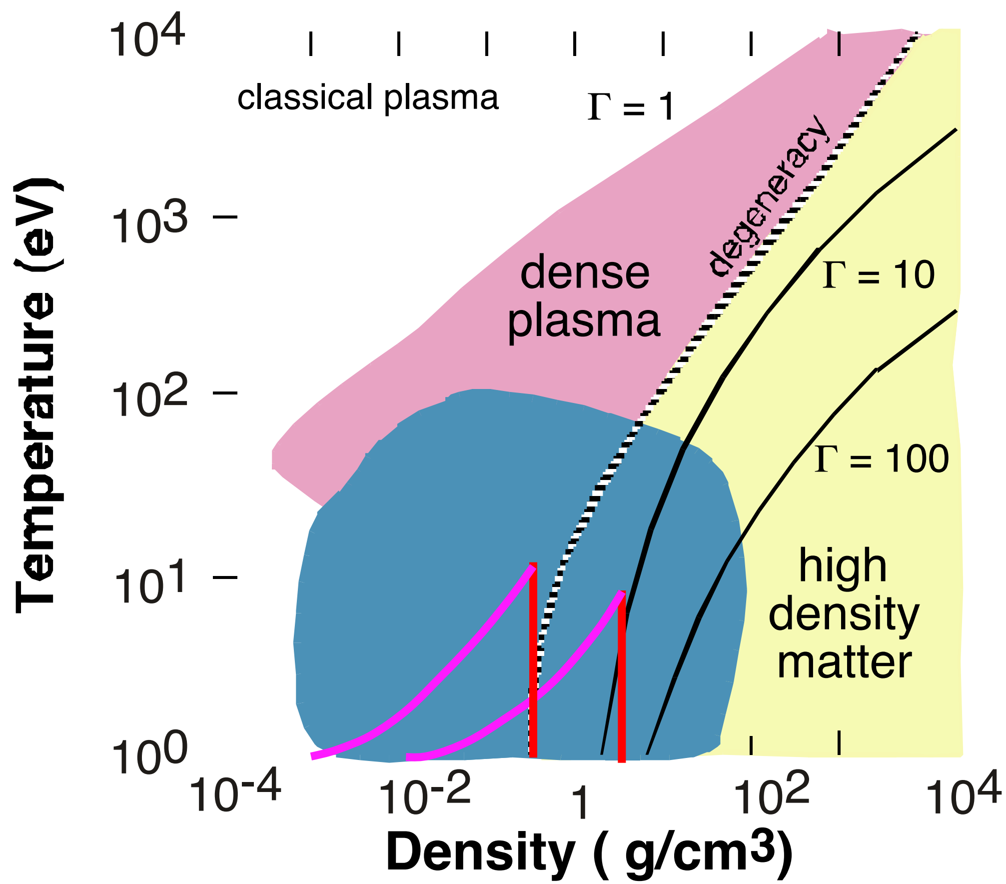
- e.g., time (temperature) dependent electron-phonon coupling, phase transitions
- Optimal x-ray probe source bandwidth = 10 %, beyond current FELs!

HED matter can be created by isochoric heating using x-ray FELs (with isentropic expansion to sample the phase space)

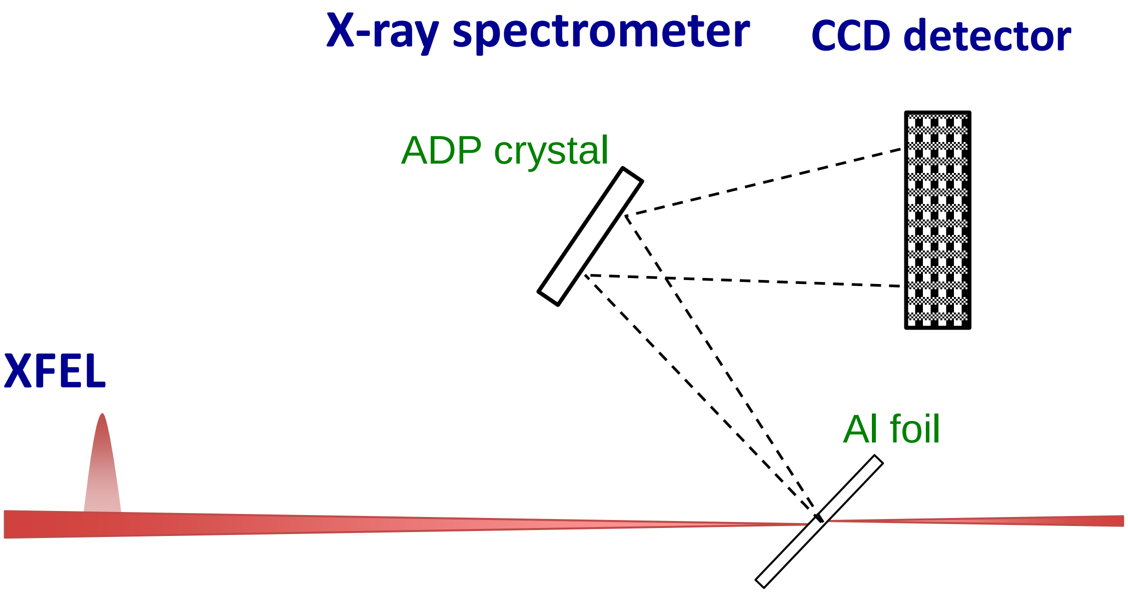
X-rays can create greater uniformity and depth than optical pulses, with relatively hot initial electrons

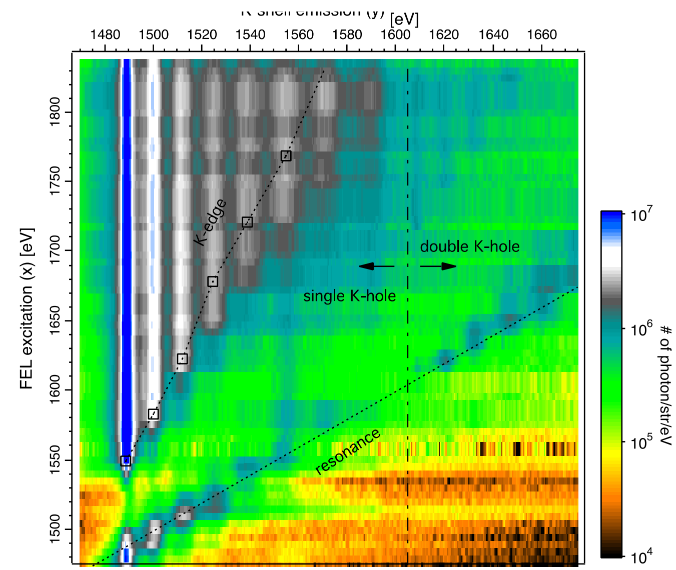


- XFEL can heat matter rapidly
  - Isochores (constant ρ)
  - Isentropes (constant entropy)
- Starting with underdense foams allows complete sampling
  - Isochores (constant ρ)
  - Isentropes (constant entropy)



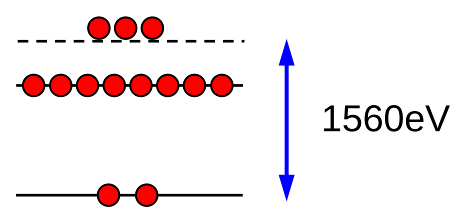
K-alpha emission from ion stages and resonant x-ray scattering reveal novel plasma conditions





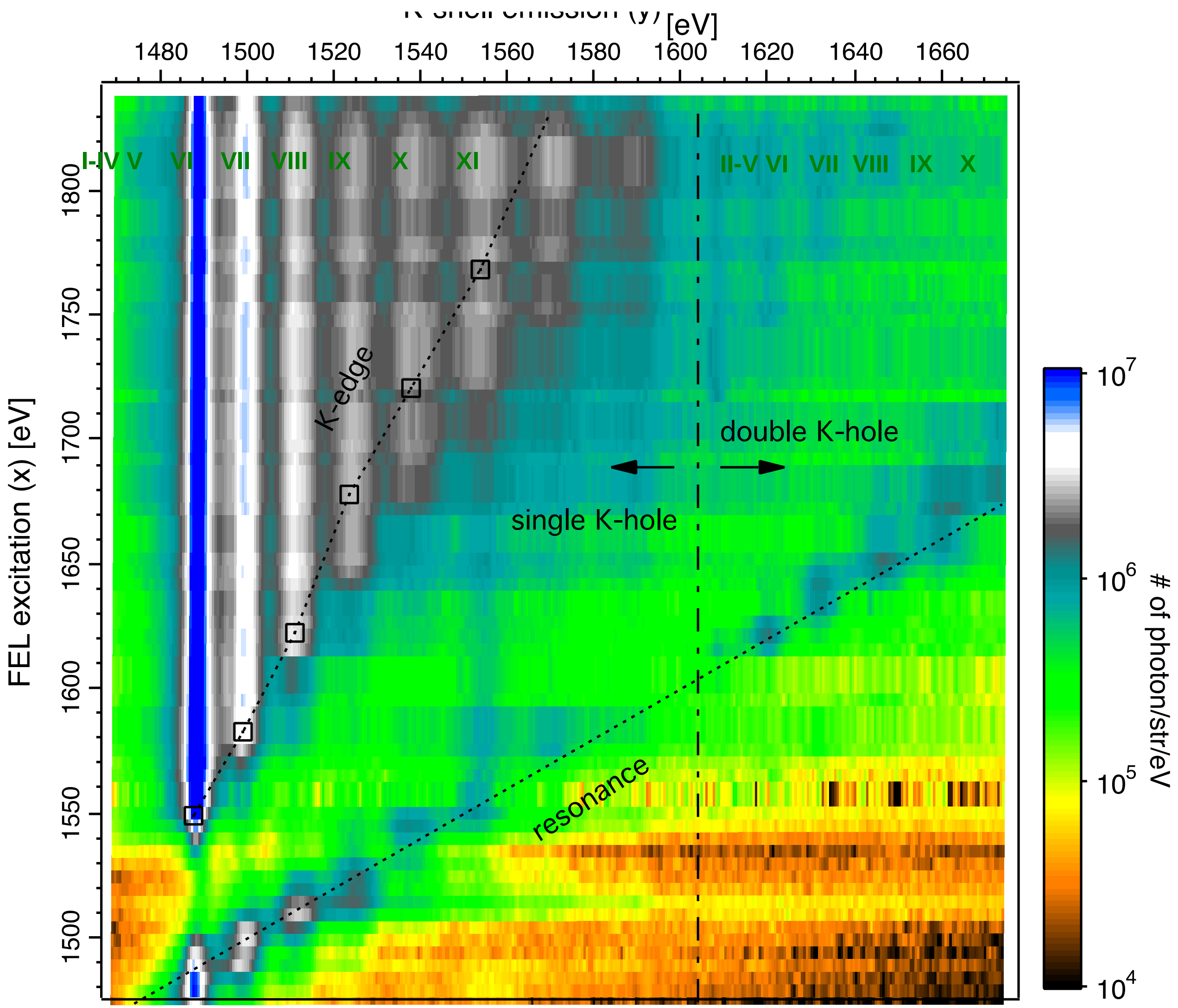
Vinko, et al Nature 482, 59-62 (2011)

multiple photons absorbed per atom per pulse



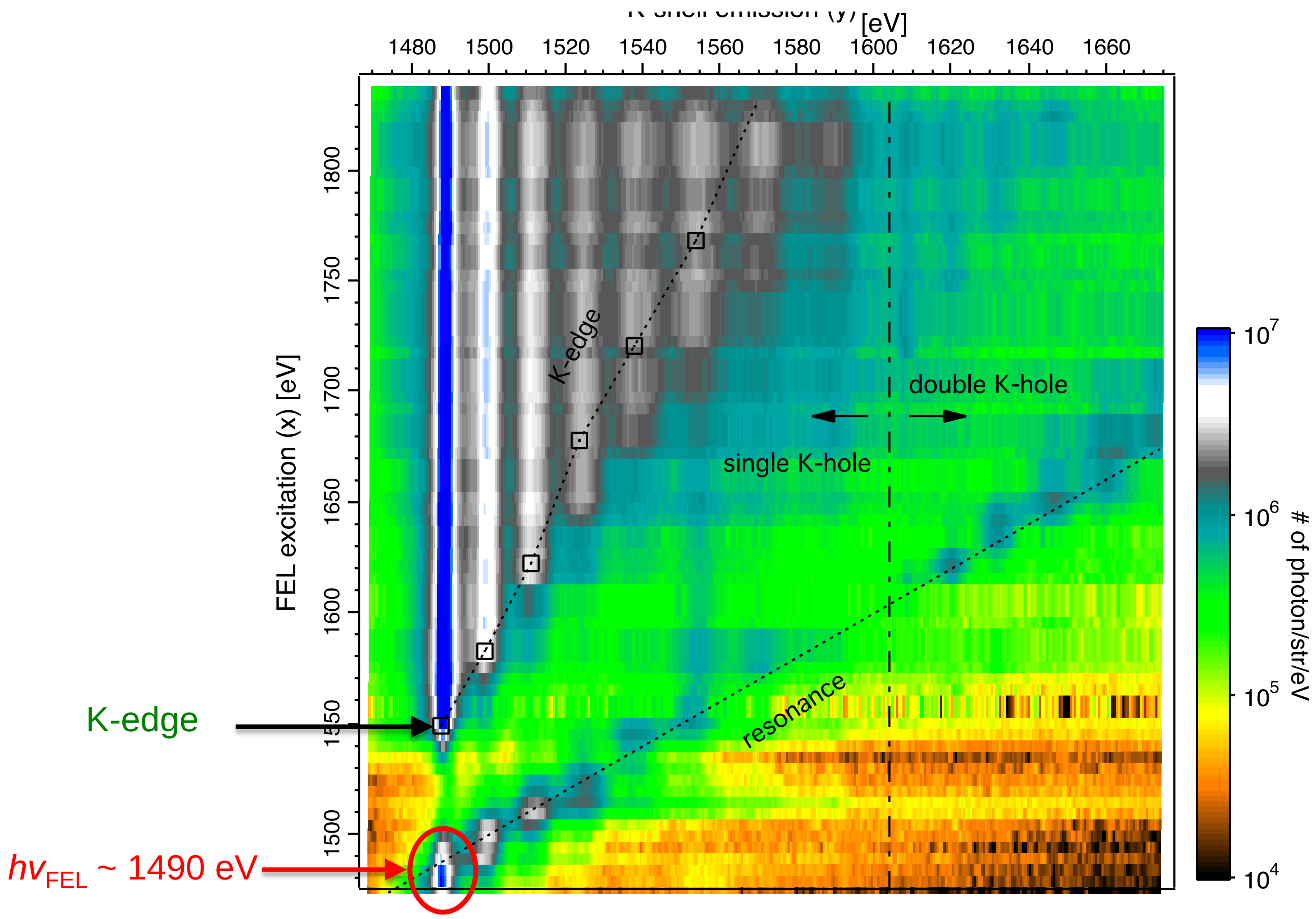
Collaboration: Oxford, LBNL, IAEA Vienna, Austria, AWE Aldermaston, Institute of Physics ASCR, Czech Republic, Physics UC Berkeley, SLAC, LLNL, DESY, Hamburg, Friedrich-Schiller-Universitat. Jena

Two types of emission patterns are observed: K-alpha from ion stages and resonance scattering



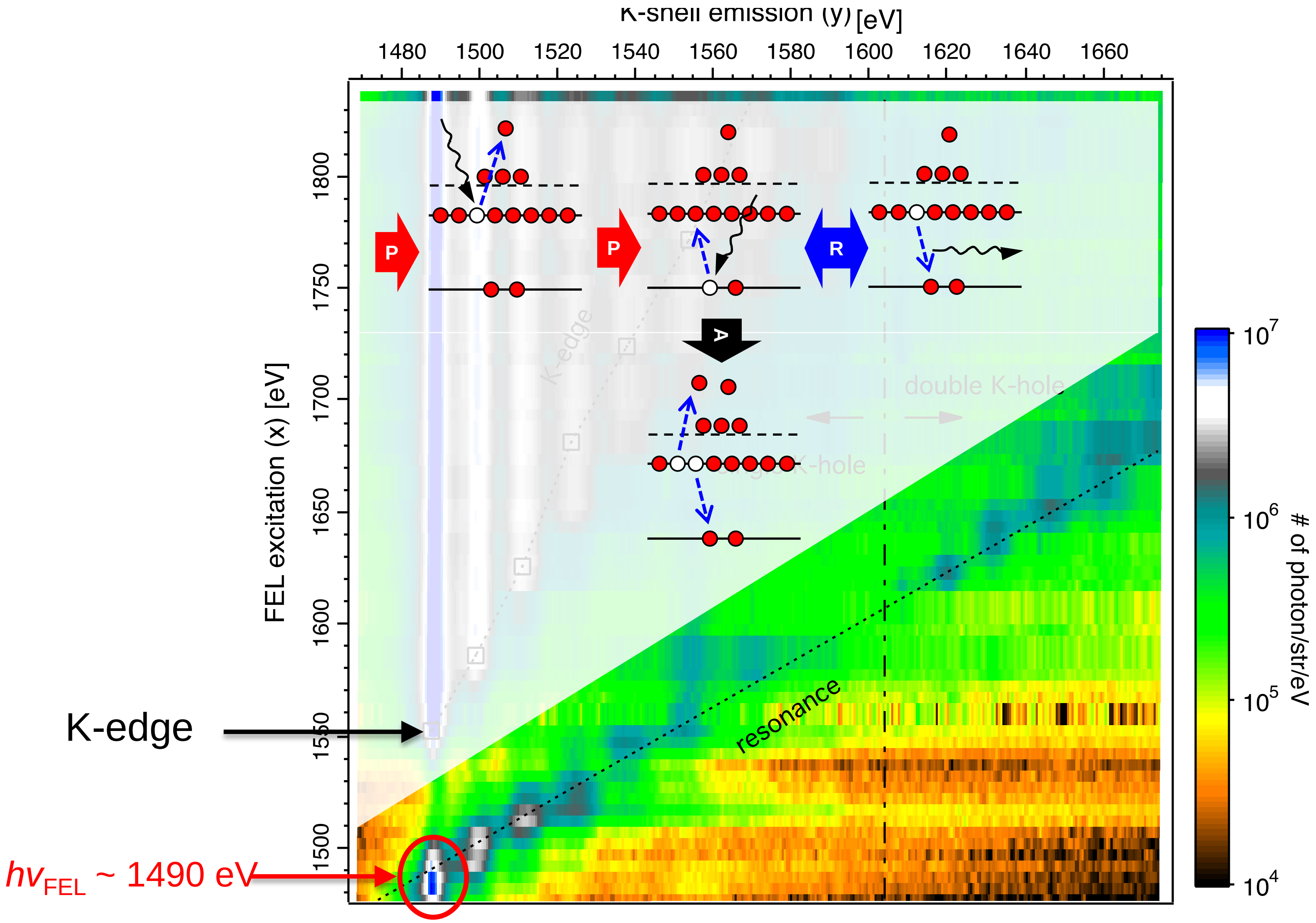
Vinko, *et al* Nature

High fluence x-ray pulse opens inner shells that are inaccessible via single photon process

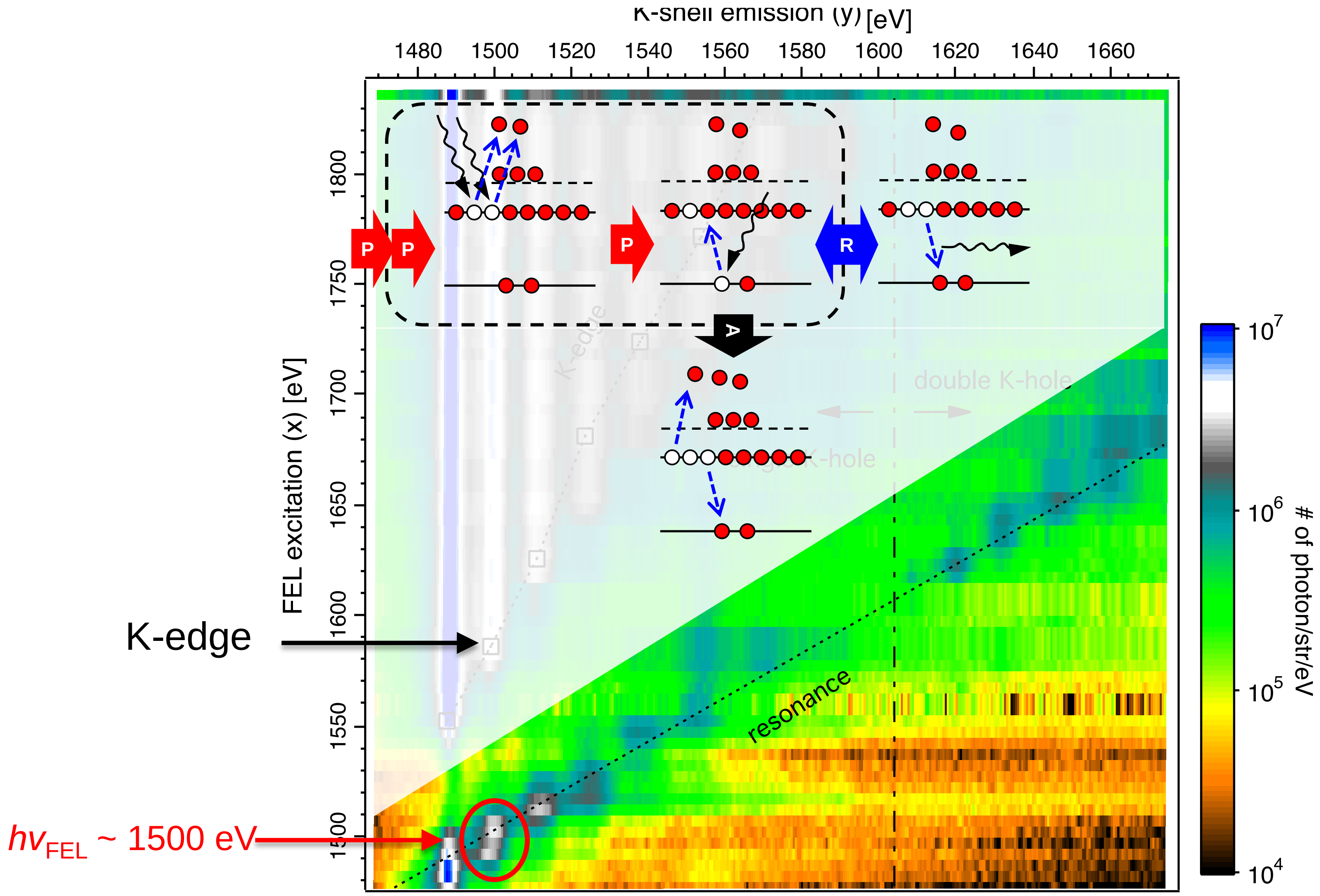


Cho, *et al* PRL

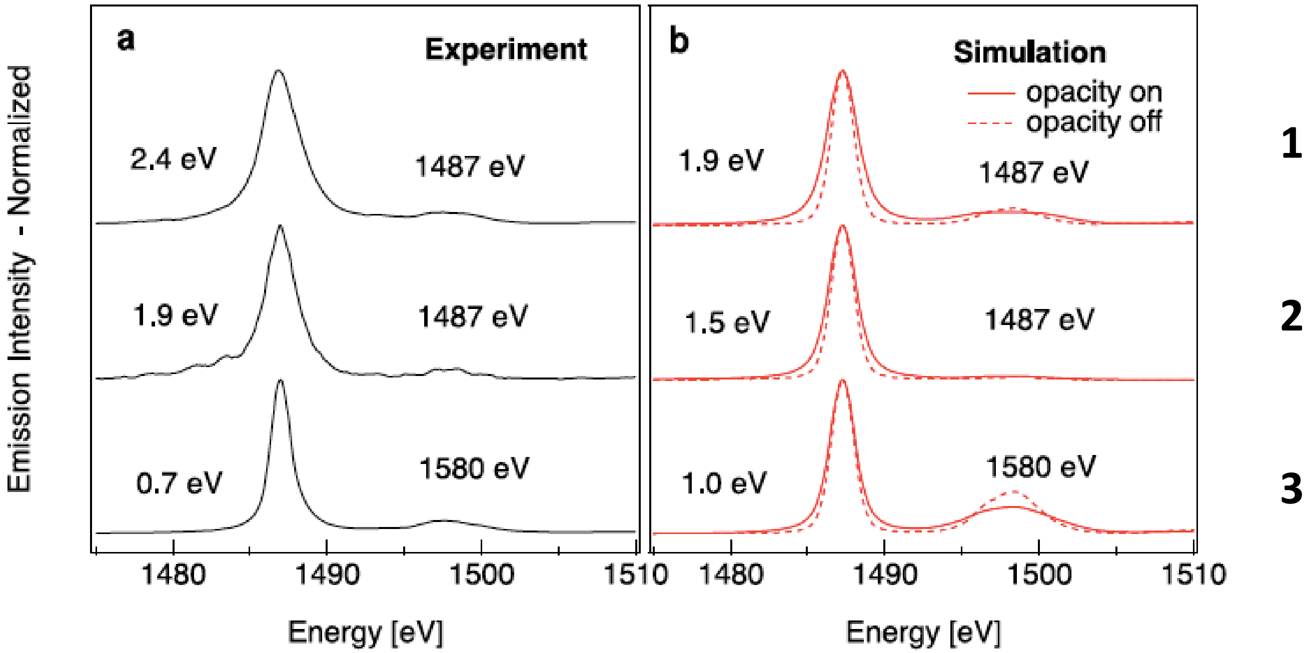
High fluence x-ray pulse opens inner shells inaccessible via a single photon



Hidden resonance revealed through multi-photon processes



Emission spectra sensitive to FEL-intensity-dependent opacity effects: L-shell holes reabsorb K-alpha

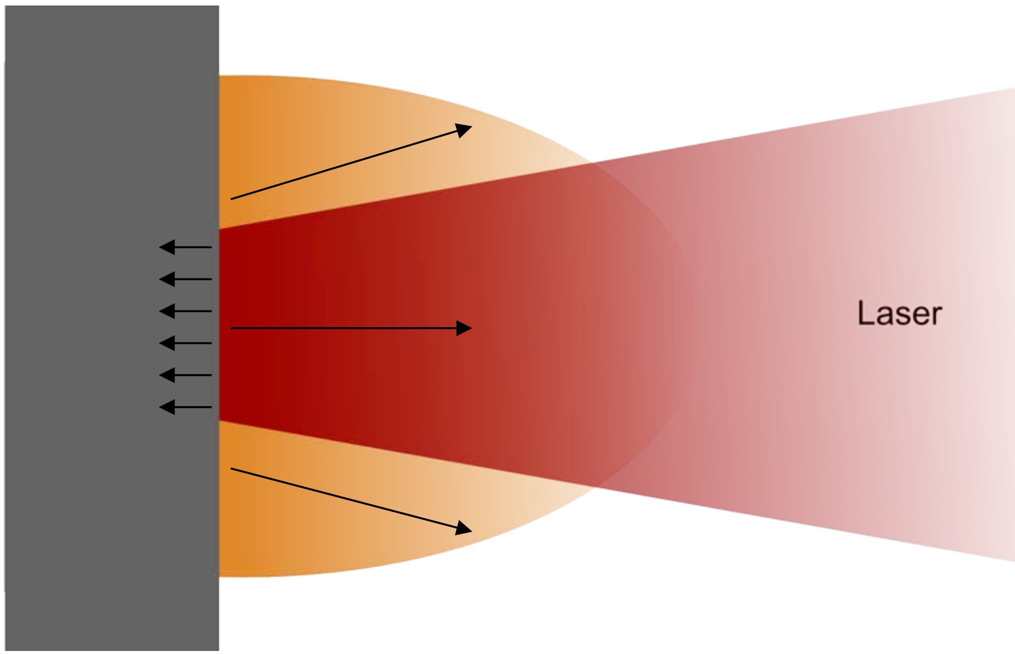


Experimental line shapes of main Kα emission

(1a) resonant pumping (1487 eV) with X-ray intensity: 1×10^{17} W/cm^2
(2a) resonant pumping: 0.7×10^{17} W/cm^2
(3a) non-resonant pumping (1580 eV)

(b) SCFLY simulations with equivalent conditions: with (solid) & without (dashed) opacity

Creating high pressures: nanosecond-laser-driven shock waves can create Mbar to multi-Mbar pressures



Intensity ~10^{13} W/cm^2

Ablation pressure:
P ~ 200 GPa = 2×10^6 bar

→ shock wave

Rankine-Hugoniot relatios:

$$\rho_1 u_1 = \rho_2 u_2$$

$$\rho_1 u_1^2 + P_1 = \rho_2 u_2^2 + P_2$$

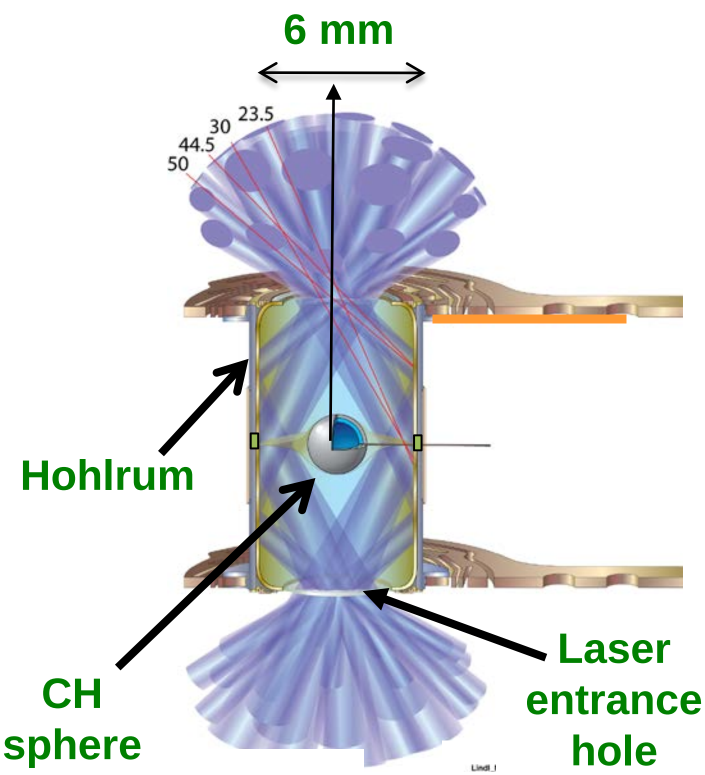
$$u_1(\rho_1 \epsilon_1 + \frac{\rho_1 u_1^2}{2} + P_1) = u_2(\rho_2 \epsilon_2 + \frac{\rho_2 u_2^2}{2} + P_2)$$

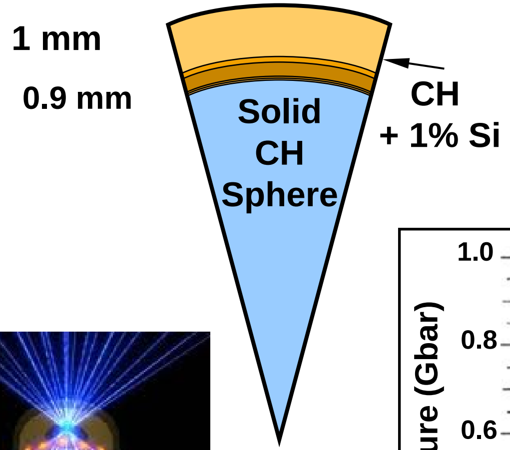
Measure 2 quantities to solve system:

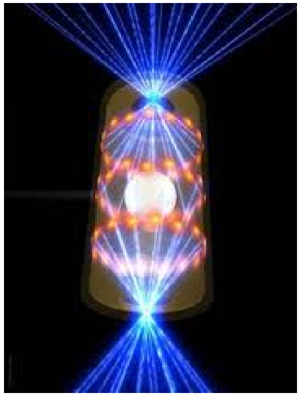
e.g. u_1, u_2 or u_1, ρ

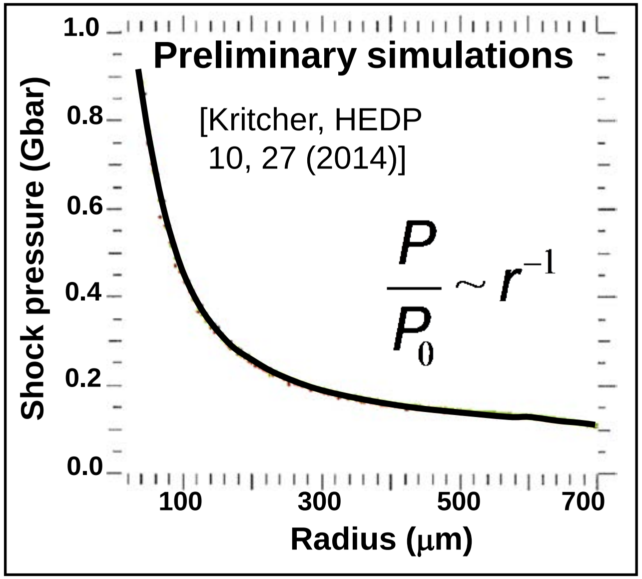
Highest energy lasers (e.g., NIF) can produce the highest pressure and density with spherical converging shocks

Use hohlraum (hollow can) with spherical material target in center





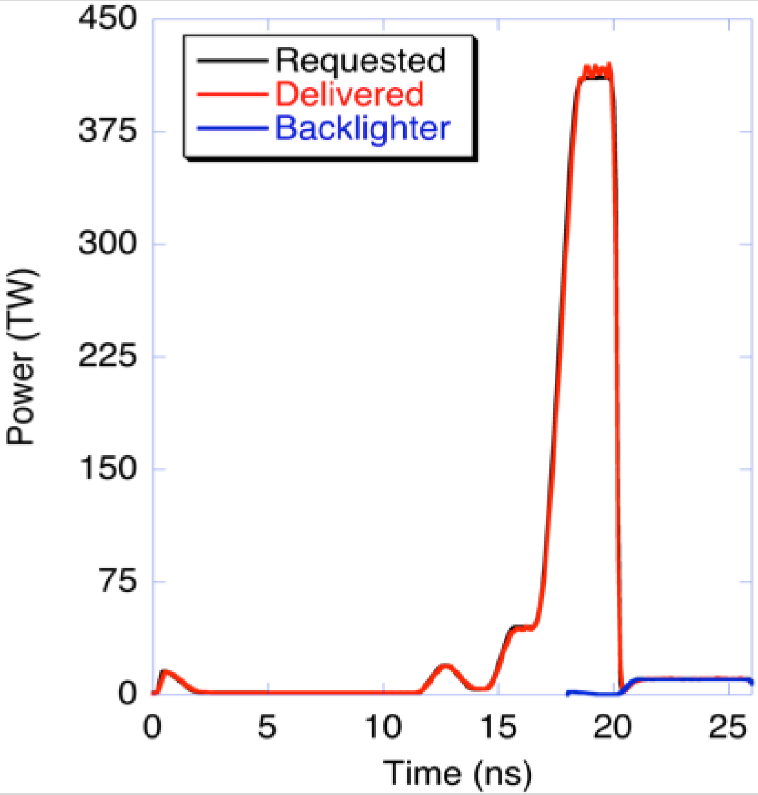




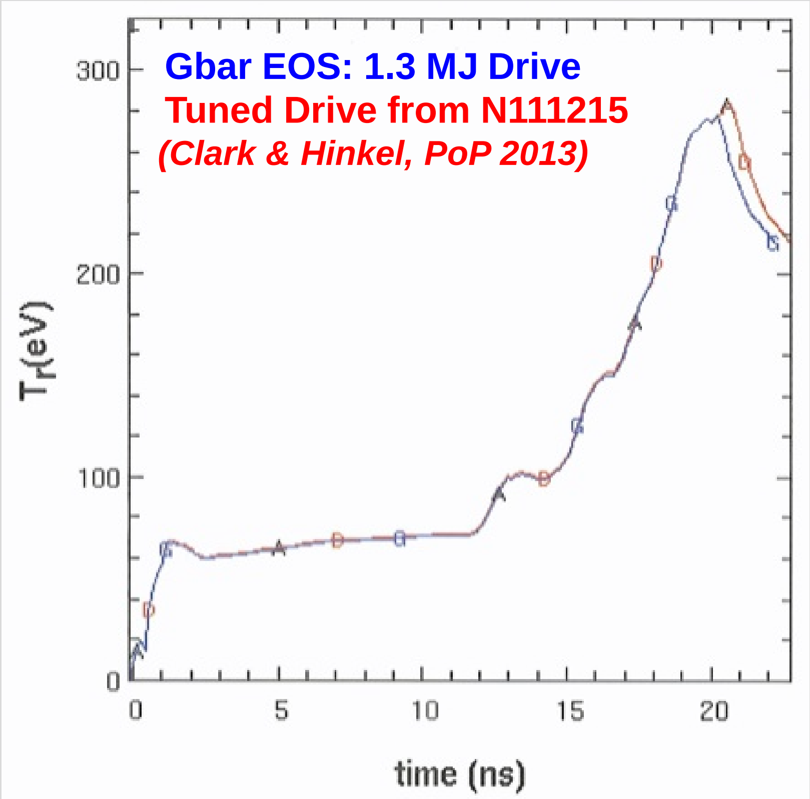
- 192 laser beams focused inside cm-sized hohlraum to > 200 eV BB temperature
- Surface of a mm-sized target in the center vaporizes, and ablates
- Converging spherical shock wave generates Gbar pressures and compresses the target

Hohlraum peak radiation temperature is ~ 280 eV

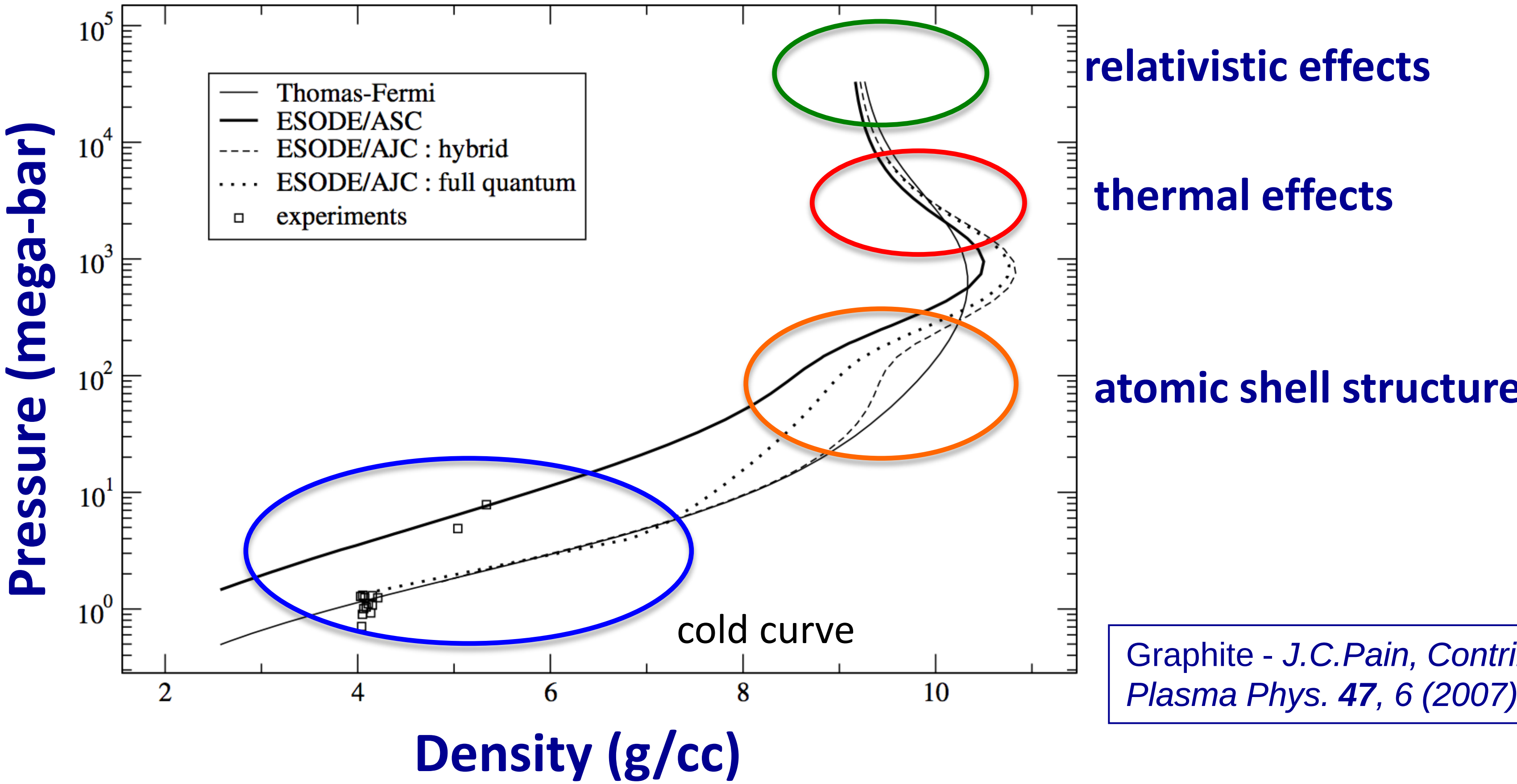
Laser Pulse Shape



Radiation Drive



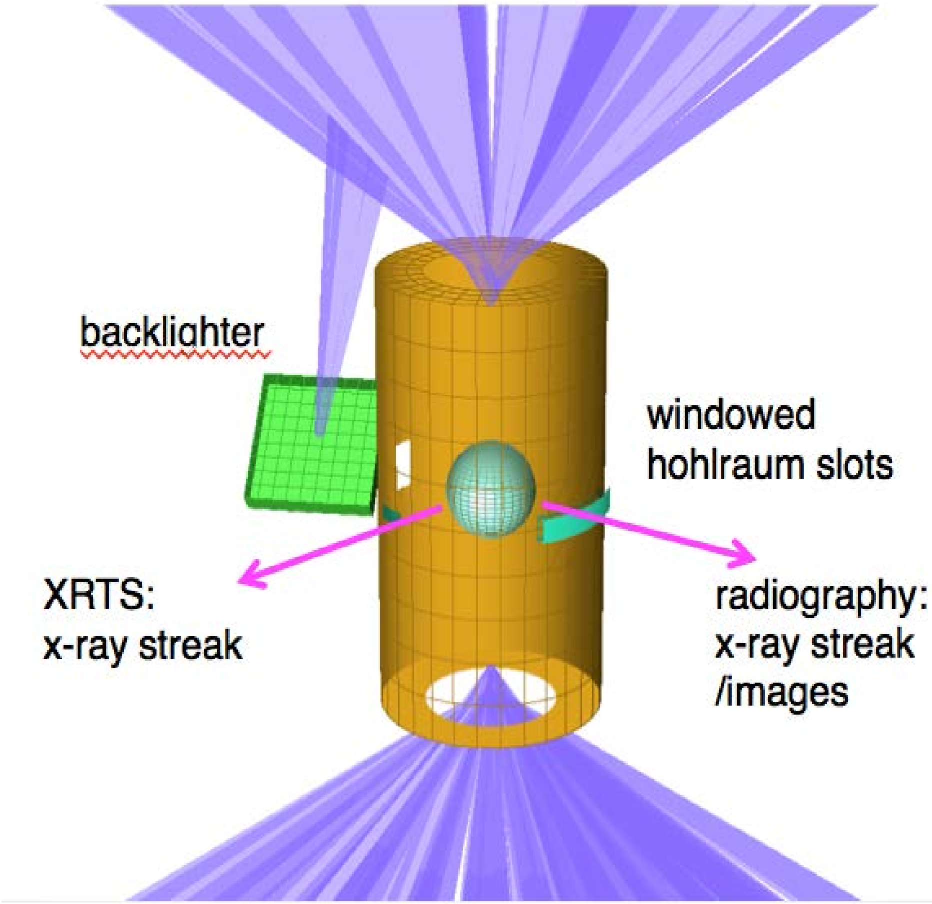
By measuring shock speed and density determine equation of state of material at GBar pressures



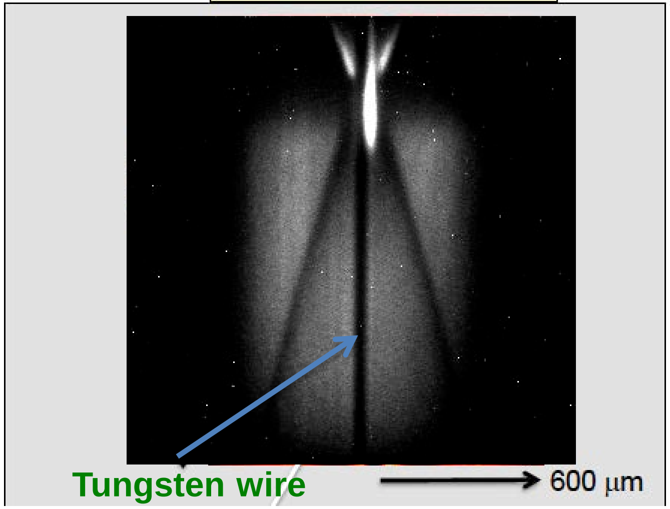
Graphite - *J.C.Pain, Contrib. Plasma Phys.* ***47***, *6 (2007)*

- **Important for understanding planetary & stellar physics, inertial confinement fusion**
- **Little data (not adequate to constrain models; average atom vs detailed configuration)**

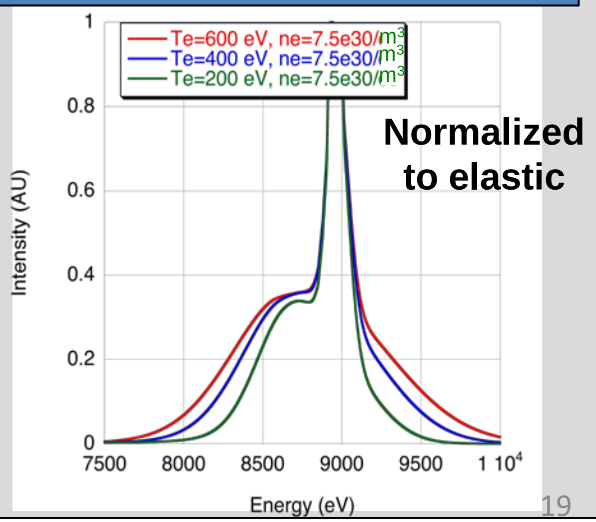
Measurements at high pressures include simultaneous x-ray radiography and inelastic x-ray scattering



Composite "streak"



X-ray Thomson Scattering



- **Streaked radiographic imaging of shock**
- **X-ray scattering from compressed matter**

Plasma X-Ray Source for Backlighting and Thomson Scattering

Current platform

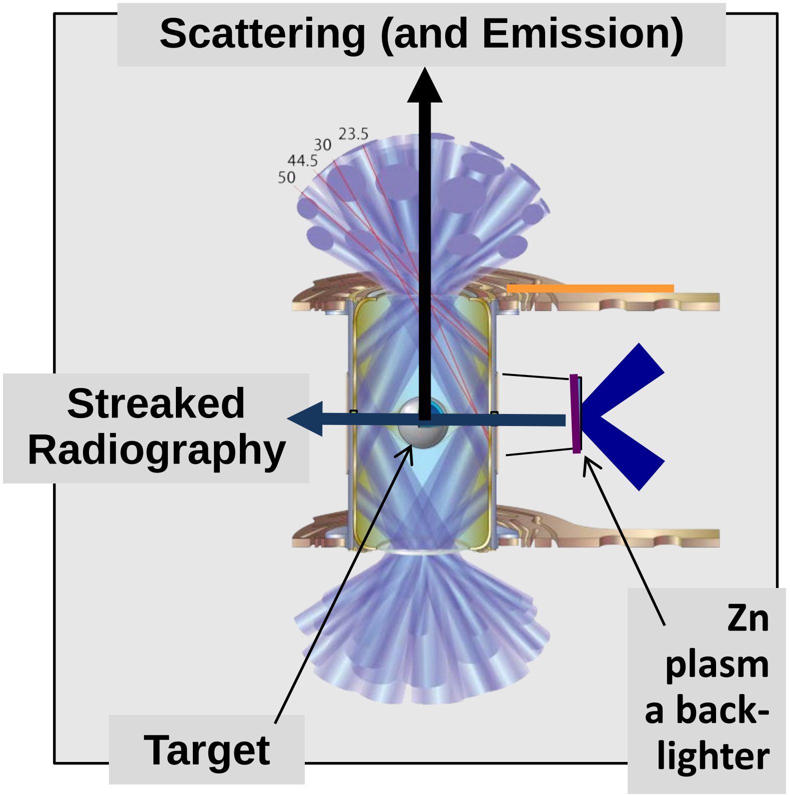
16 x TW beams on Zn foil = 100 kJ
1% conversion to 9 keV x-rays = 1 kJ / 4pi
Source distance from sample = 7.5 mm
Solid angle subtended = 0.006 sr
X-rays on sample = 0.5 J
Gated-imager sets integration time = 100 ps

Improved platform

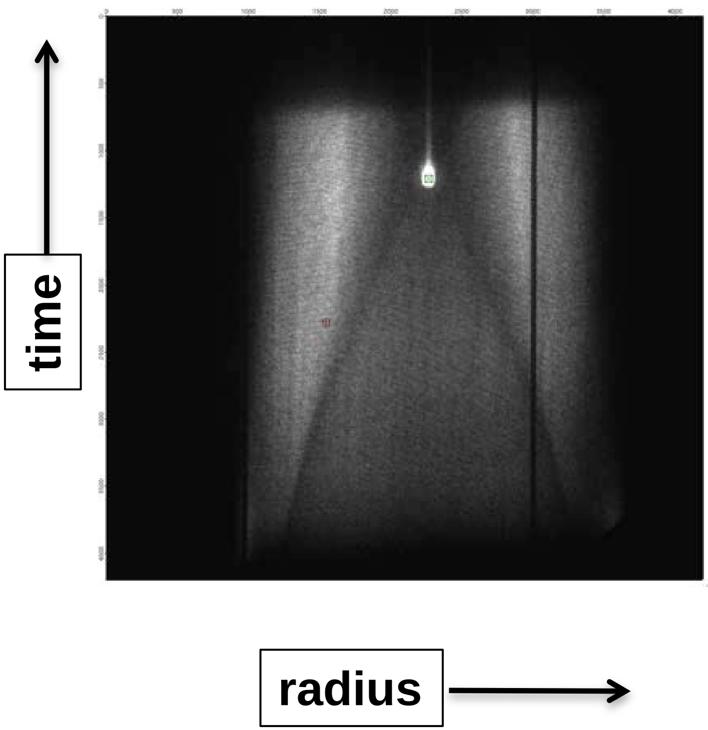
Source 2.5 mm from sample
4x improvement in collected x-ray power

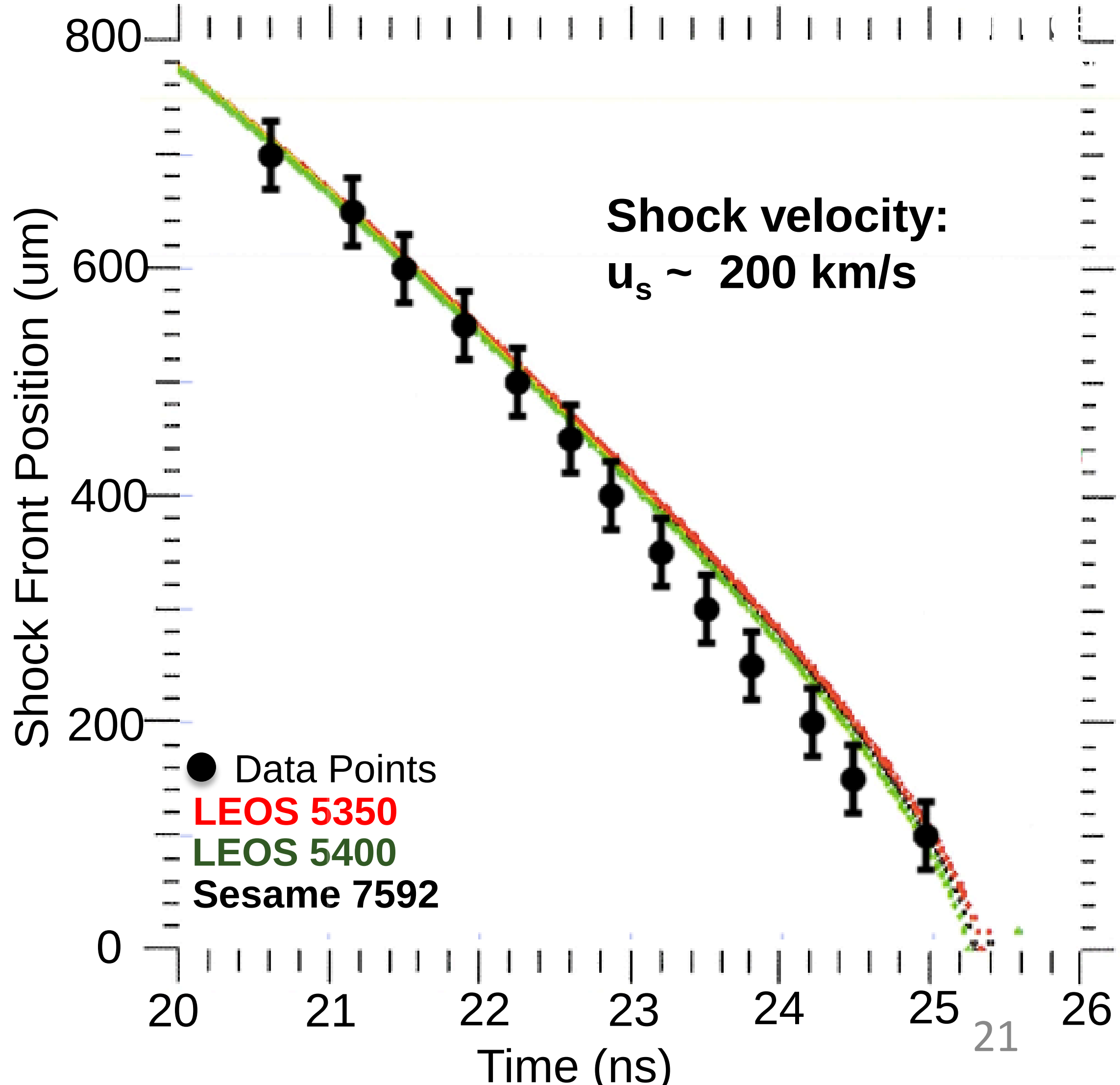
- **0.5 GW x-ray beam**
- **On 200 micron sample at shock coalescence with GBar pressure**
- **50 mJ x-rays in 100 ps gating time**

  ....but still... 100 ps gating time



Shock speed appears higher than predicted





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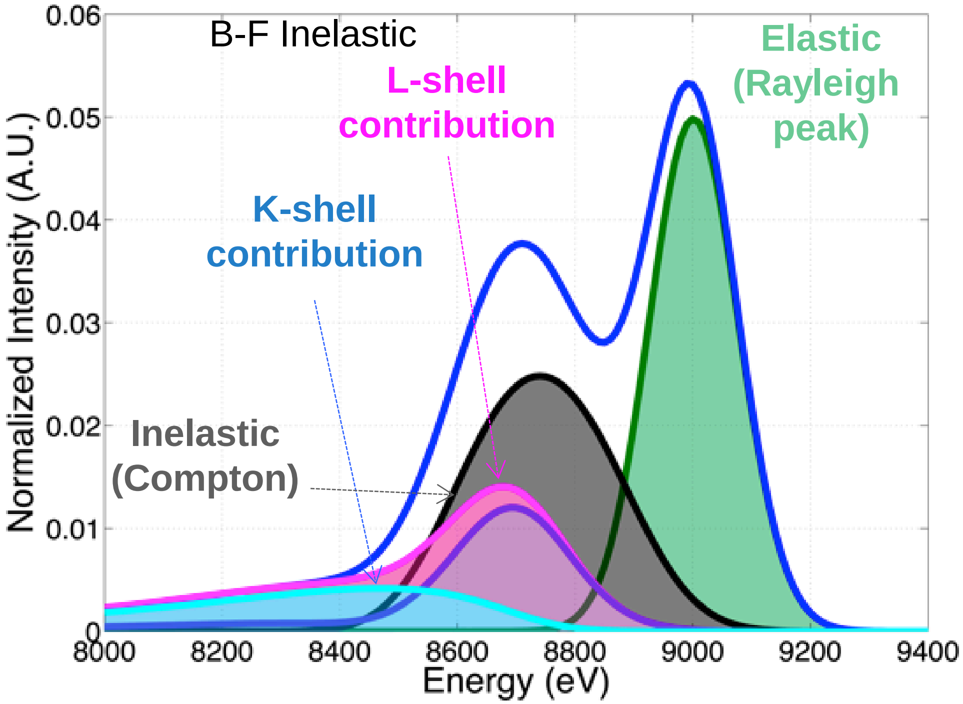
Analyzing x-ray Thomson scattering data

Contributions to the dynamic structure factor:

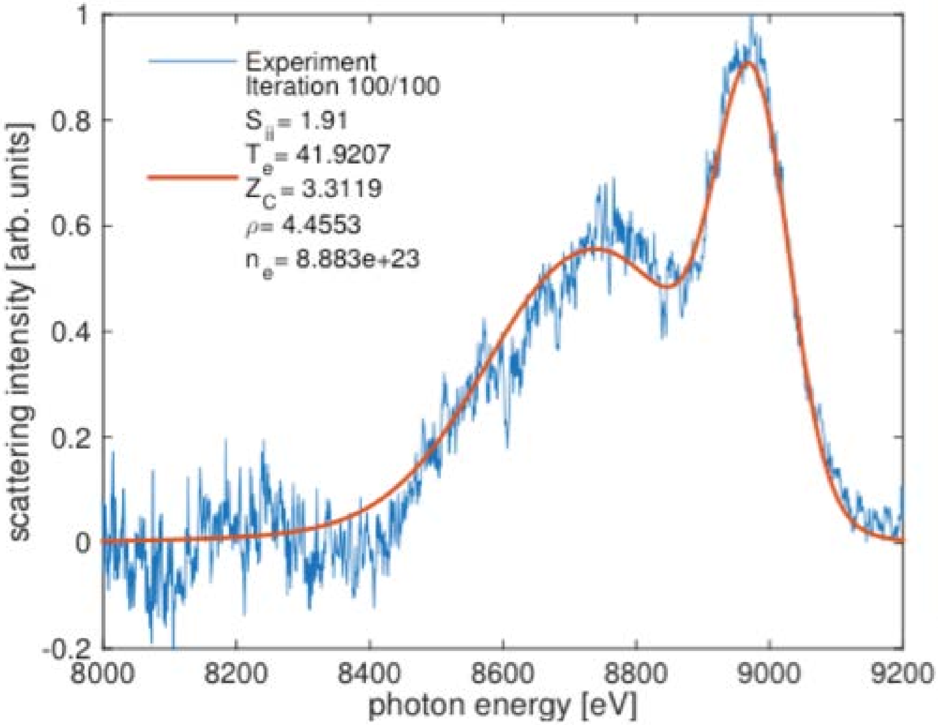
ion feature | electron feature | bound-free feature

$$S(k,\omega) = |f_I(k) + q(k)|^2 S_{ii}(k,\omega) + Z_f S^0_{ee}(k,\omega) + Z_c \int \tilde{S}_{ce}(k,\omega-\omega')S_s(k,\omega')d\omega'$$

Z_f =free e$^-$; Z_c=core e$^-$; f_i = ion form factor; $q(k)$= free & valence e$^-$ screening cloud



Early inelastic scattering results from CH

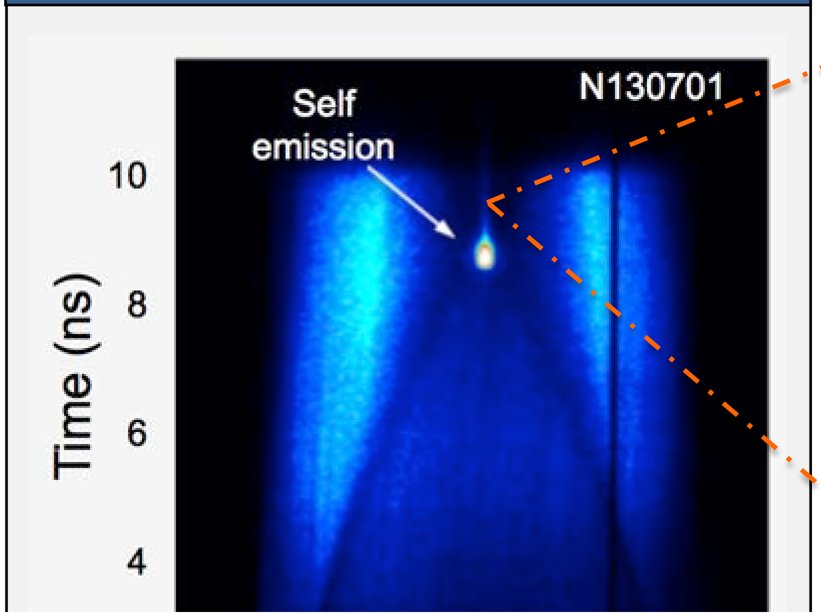


X-ray scattering:

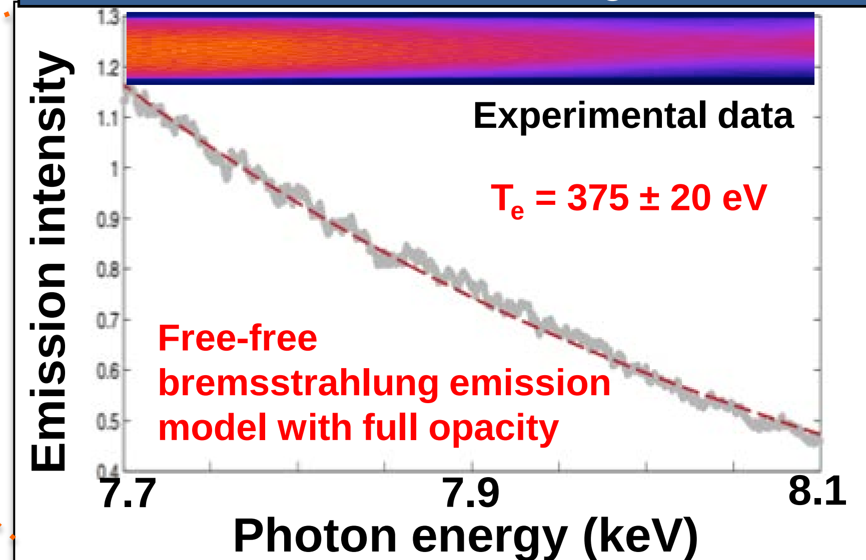
- temperature
- Ionization
- 4 g/cc,
- Ne = 8 E23
- Te = 40 eV
- Integration to r = 500 um

Temperature measurement at from core at 30 g/cc CH ~ 375 eV from Bremsstrahlung consistent with modeling

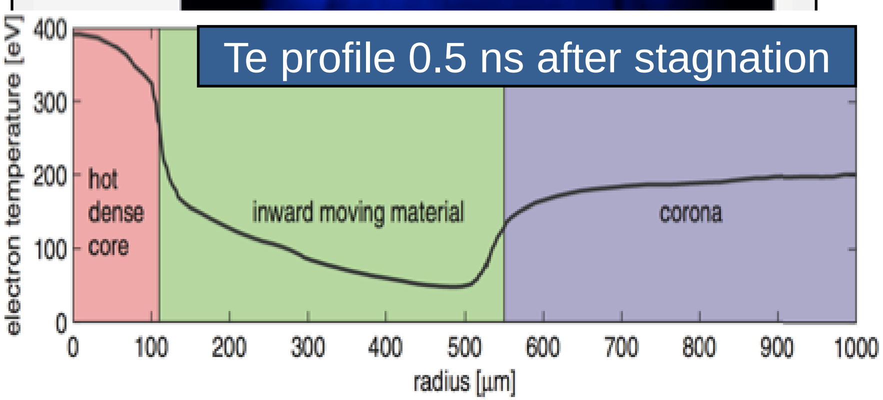
Streaked radiography data (CH)



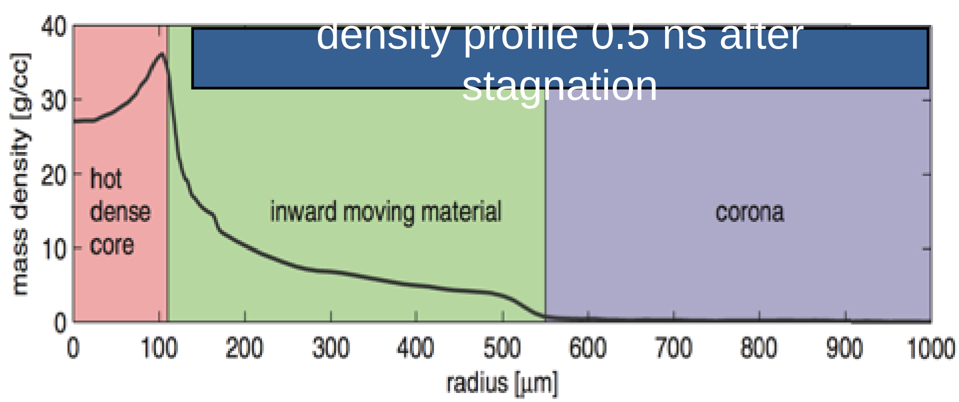
Bremsstrahlung emission spectrum 0.5 ns after shock stagnation



Te profile 0.5 ns after stagnation

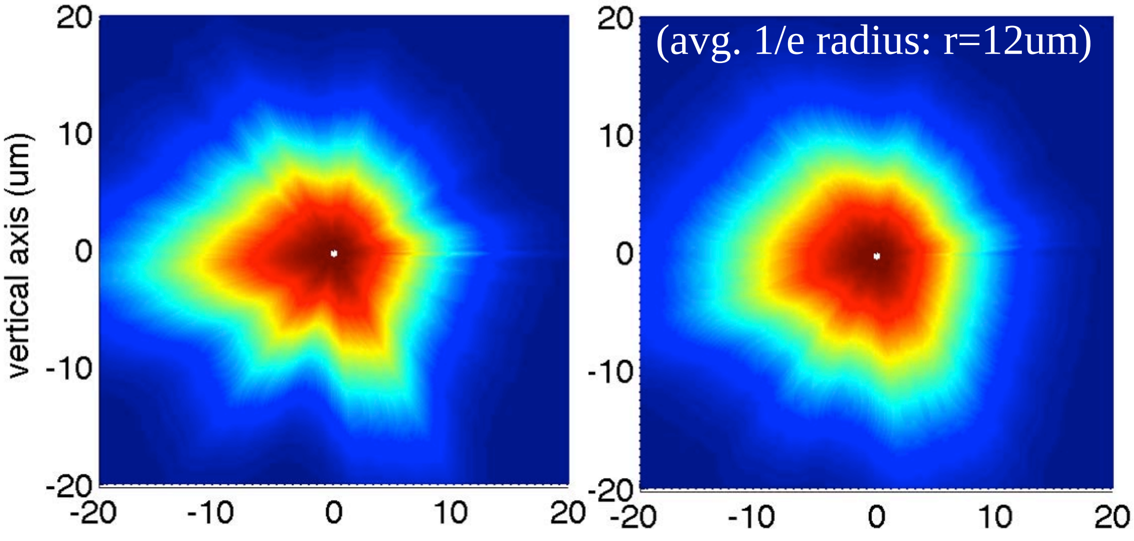


density profile 0.5 ns after stagnation



Kraus et al. RSI 85, 11D606 (2014)

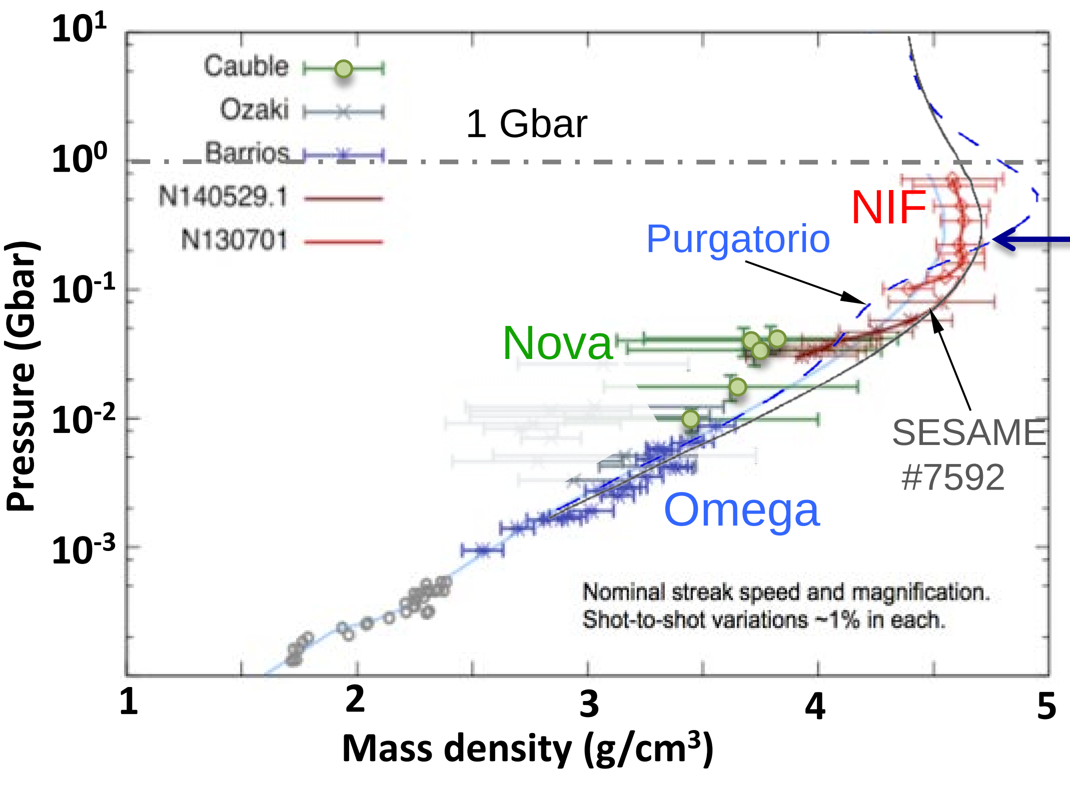
Hot spot measurement by penumbral imaging



Small and round emitting area indicates symmetric shock convergence

Analytical reconstruction
(assume error-function shape for circular lineouts)

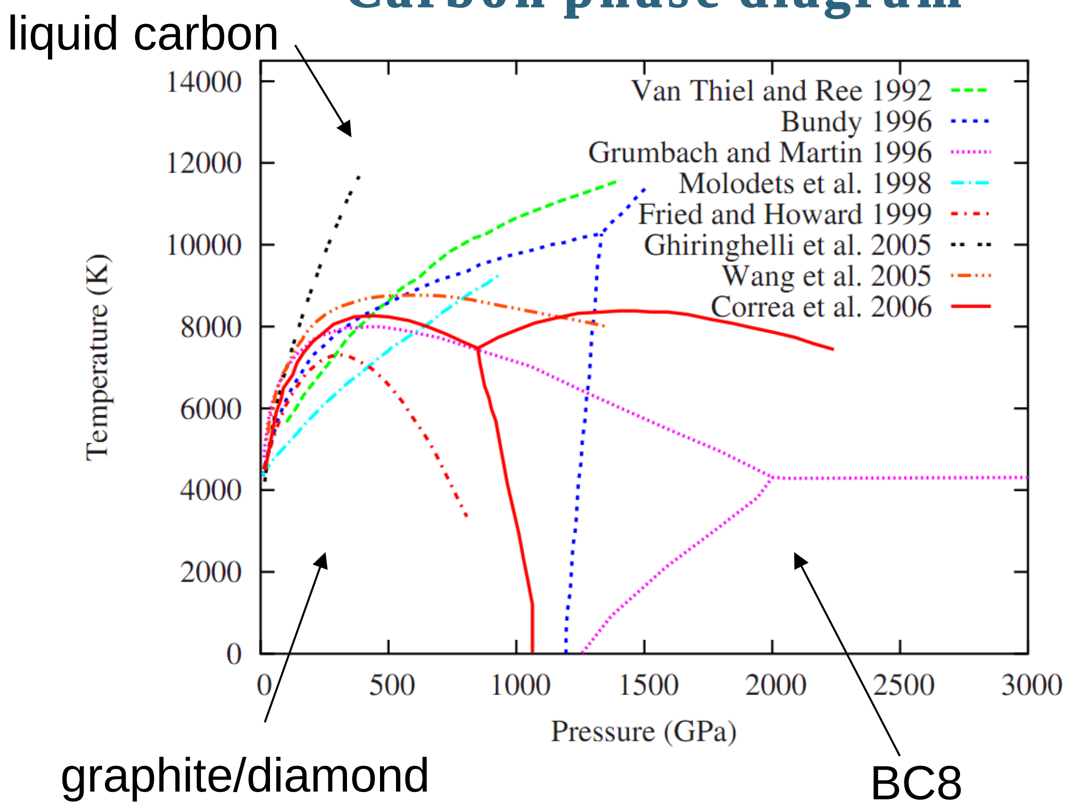
Conclude: high energy lasers can reach the Gbar regime of material performance; diagnostics require x-ray probes



Carbon K-shell Ionization effects

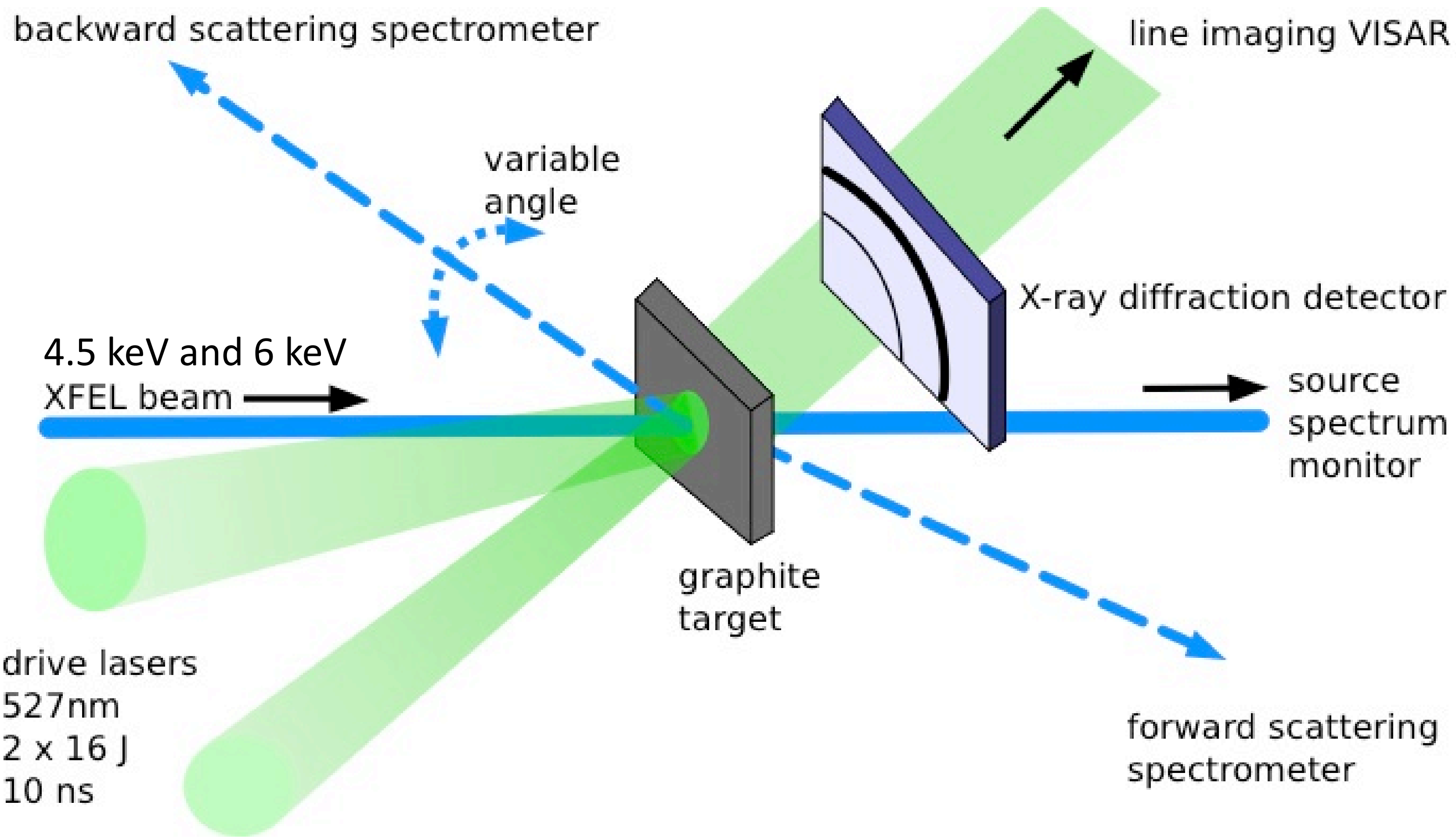
LCLS experiment | At lower pressure, can observe shock-induced transition from graphite to warm dense diamond and liquid carbon

Carbon phase diagram



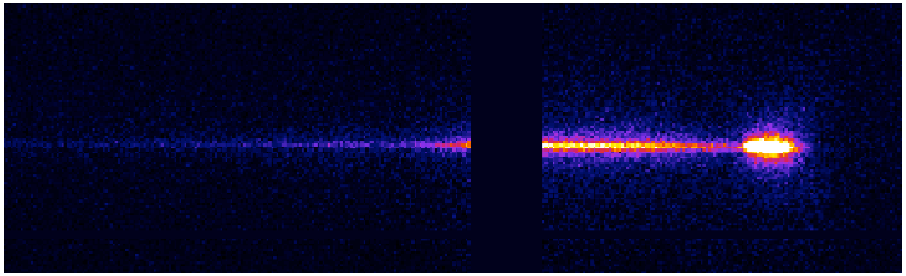
Correa et al. Phys. Rev. B 78, 024101 (2008)

LCLS experiment | Setup

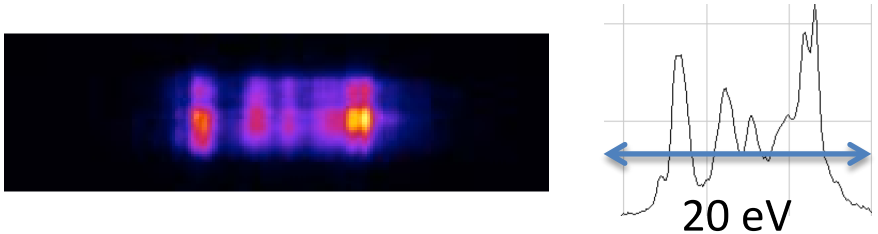


LCLS experiment | High quality scattering spectra – single shot

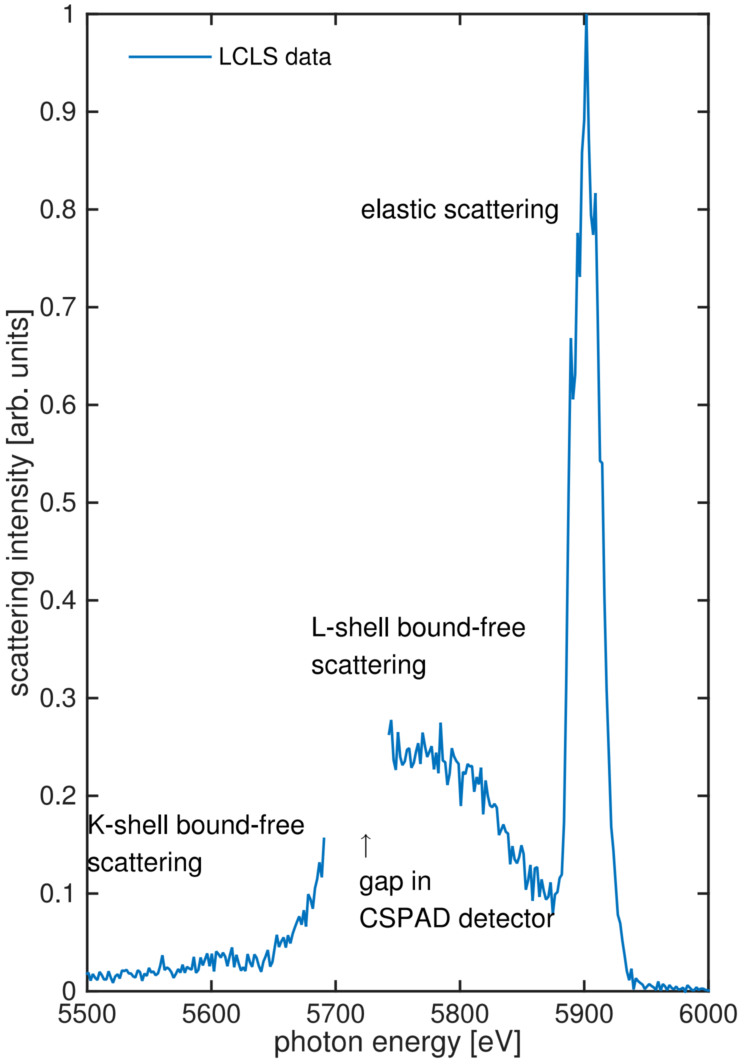
XRTS: HAPG in von Hamos geometry



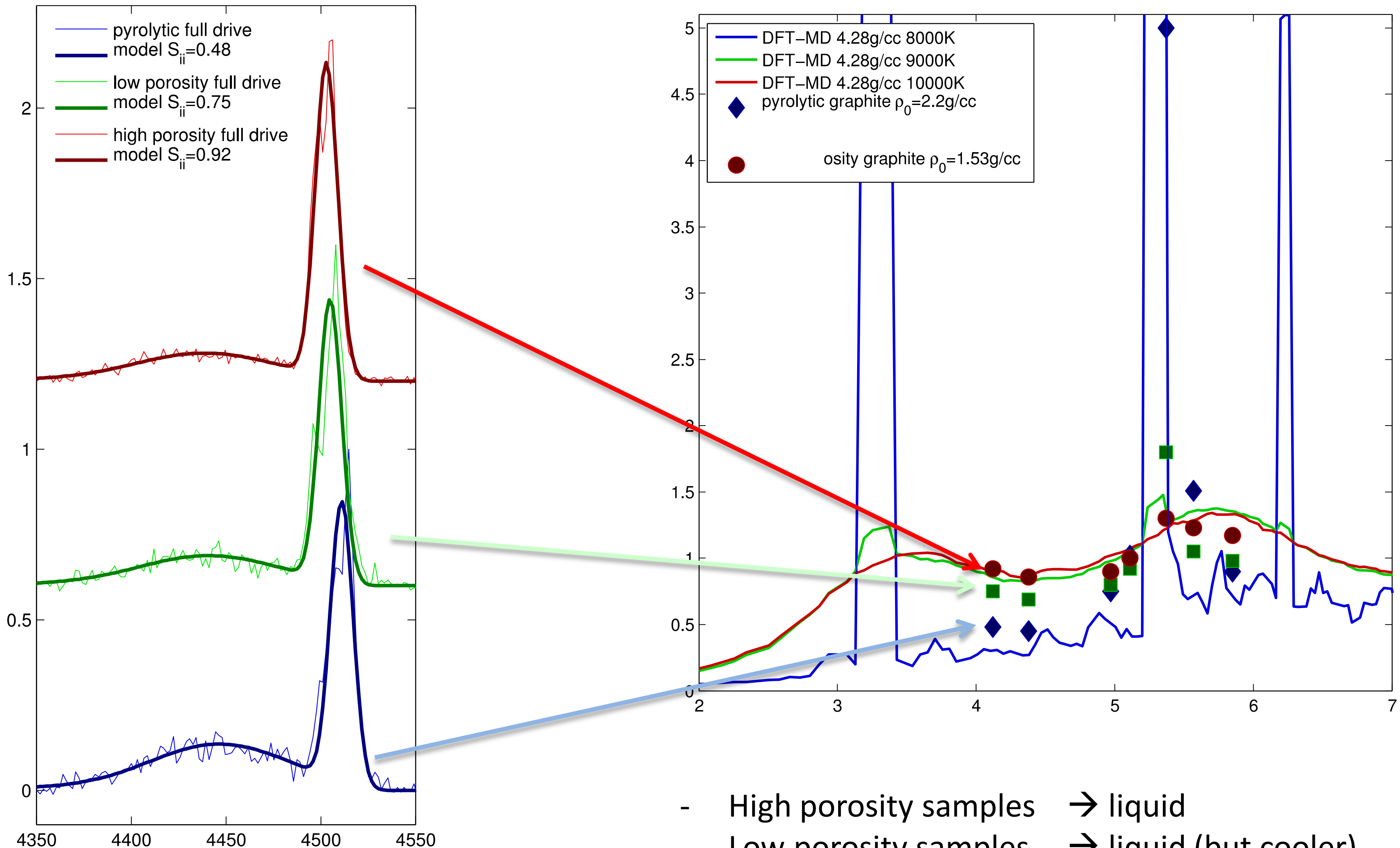
SASE Source spectrum is measured for each shot



- Possibility to check bound-free models with high accuracy
- Even K-shell bound-free feature visible
- High accuracy for S_{ii}

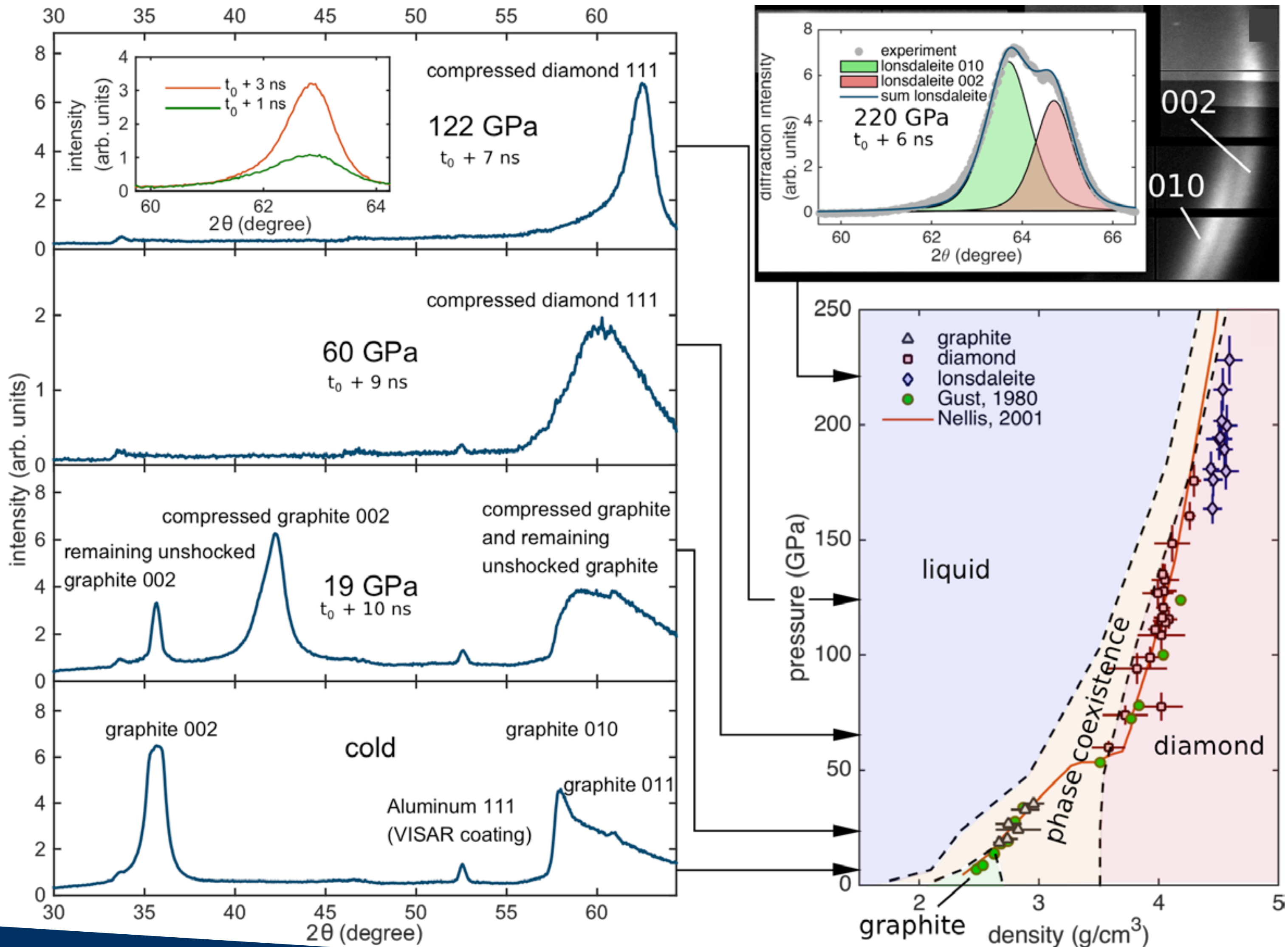


LCLS experiment | Solid and liquid structure at ~180 Gpa = 1.8 Mbar

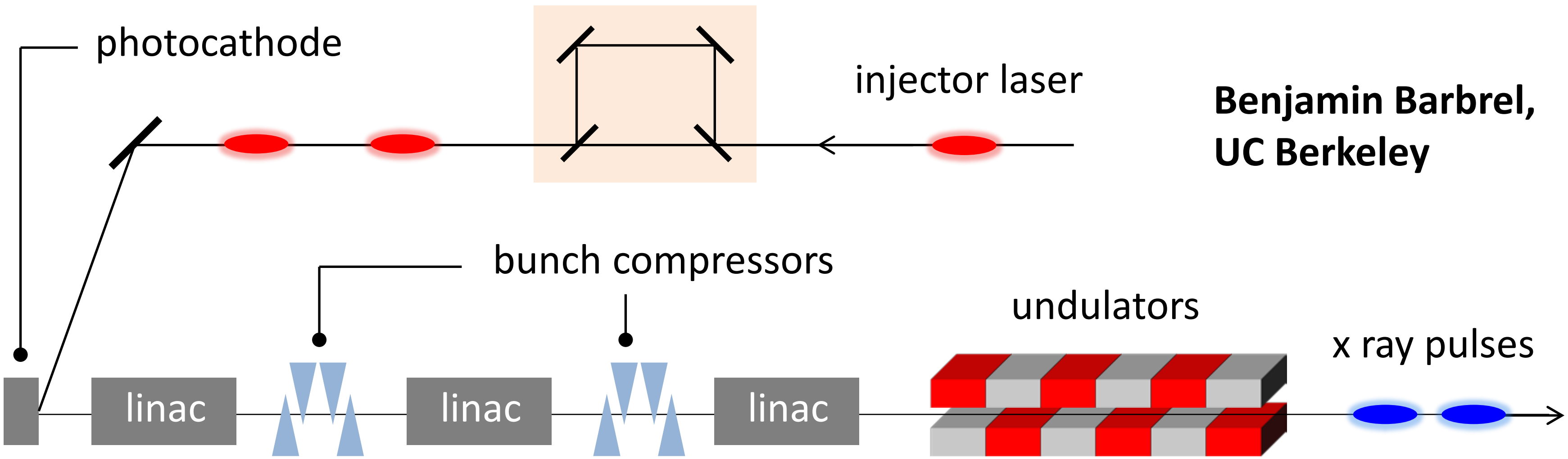


- High porosity samples → liquid
- Low porosity samples → liquid (but cooler)
- Pyrolytic graphite → solid, close to melting

LCLS experiment | X-ray Diffraction: ns-formation of diamond

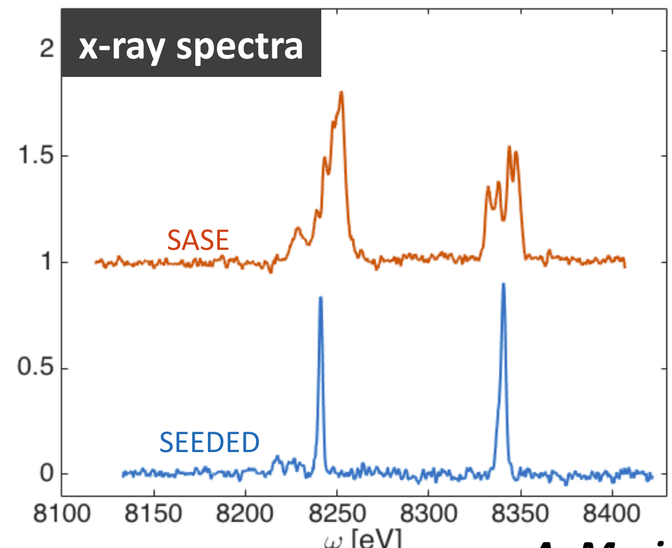


LCLS experiment | XFEL heating – two pulses – two colors



Benjamin Barbrel,
UC Berkeley

Beam Parameters (2x75pC, 8keV)	
energy separation	≤ 100eV
time separation	5 – 150fs
energy /color (SASE)	300µJ
pulse duration	12fs
self-seeding avail.	✔ ☐

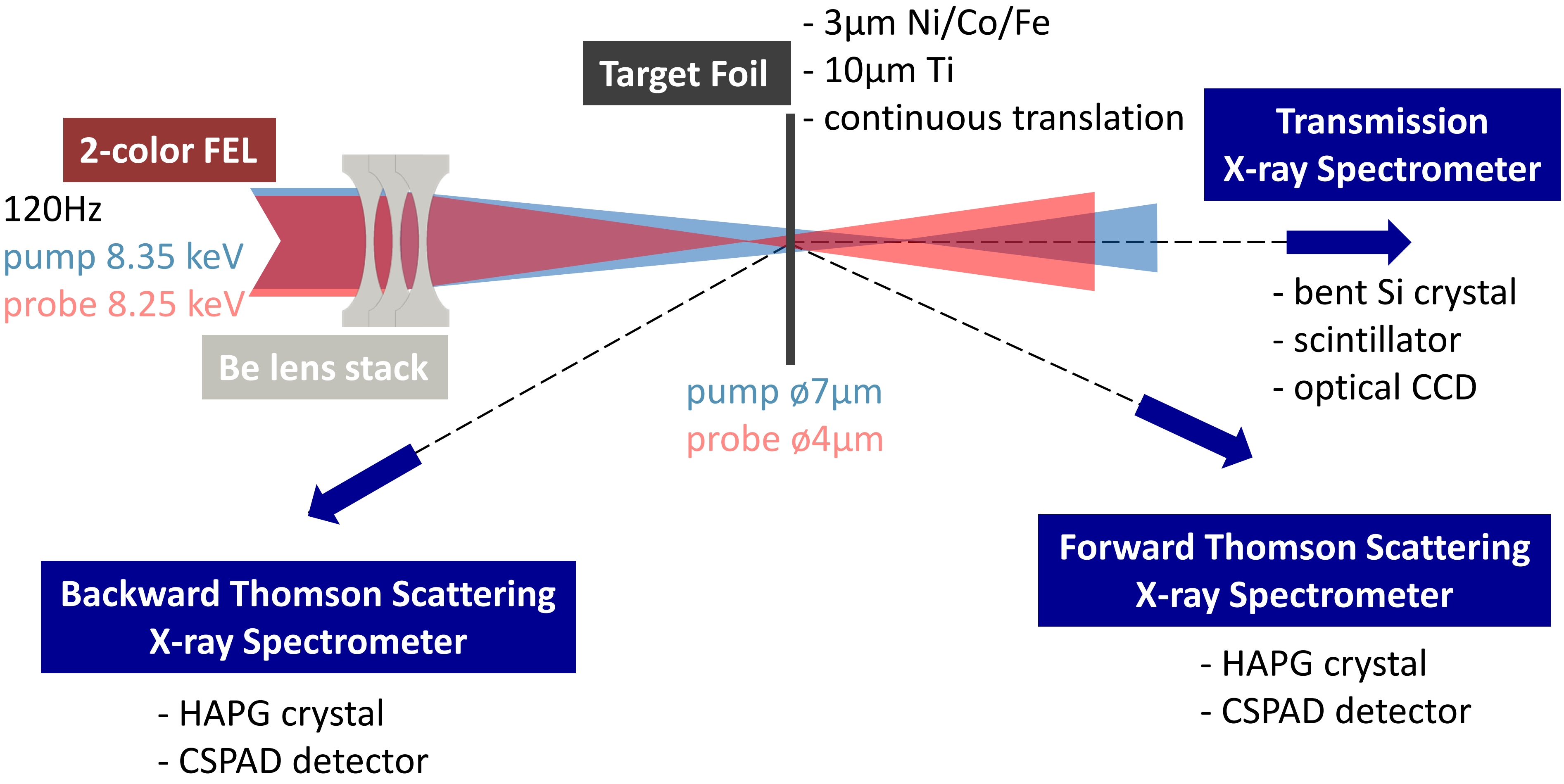


A. Marinelli

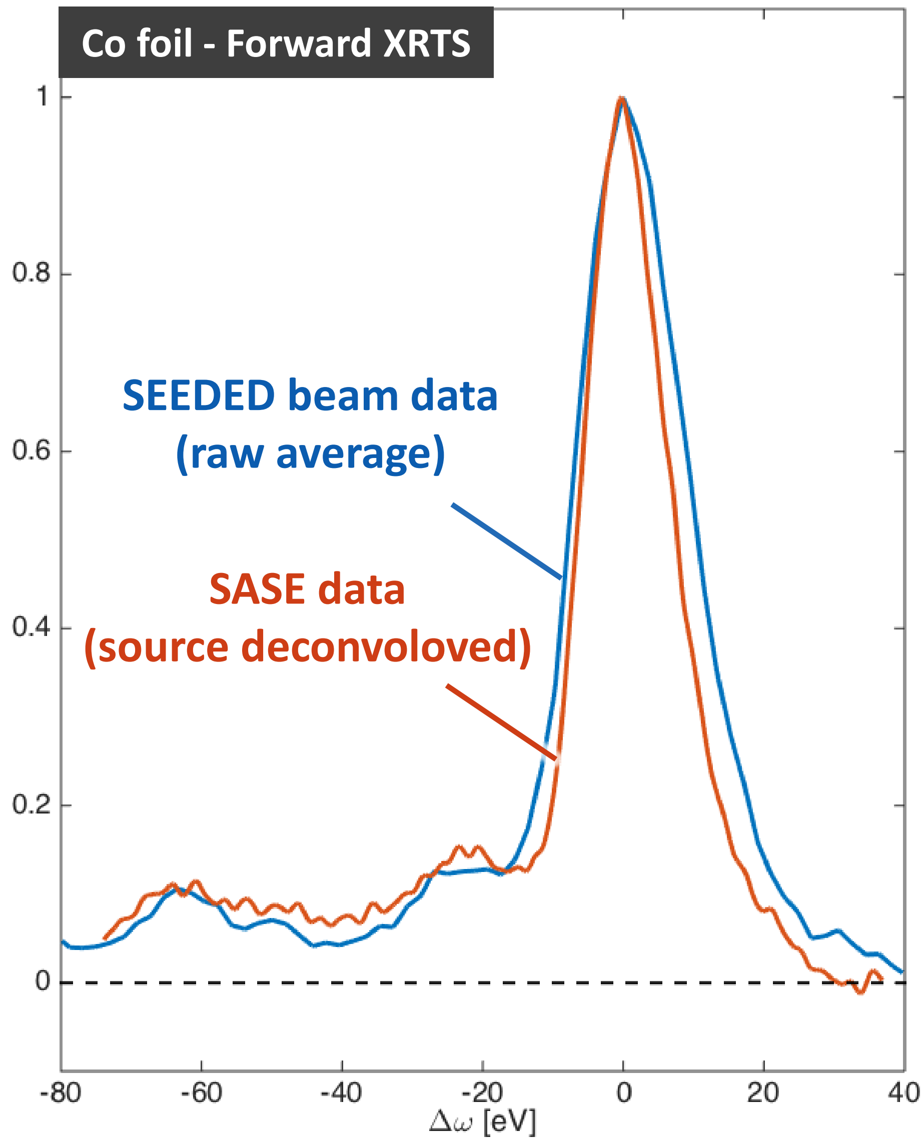
X-ray Pump / X-ray Probe

- 2-color XFEL for time-resolved XRTS in warm dense matter (Barbrel)
- isochoric heating of solid density target
- create non-equlibrium states
- femtosecond time resolution for fast processes:
  ion relaxation, electron thermalization
- various techniques
  - absorption (FEL is difficult, broadband sources = betatron?)
  - diffraction
  - inelastic x-ray scattering

Experiment Setup



Reconstruction Using Seeded Radiation



Use of 2-color seeded beam not realistic yet for an experiment

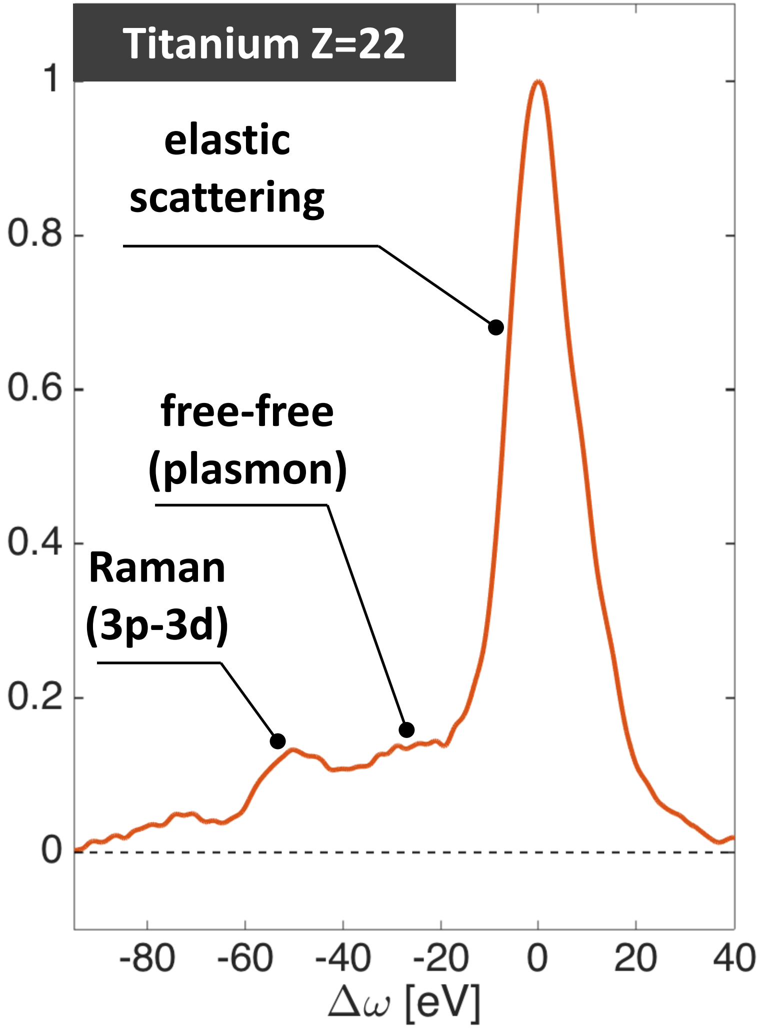
- machine tuning time very long
- lower energy (50% at best)

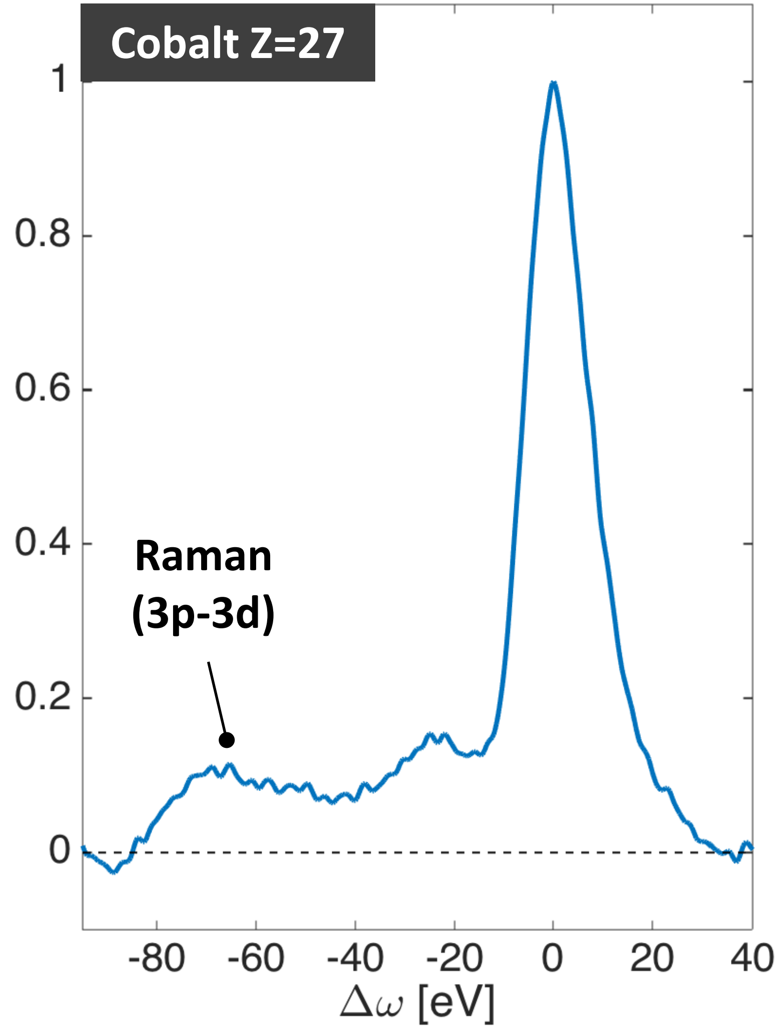
Seeded beam can be used in a limited number of cases to make sure data reconstuction is accurate:

- higer energy resolution
- no photon energy jitter

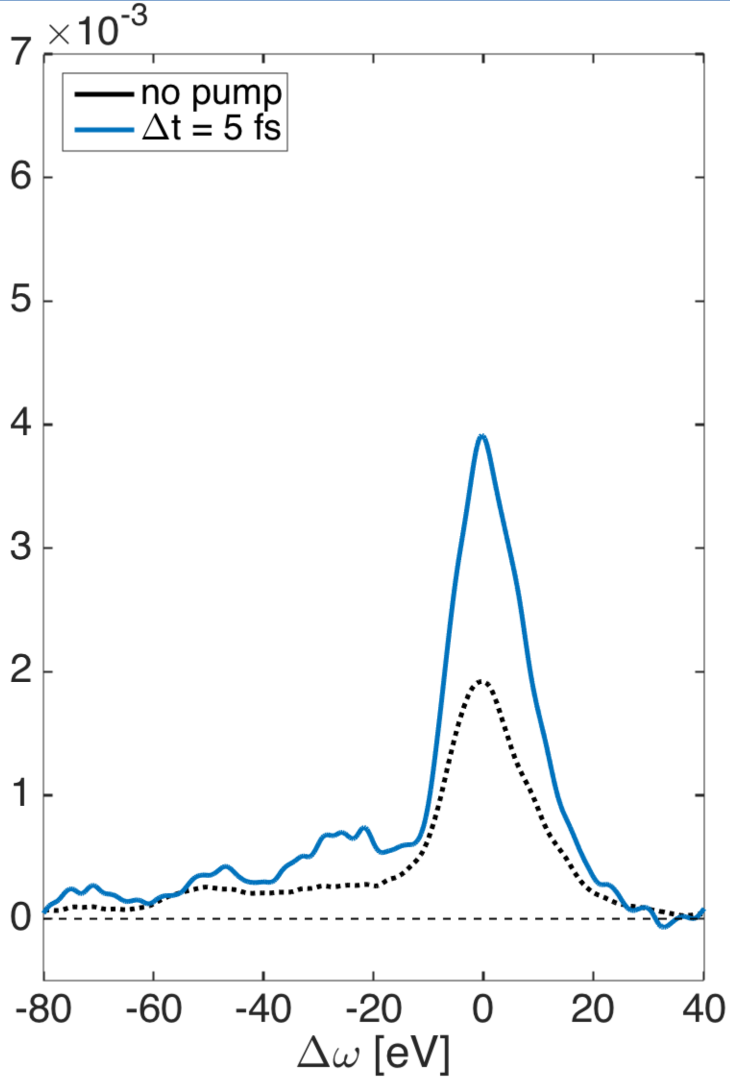
	ΔE/E	ΔE@8keV
SASE FEL	2.10^{-3}	19 eV
SEEDED FEL	10^{-4}	0.8eV

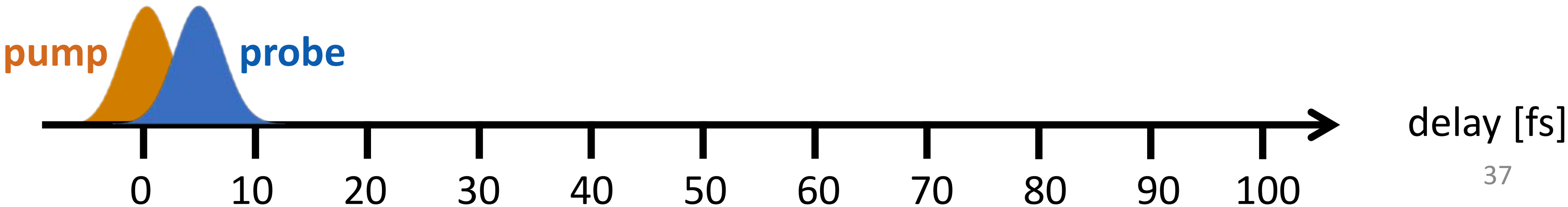
Single-Color (No Pump) Forward Scattering



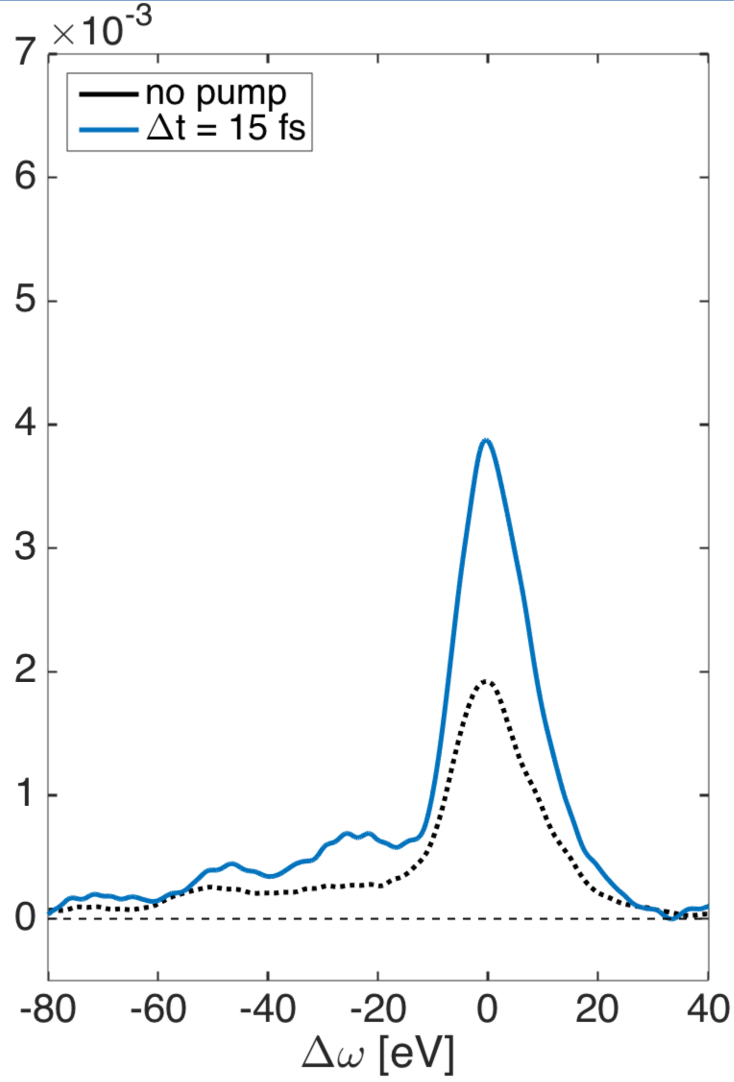


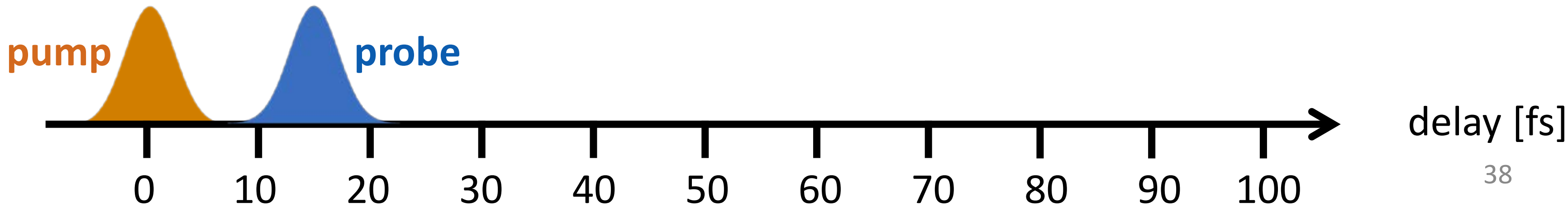
Titanium : Time-Resolved Forward Scattering





Titanium : Time-Resolved Forward Scattering





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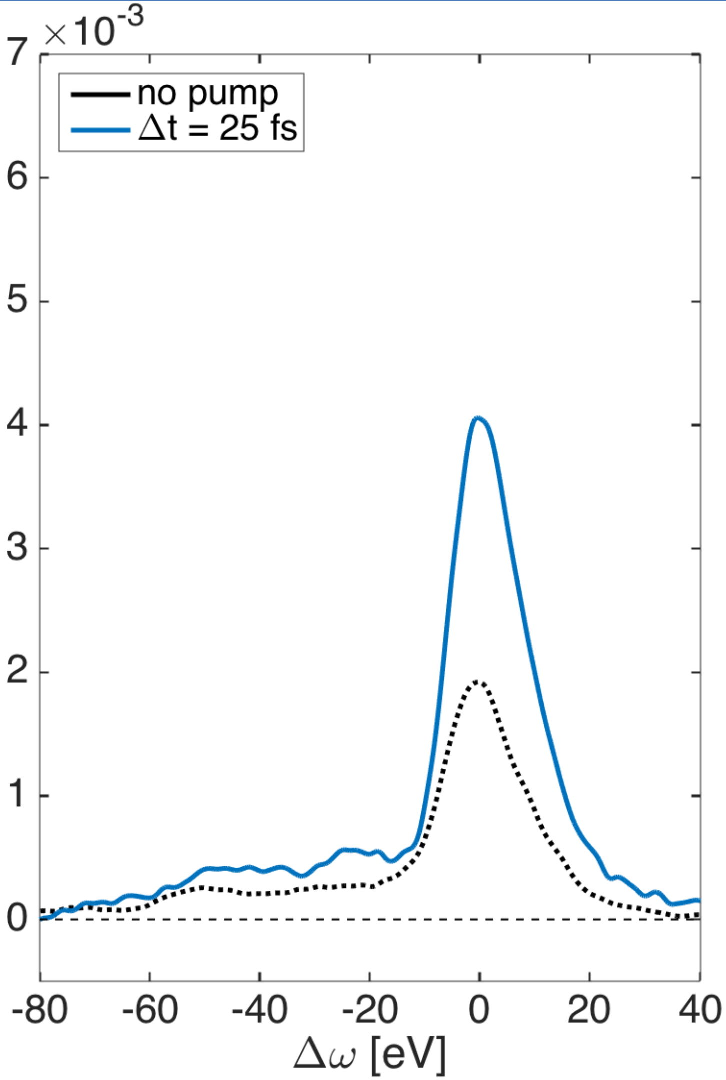

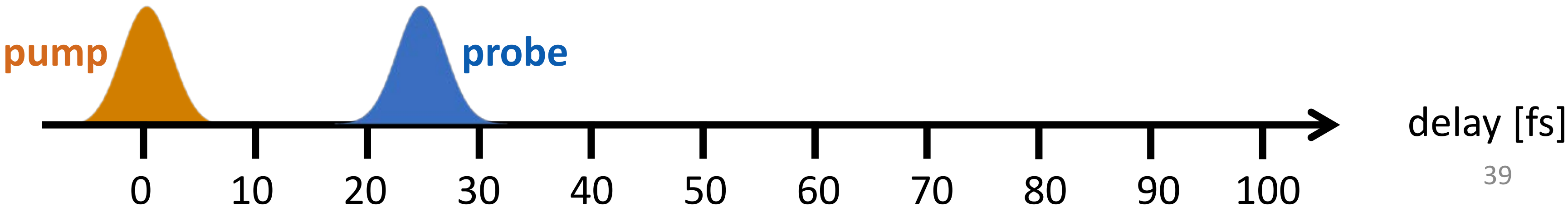

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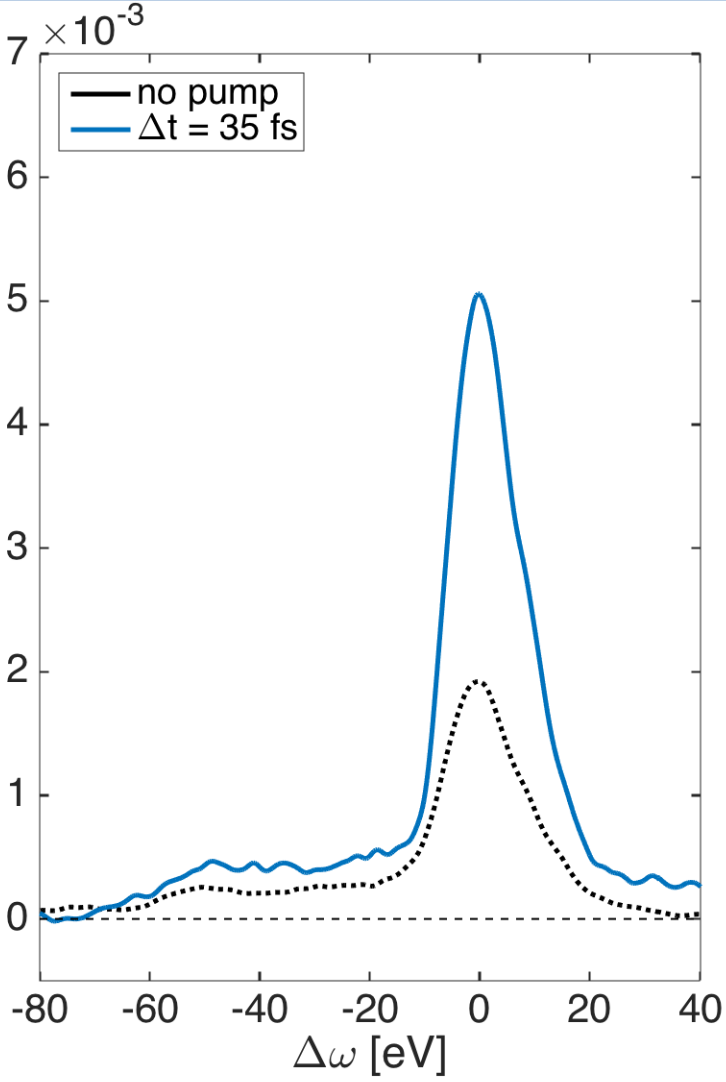

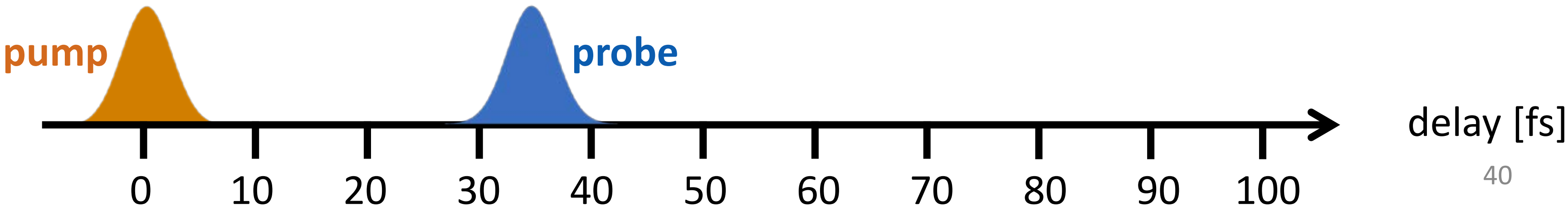

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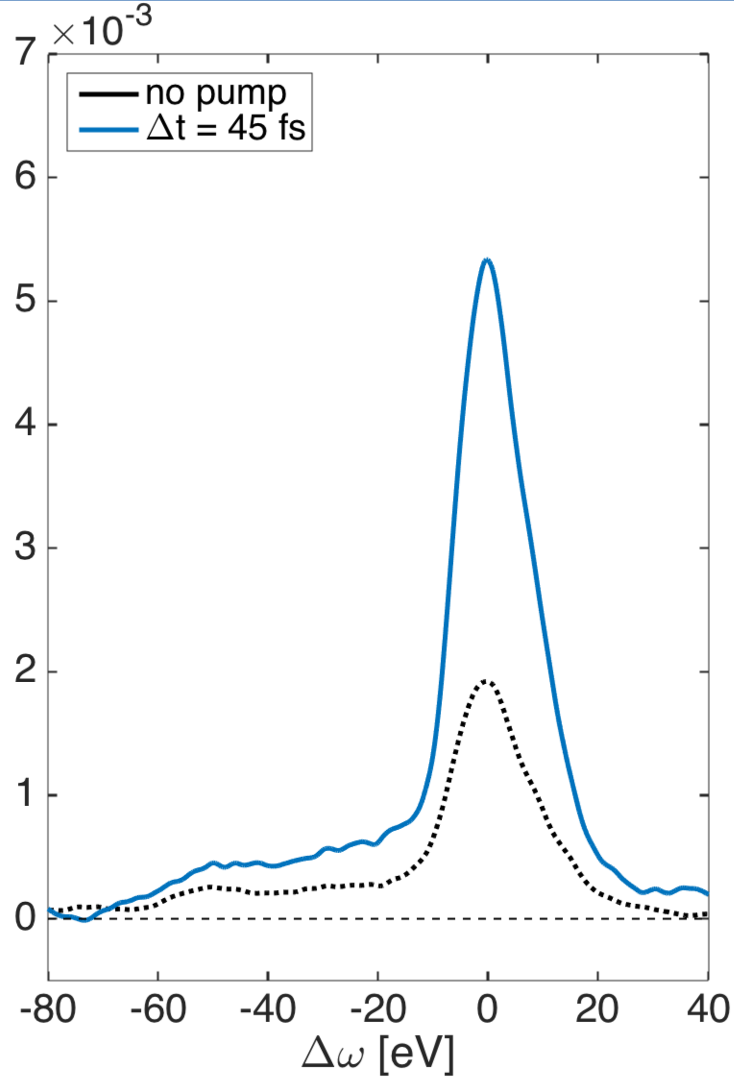

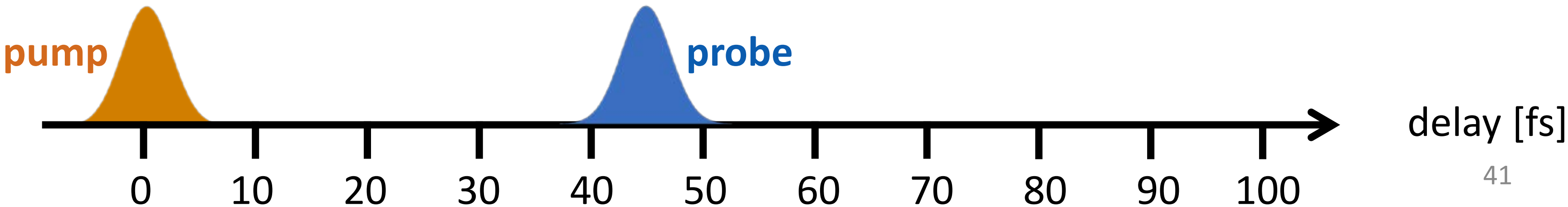

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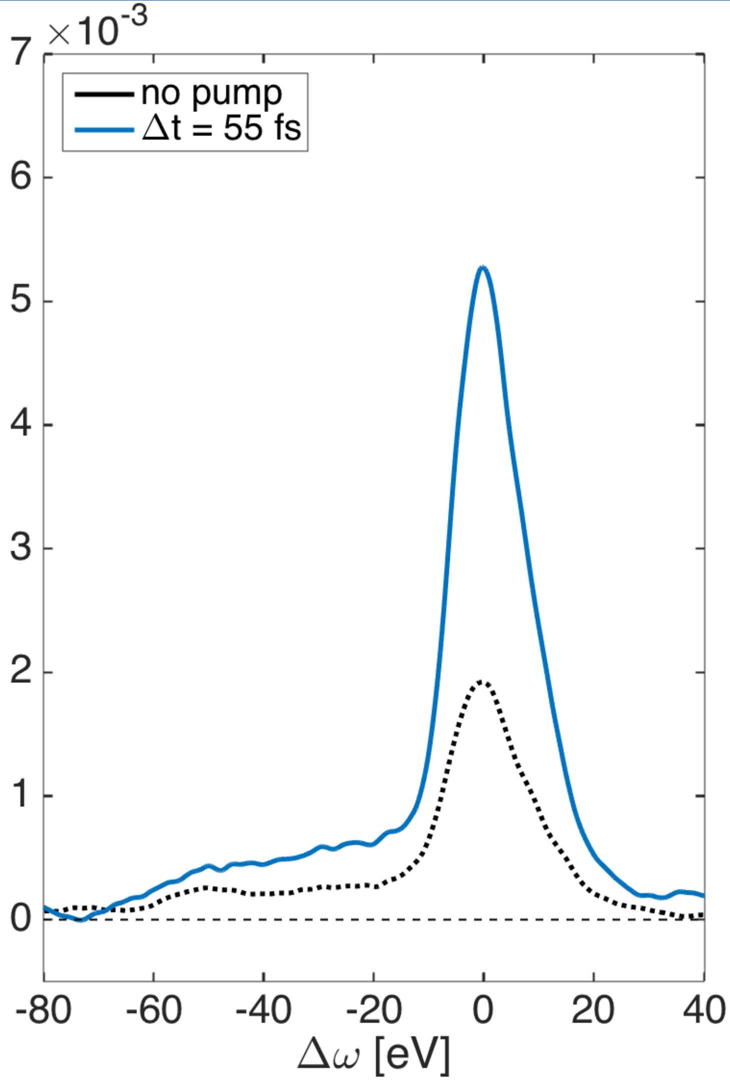

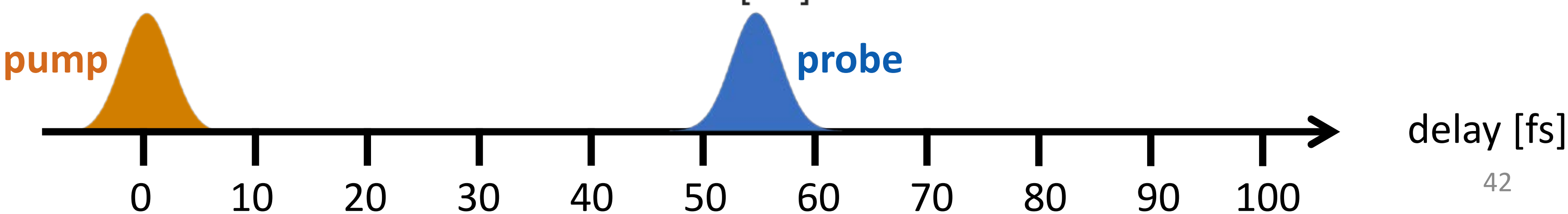

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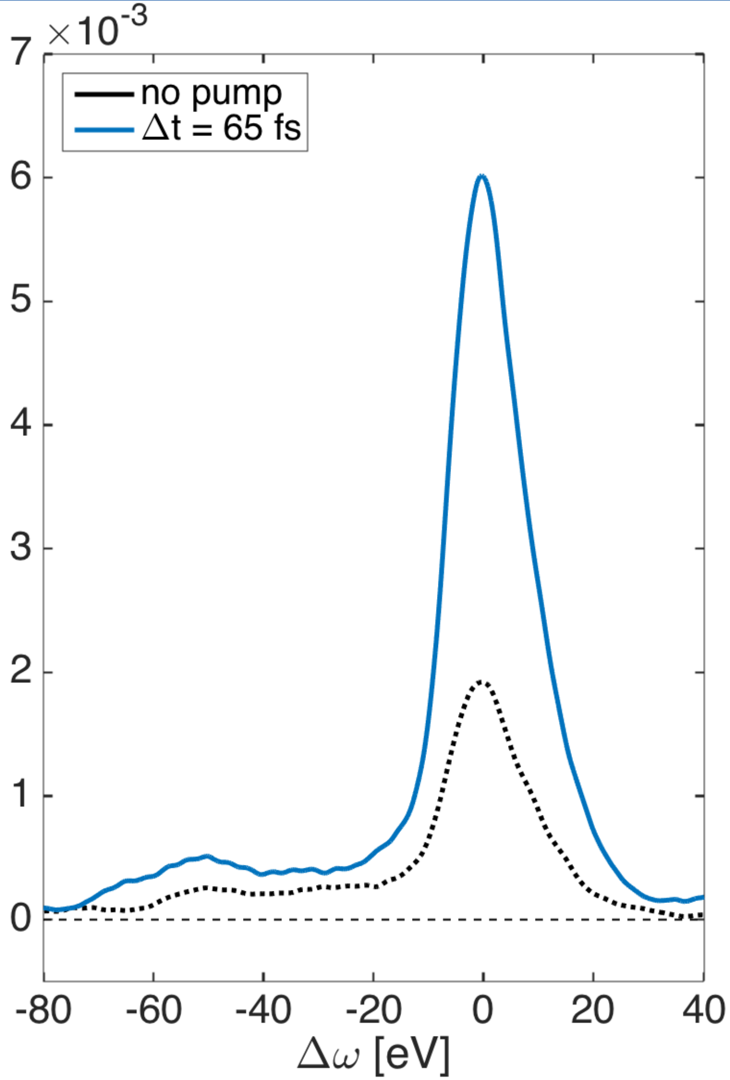

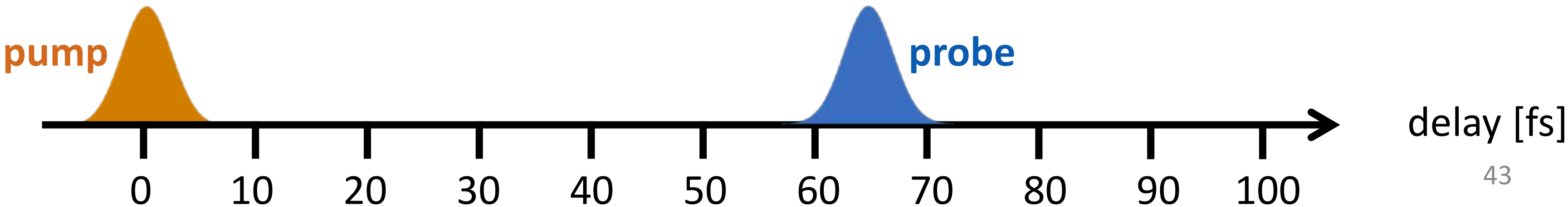

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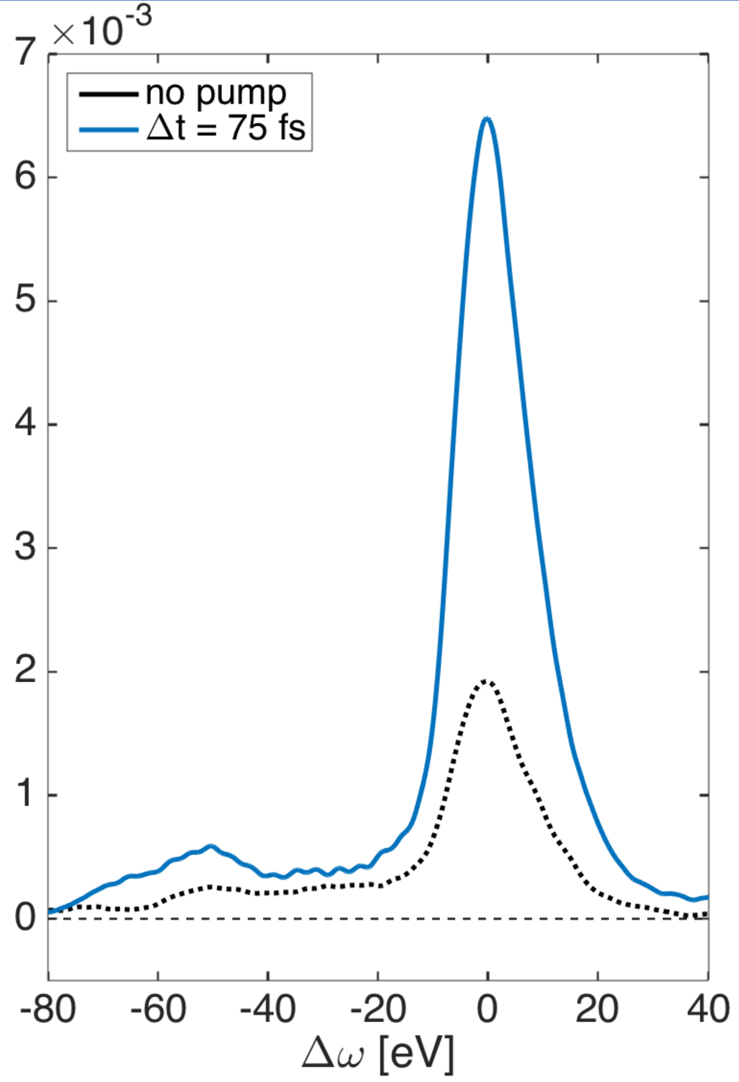

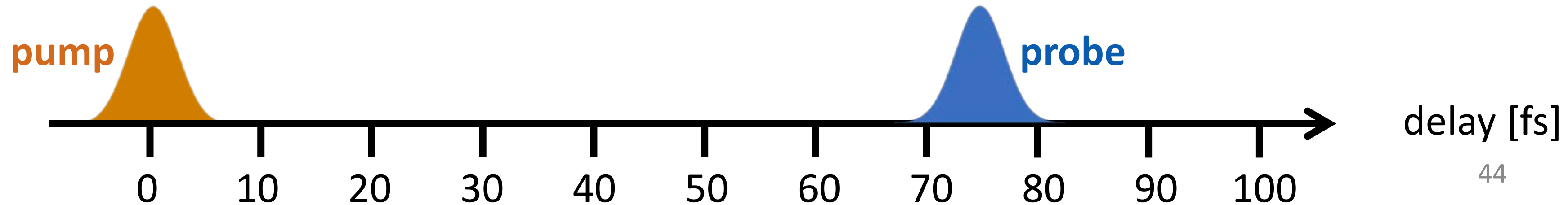

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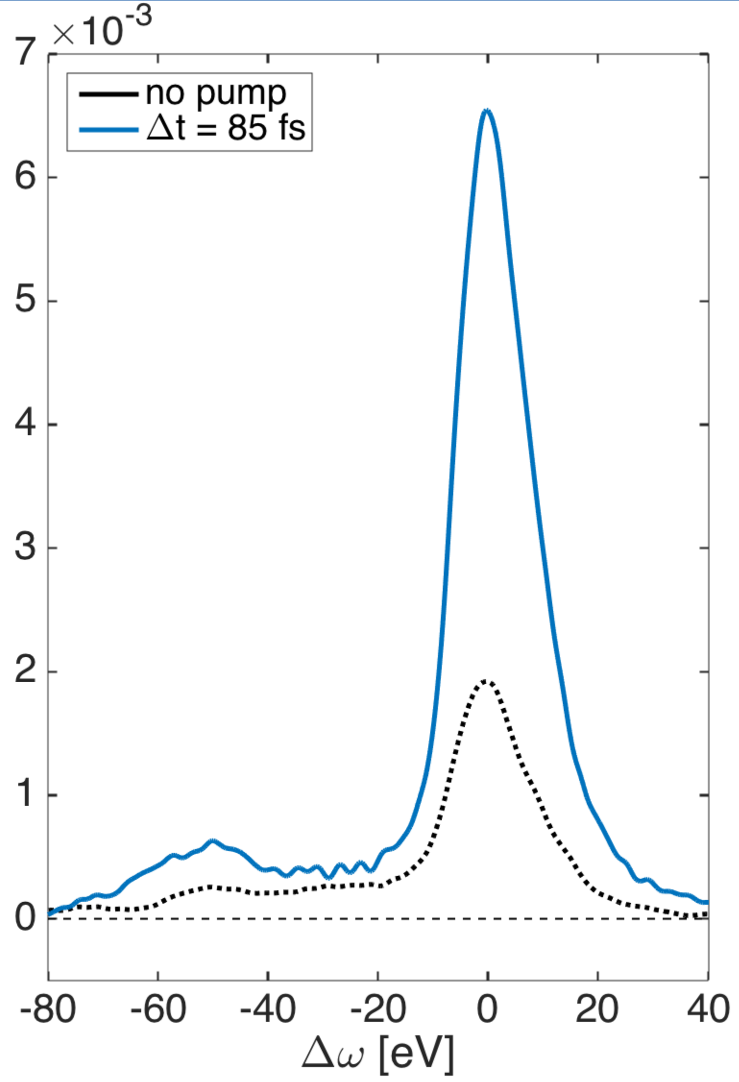

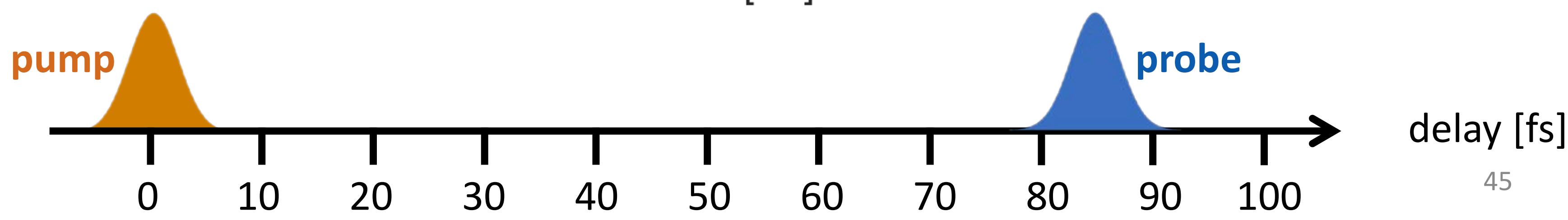

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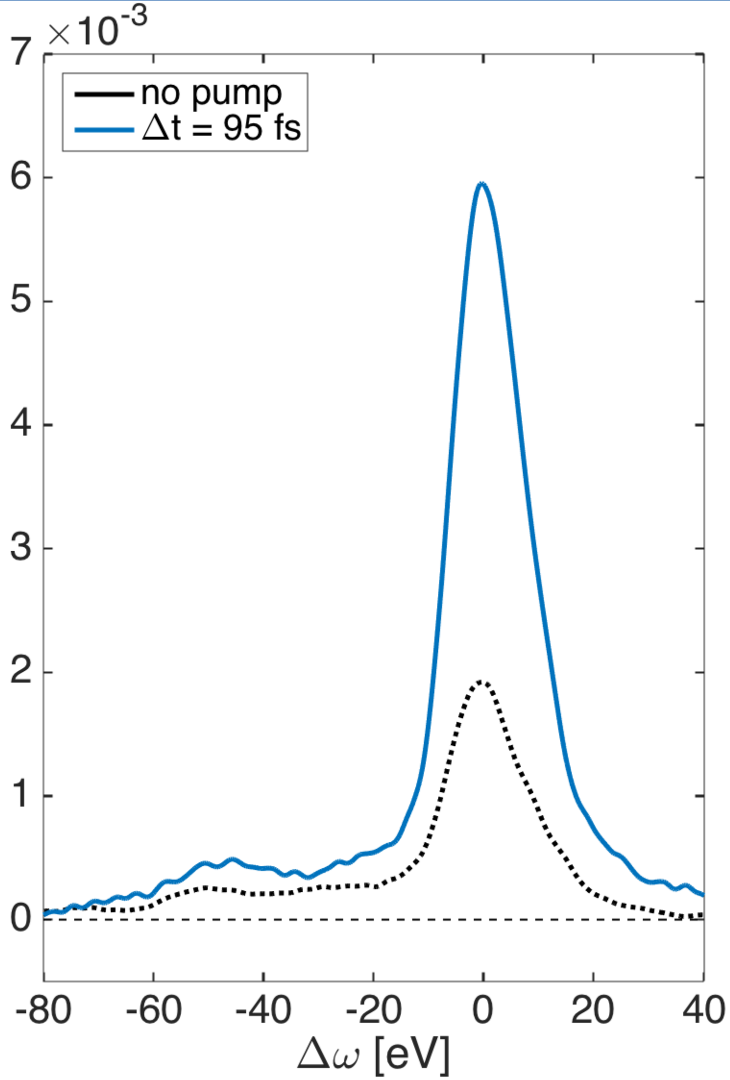
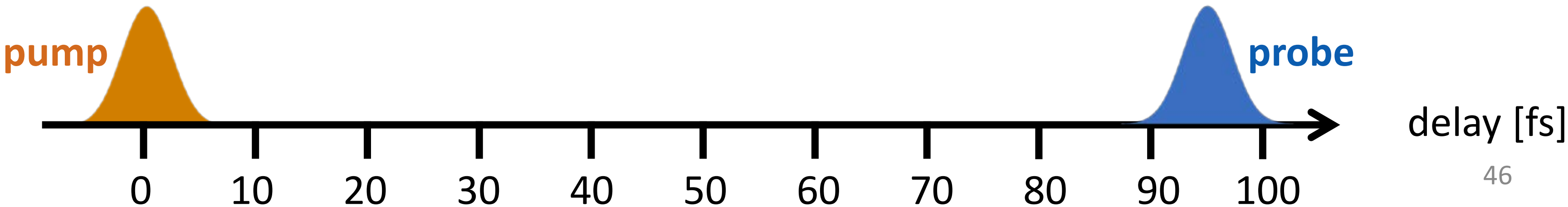
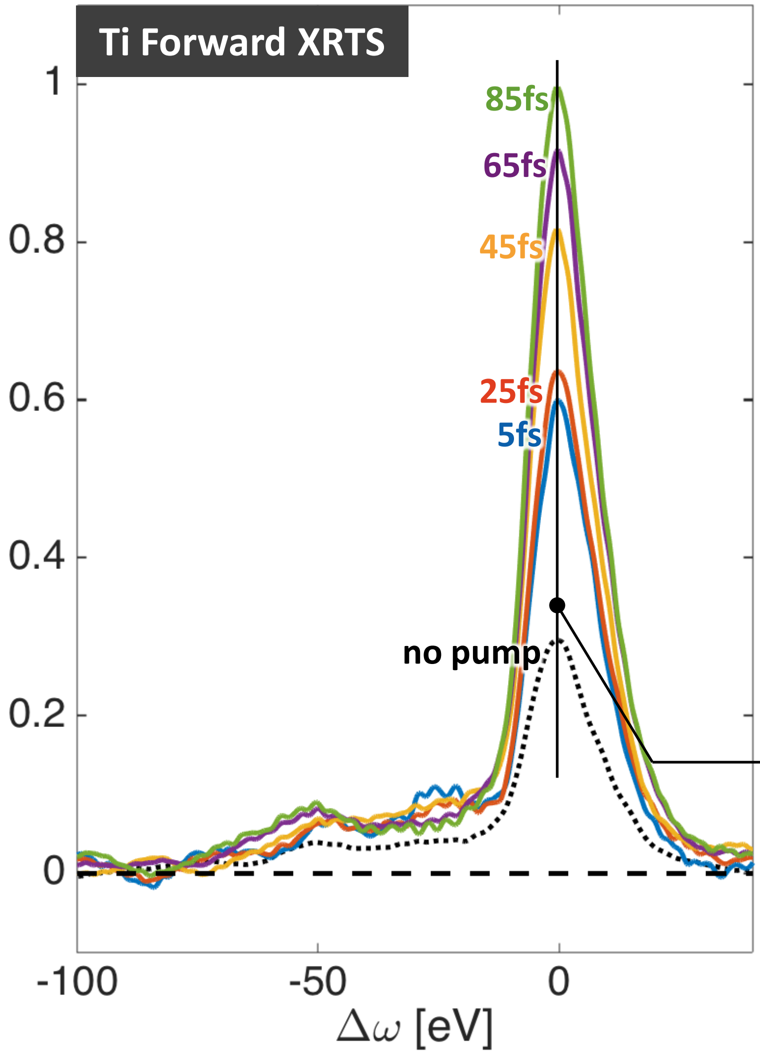
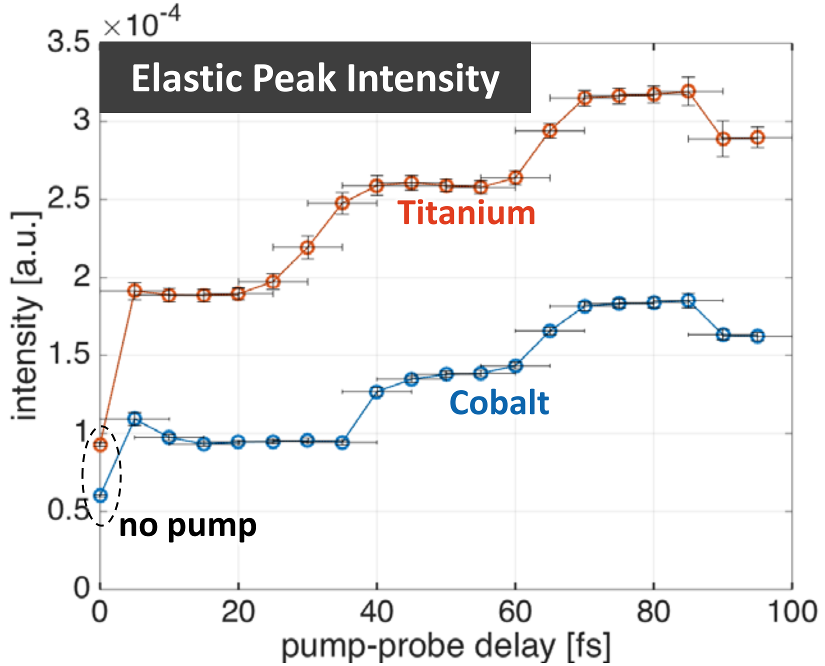




Rayleigh Scattering





strong increase of elastic scattering intensity

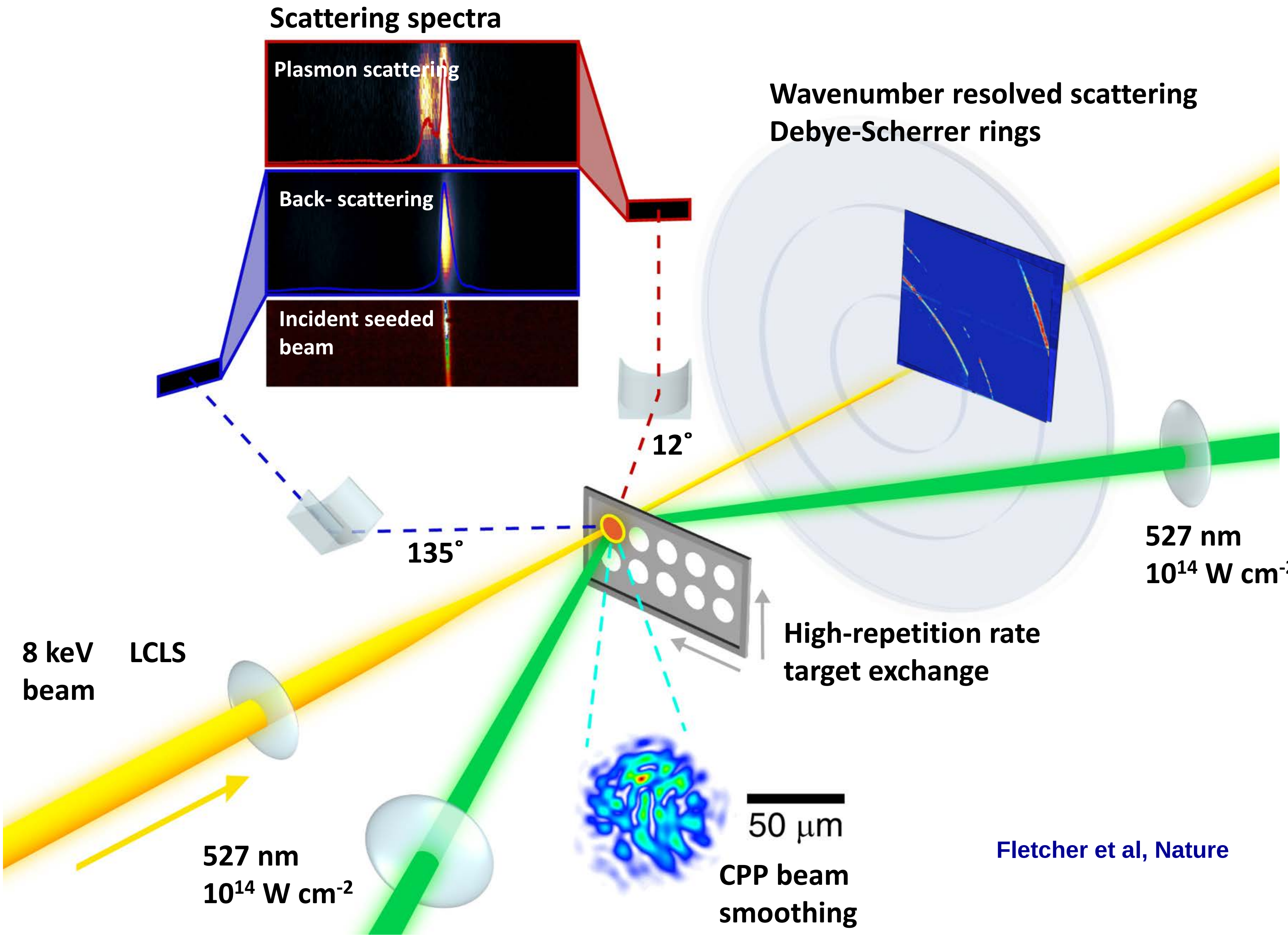
$$S(k) = [f_i(k) + q(k)]^2\, S_{ii}(k)$$

- ion structure factor
- screening free electrons
- bound electrons

current hypotheses

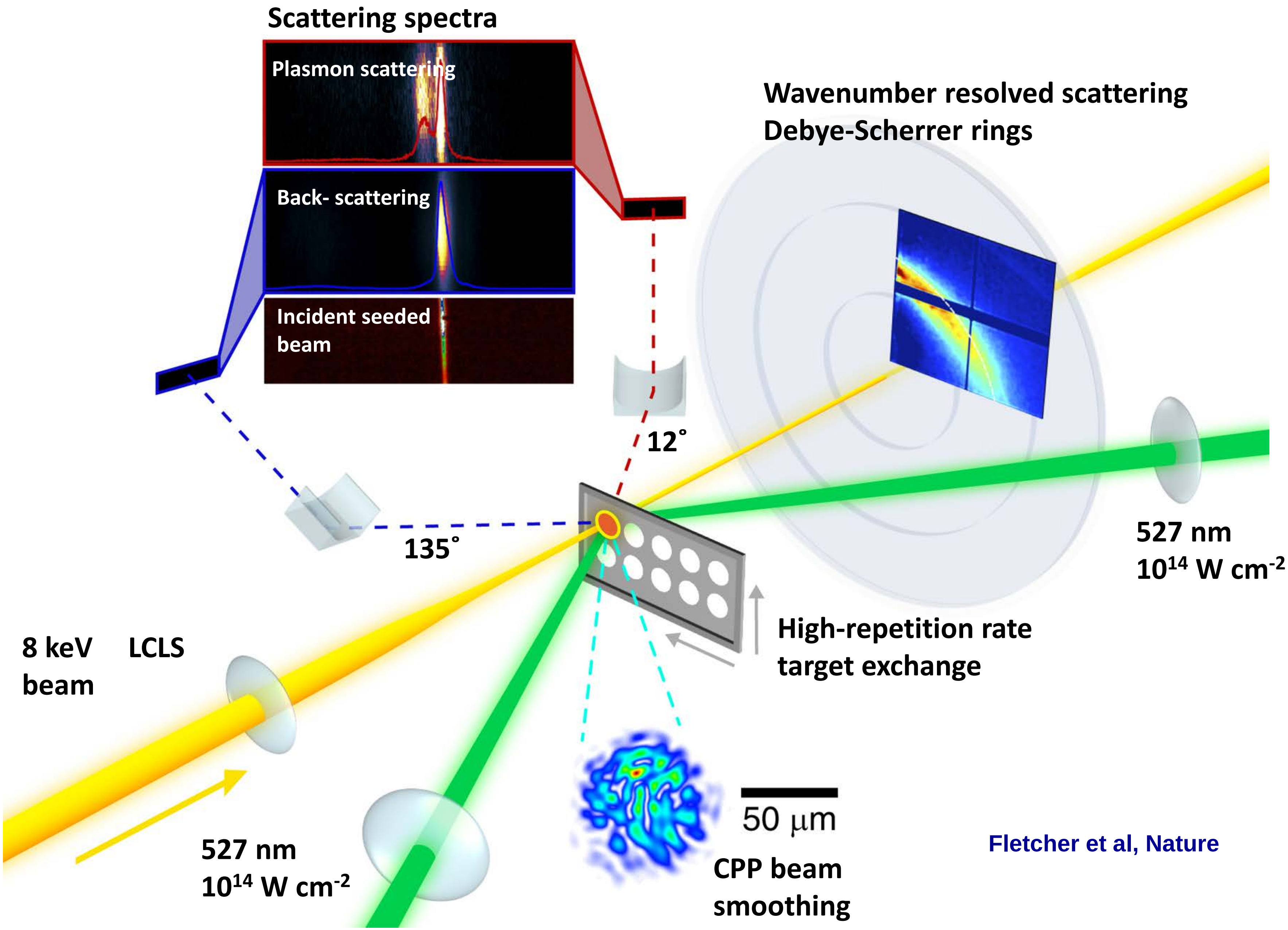
- variation of ionization state
- modification of screening length of heated e-
- ultrafast heating of ion lattice

LCLS precision characterization of HED matter



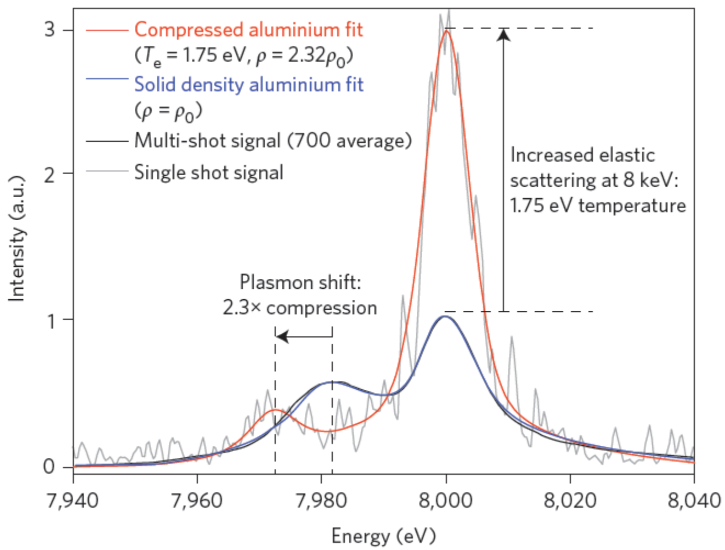
Fletcher et al, Nature

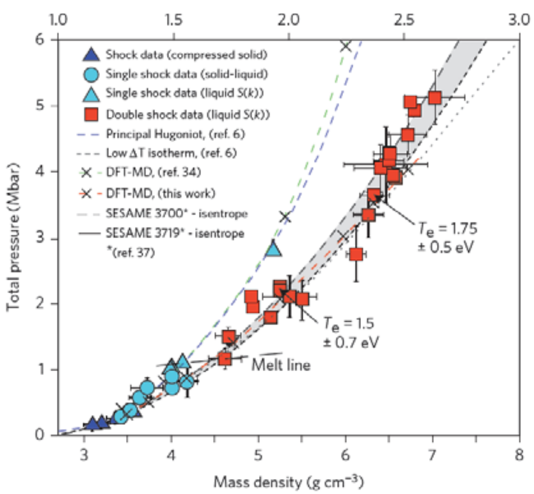
LCLS precision characterization of HED matter

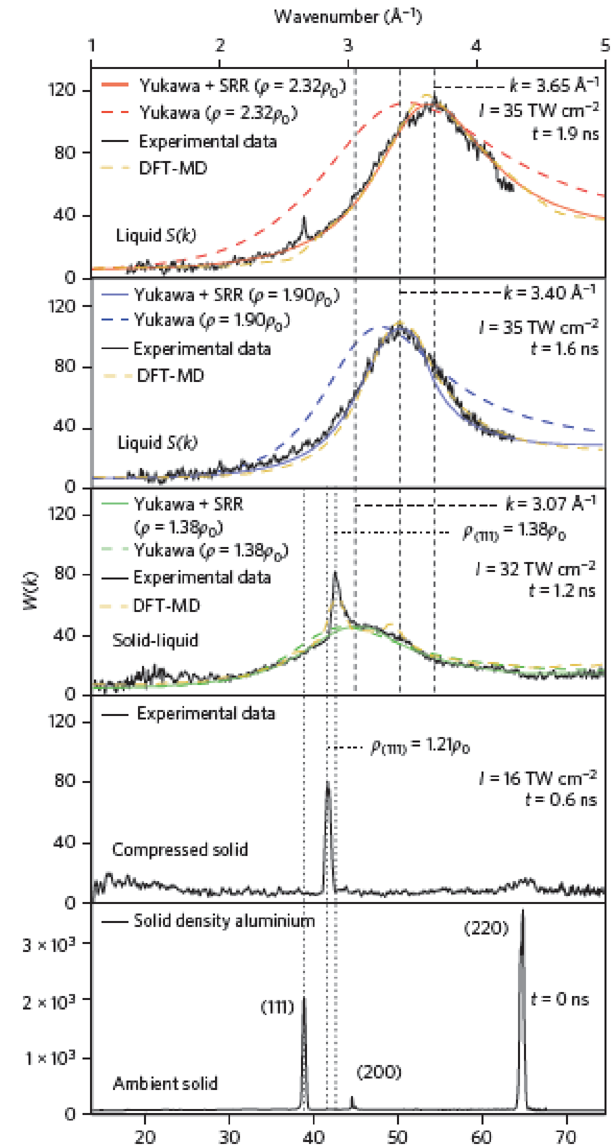


Fletcher et al, Nature

LCLS precision characterization of HED matter







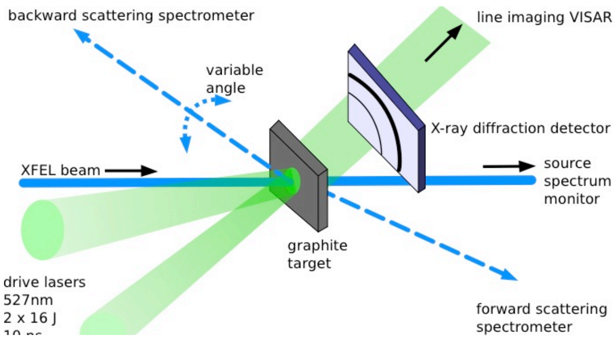
Fletcher et al, Nature

No Bragg peaks for compressed Al above P = 1.2 Mbar, (
with previous melt line measurements on the Hugoniot.

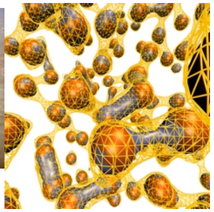
Summary

LCLS experiments:

- **Shock compressed material transformations measured by diffraction and inelastic scattering**
- **Two color x-ray pump and probe of dense plasmas**
- **Solid density plasma emission spectroscopy under intense excitation**

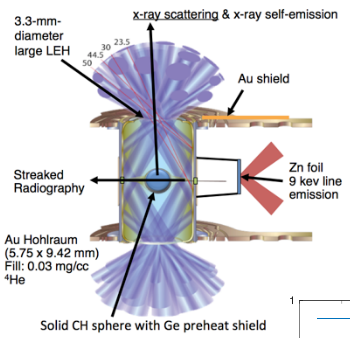


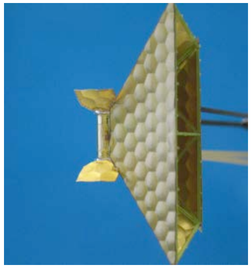


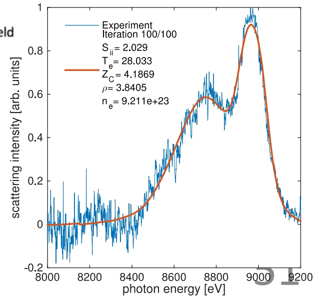


NIF experiments:

- **Measurement of temperature, density and ionization of extremely high pressure**







Challenges:

- **Bring multi-ten's of Mbar pressures to XFEL science at modest rep-rate**
- **Develop x-ray imaging, inelastic scattering, and near edge absorption structure**

Collaboration on warm and dense materials at ALS, LCLS, and NIF

- **UC, Berkeley and LBNL**
  D. Kraus B. Barbrel, K. Engelhorn,, A. Saunders, P Heimann, B. Cho

- **SLAC**
  S. Glenzer, H. J. Lee, L. Fletcher, B. Nagler, E. Galtier, M. Gauthier, E. Gamboa, M. MacDonald, S. Goede, D. Milathianaki, A. Ravasio, W. Schumaker

- **LLNL**
  T. Doeppner, A. L. Kritcher, D. Swift, B. Bachmann, G. Collins, K. Boehm, A. Pak, O. Landen, J. Nilsen, S. Le Pape, S. Hammel, T. Ma, P. Celliers, J. Eggert

- **GSI, Darmstadt**
  P. Neumayer

- **TU Darmstadt**
  M. Roth, G. Schaumann, S. Frydrych, J. Helfrich

- **MPI PKS, Dresden**
  J. Vorberger

- **AWE, UK**
  S. Rothman, D. Chapman

- **University of Warwick**
  D. O. Gericke, D. A. Chapman

- **University of Oxford, UK**
  G. Gregori, J. Wark

- **Univ. of Rostock, Germany**
  R. Redmer, et al.

- **Washington State U**
  J. A. Hawreliak

- **LANL**
  J. L. Kline, A. Yi

Collaboration on LCLS

UC Berkeley
Dominik Kraus
Kyle Engelhorn
Alison Saunders

LULI
Alessandra Ravasio

University of Trento
Giulio Monaco

University of Jena
Ulf Zastrau

GIST
Byoung-ick Cho
Minju Kim

SLAC
Siegfried Glenzer
Jerry Hastings
Eliseo Gamboa
Will Schumaker
Mac MacDonald
Maxence Gauthier
Luke Fletcher

MEC
Phil Heimann
Eric Galtier
Hae Ja Lee
Bob Nagler

LCLS
Agostino Marinelli
Timothy Maxwell

IAEA
Hyun-Kyung Chung