Atomic and Electronic Structure of Warm Dense Silicon

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Motivation

We are developing a new platform to directly measure and understand the structure and transport of warm dense matter.

Challenges

Warm Dense Matter
High temperature (eV), high pressure (Mbar)

- Challenging to model
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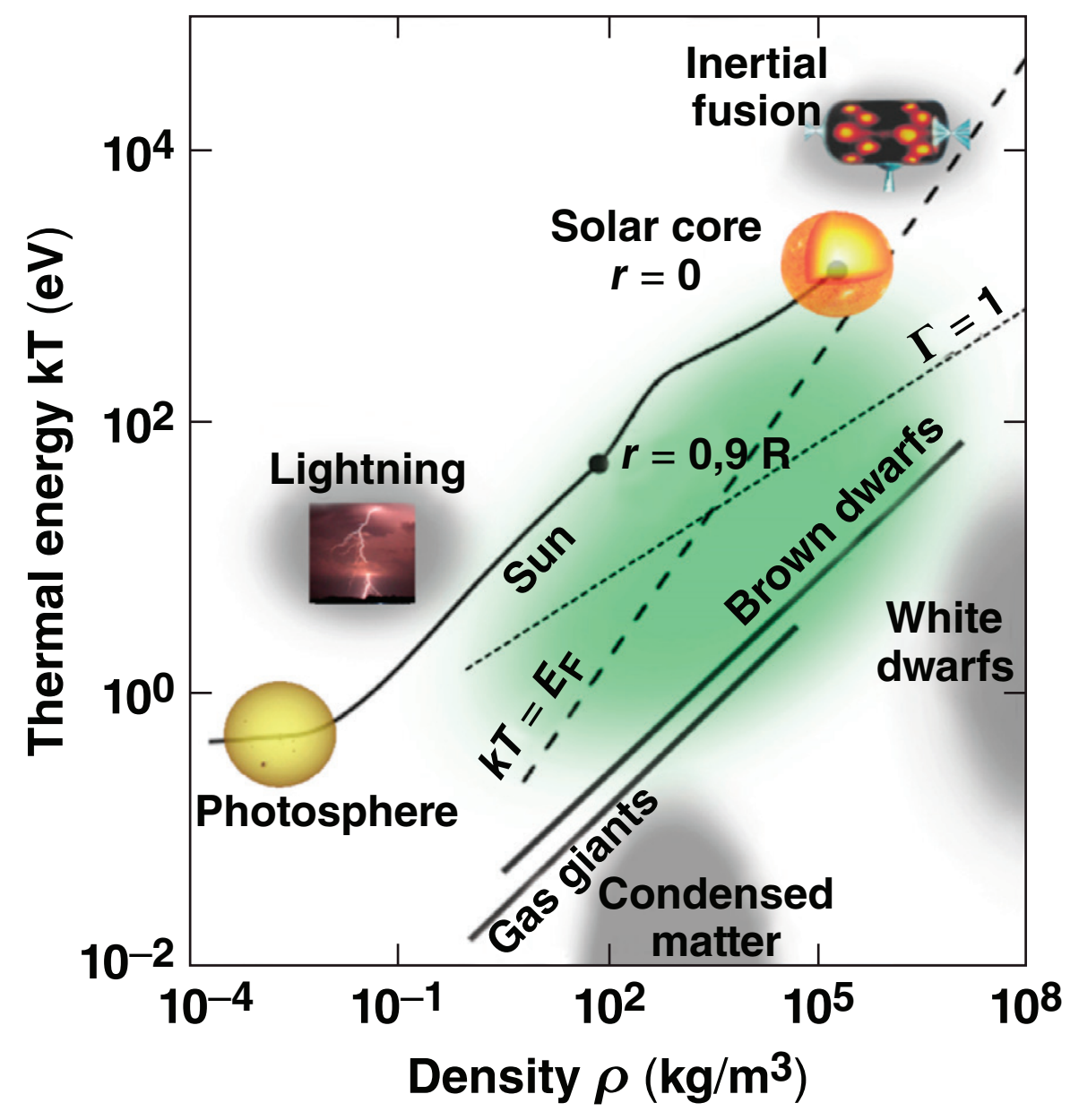

Phase diagram including the approximate location of warm dense matter (green area)

https://en.wikipedia.org/wiki/Warm_dense_matter#/media/File:Temperature-density-phasediagram.png

Goals

1. Measure density and temperature states by constraining the assumptions with multiple simultaneous diagnostics.
2. Calculate structure factors without model assumptions.
3. Estimate ionization from Thomson-scattering data and constraint those with VISAR optical reflectivity measurements.

VISAR: velocity interferometry for any reflector

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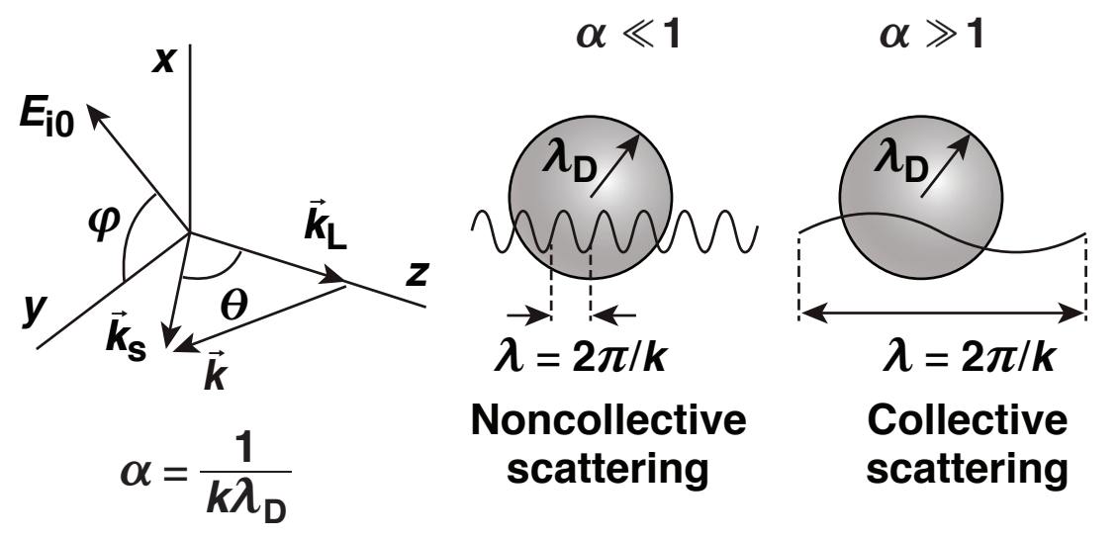
Introduction

Thomson scattering
- Elastic scattering of electromagnetic radiation from free charged particles
- Electrons accelerate in the radiation's electric field, causing the electrons to reradiate
- Scattered intensity is proportional to the electron density and temperature

Rayleigh scattering
- Elastic scattering of electromagnetic radiation from ions

Compton scattering
- Energy downshift of the scattered spectrum as a result of electron recoil

Scattering geometry

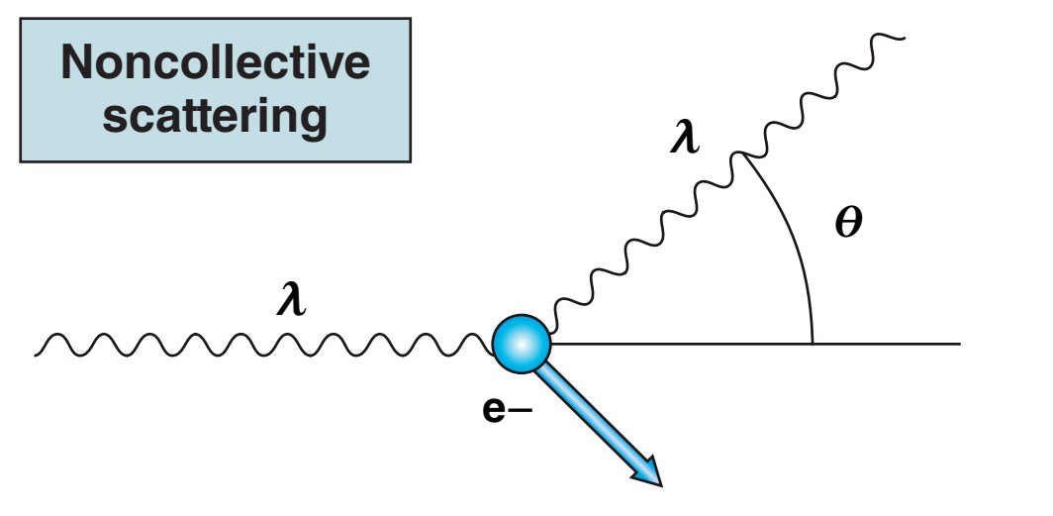


$\alpha = \frac{1}{k\lambda_D}$

Noncollective scattering

Collective scattering

Noncollective scattering



Incoherent scattering process: incoming radiation interacts with each electron

The total dynamic structure factor [1]

$$S(k,\omega) = \underbrace{\left| f_1(k) + q(k) \right|^2 S_{ii}(k,\omega)}_{\text{Elastic scattering component}} + \underbrace{Z_f\, S^0_{ee}(k,\omega)}_{\text{Inelastic scattering contribution from free electrons}} + \underbrace{Z_b \int \tilde{S}_{ce}(k,\omega-\omega')\, S_s(k,\omega')\, d\omega'}_{\text{Inelastic scattering contribution from strongly bound core electrons}}$$

$S_{ii}(k,\omega)$ = ion–ion dynamic structure factor
$q(k)$ = Fourier transform of free-electron cloud
Z_f = number of free (or valence) electrons

The terms are dependent on model assumptions. We will make direct measurements.

$S^0_{ee}(k,\omega)$ = high-frequency part of the electron–electron correlation function

Random phase approximation [2]

$f_1(k)$ = ionic form factor for bound electrons
Z_b = bound electrons

Hartree–Fock single active approximation [3]

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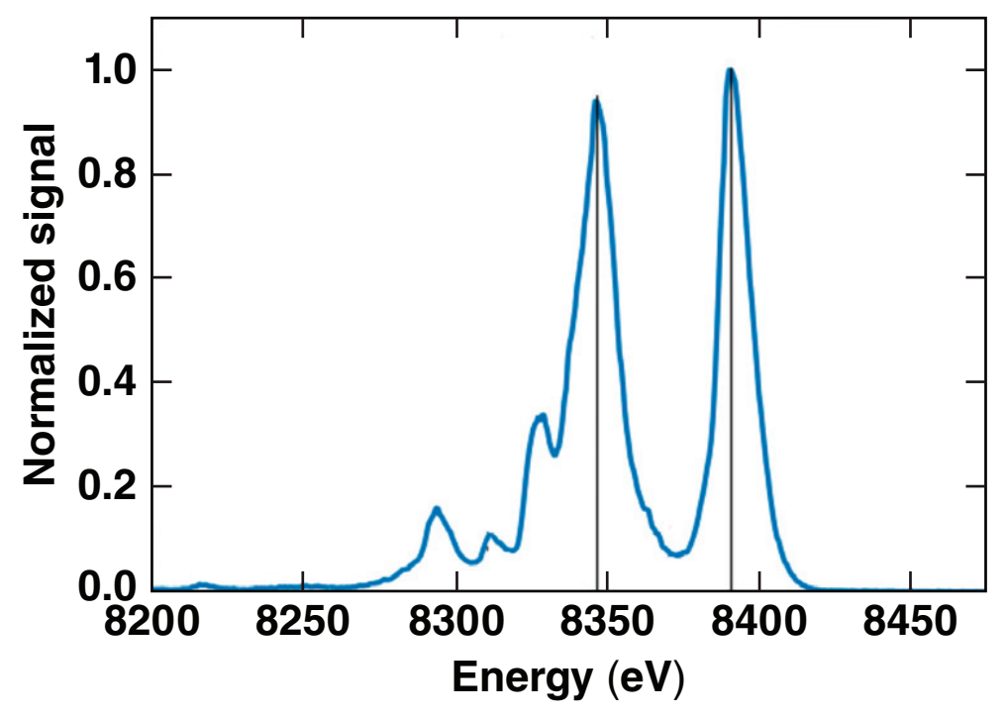
Method

Experimental Setup

X-ray source



The He-like doublet (8.35 and 8.39 keV) of a Cu backlighter

X-ray diffraction (PXRDIP)

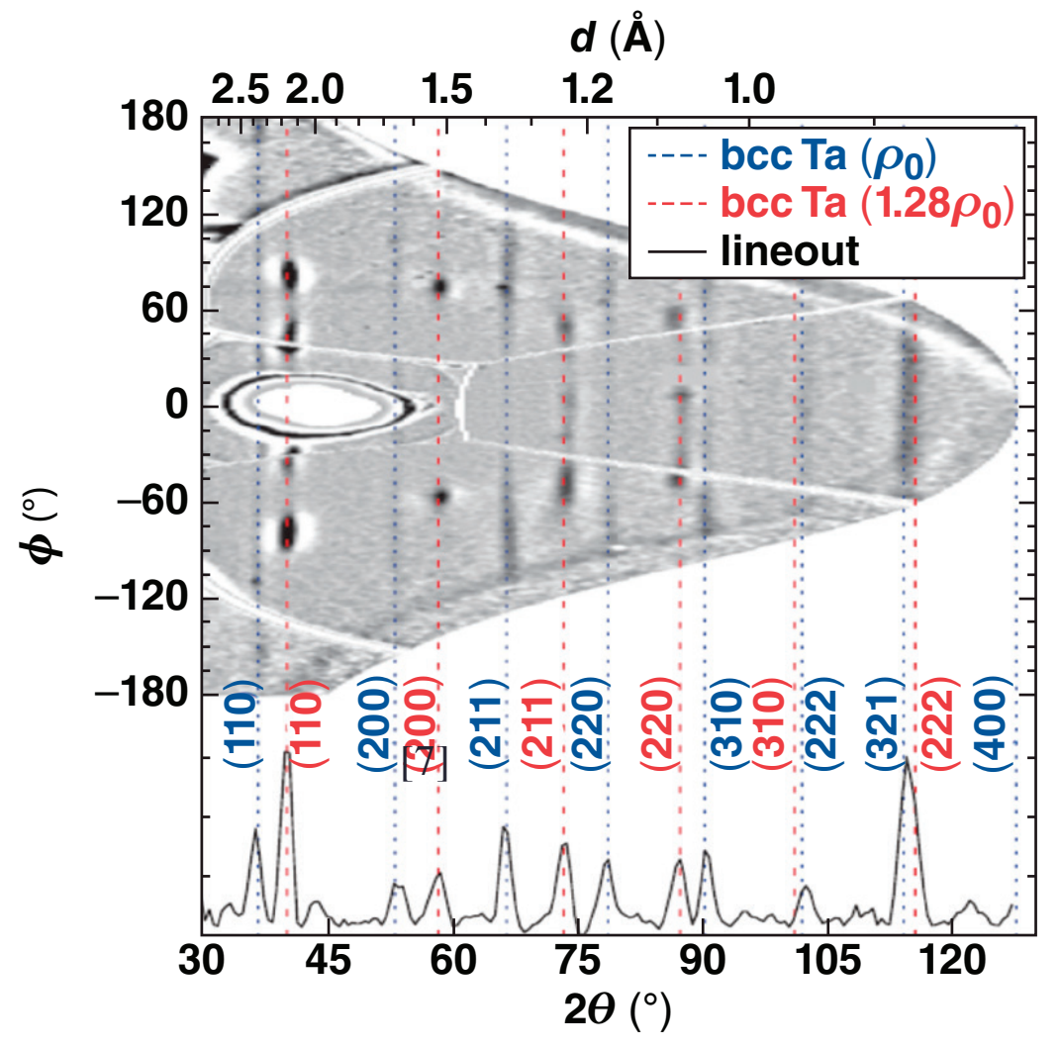
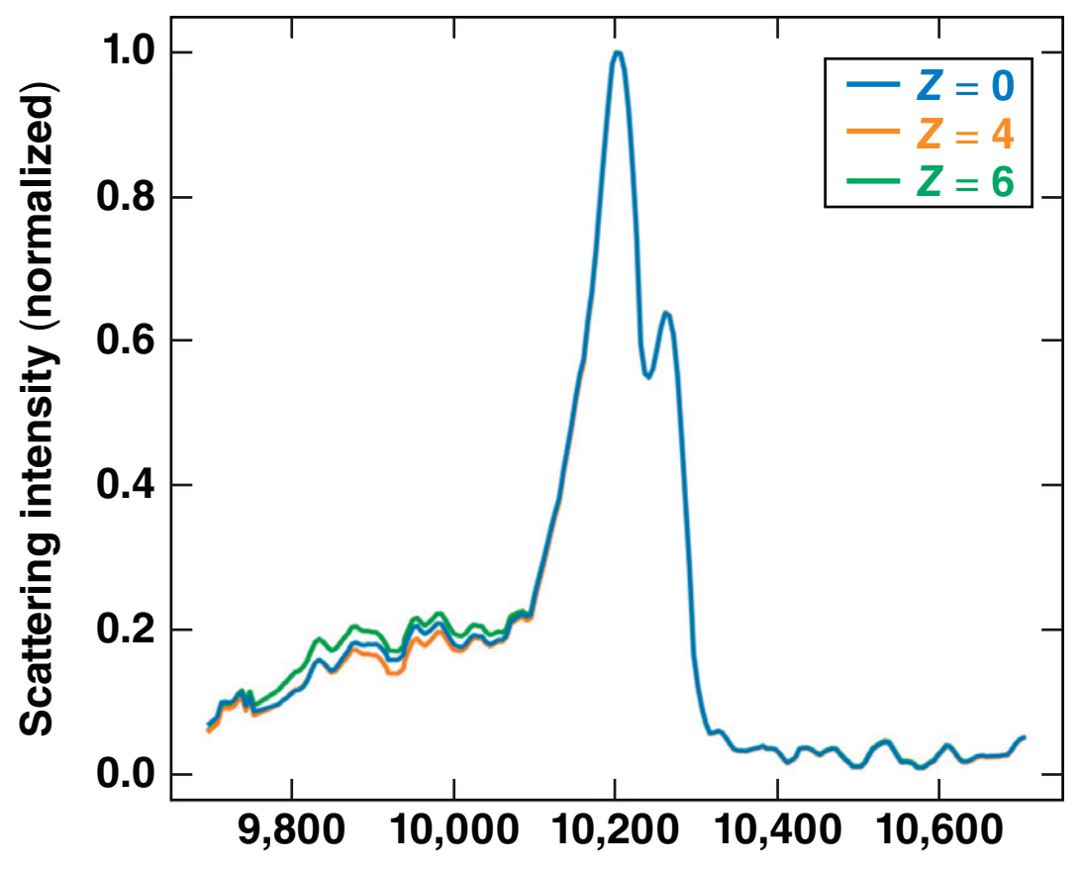


Image plates on five sides of the PXRDIP box record the angular distribution of x-ray scattering [4]

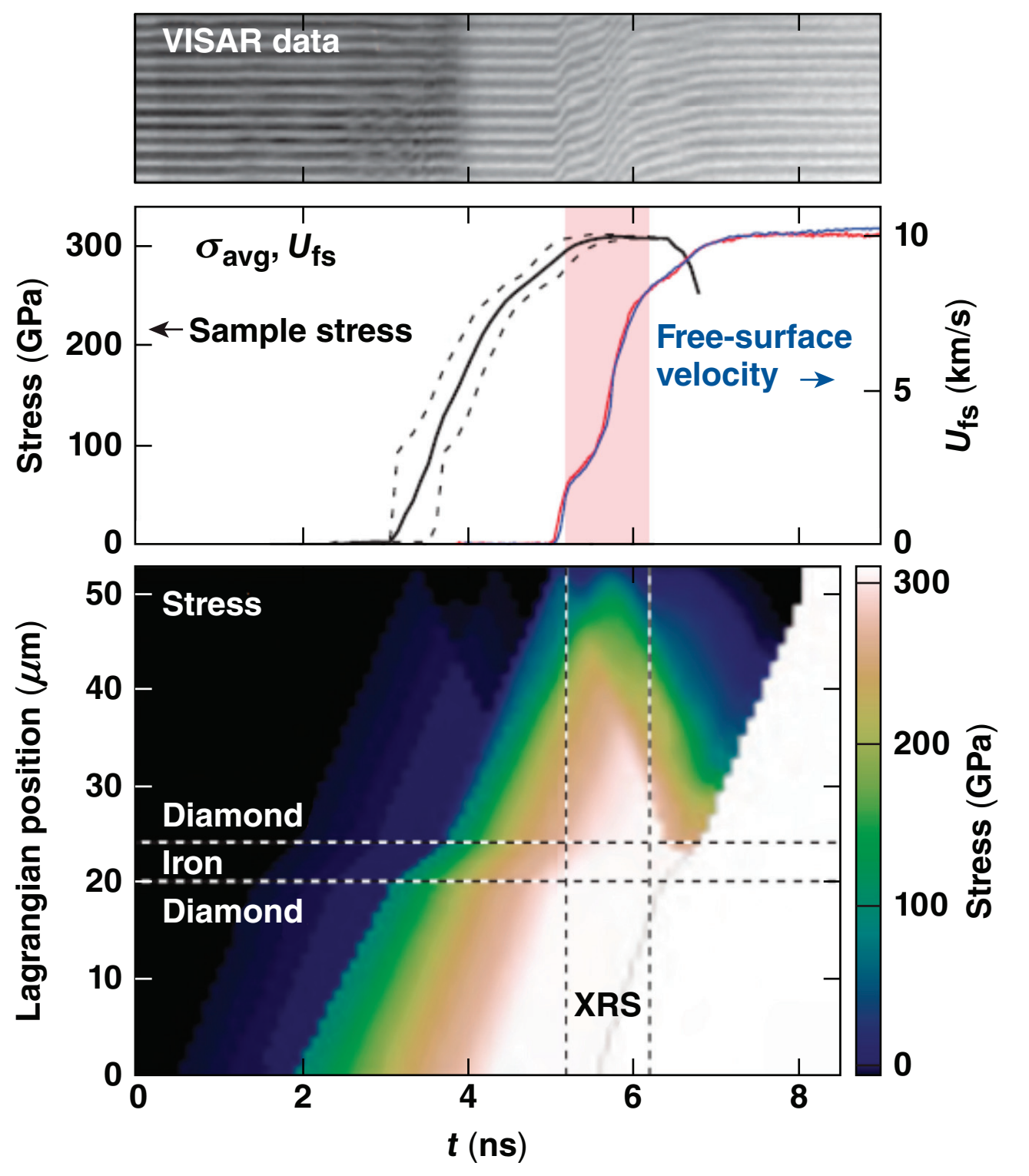
X-ray Thomson scattering (Z-Spec)

X-ray Thomson scattering for a germanium backlighter



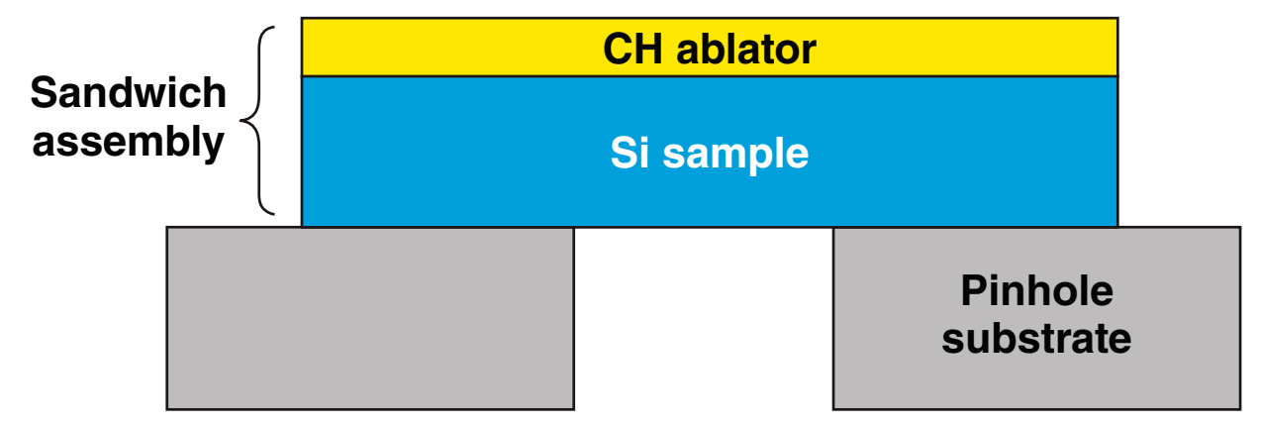
The spectrometer records spectrally resolved x-ray scattering

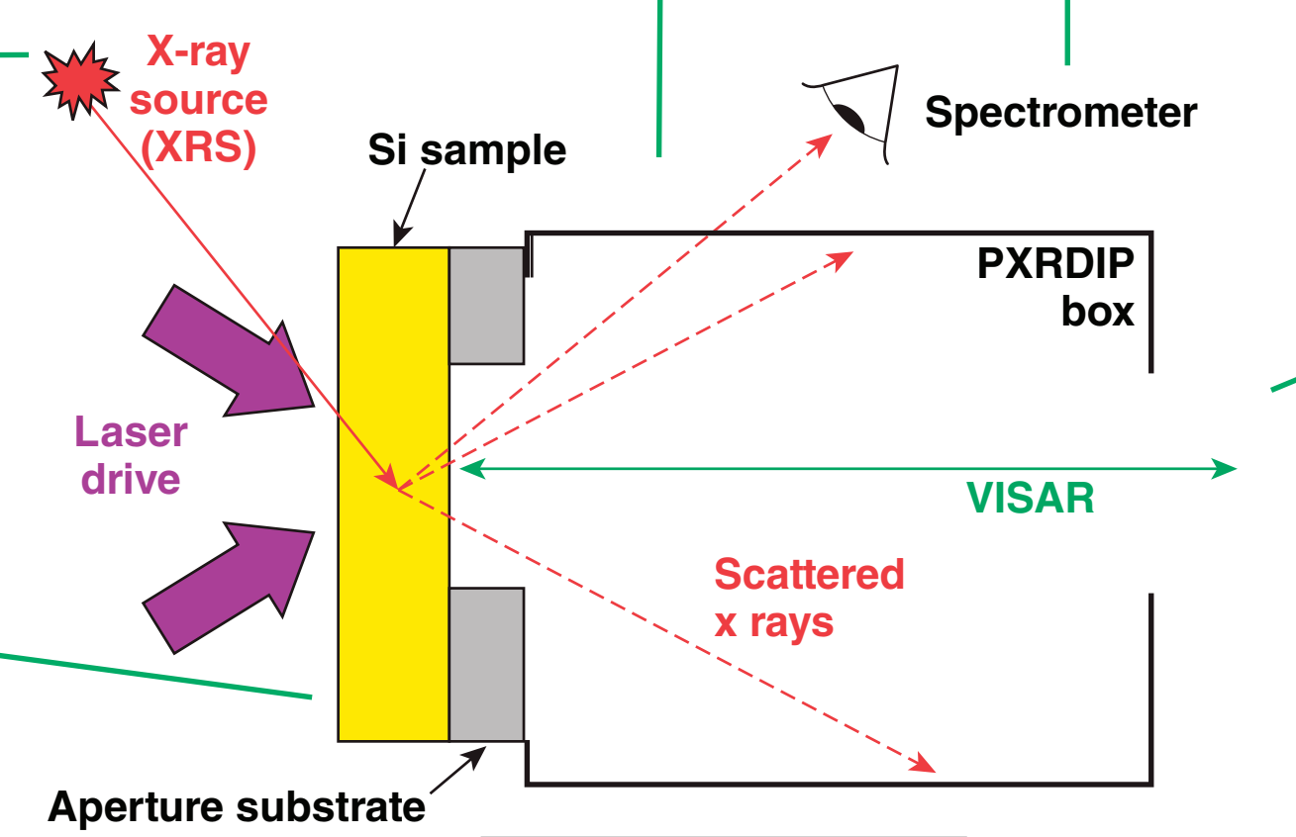
Velocimetry (VISAR)



VISAR measurement determines the breakout time and pressure versus time profile

Target assembly





Experimental Setup

PXRDIP: powder x-ray diffraction image plate

Approach

- Production of a precise and uniform state of warm dense matter by shock compression.
- Simultaneous determination of
  1. Angularly resolved x-ray diffraction with PXRDIP (for atomic structure and density),
  2. Spectrally resolved inelastic x-ray scattering with XRTS (for ionization state)
  3. Pressure and shock state with VISAR

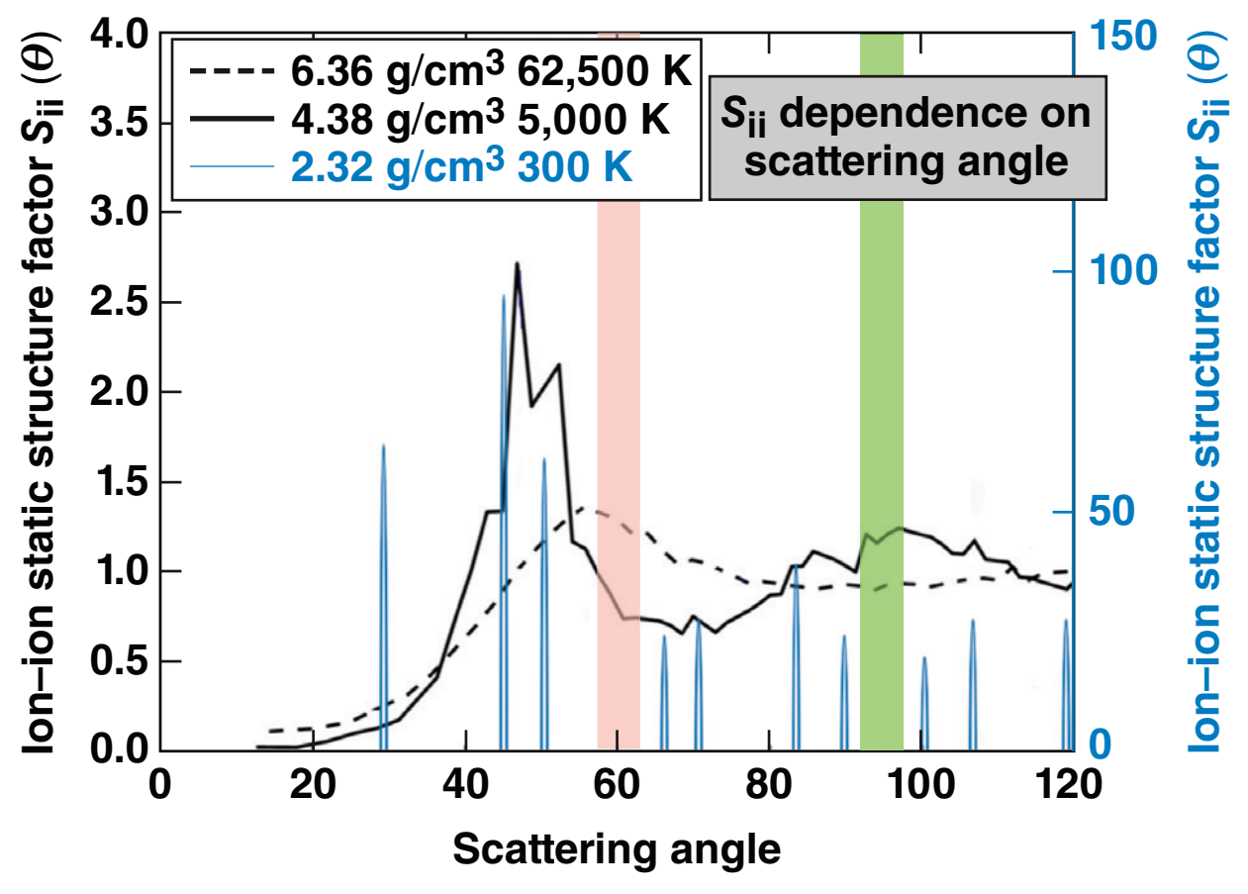
We plan to investigate
- Three conditions along the shock Hugoniot (a) solid, (b) liquid, (c) during melt
- Multiple scattering angles outside of the Bragg peak

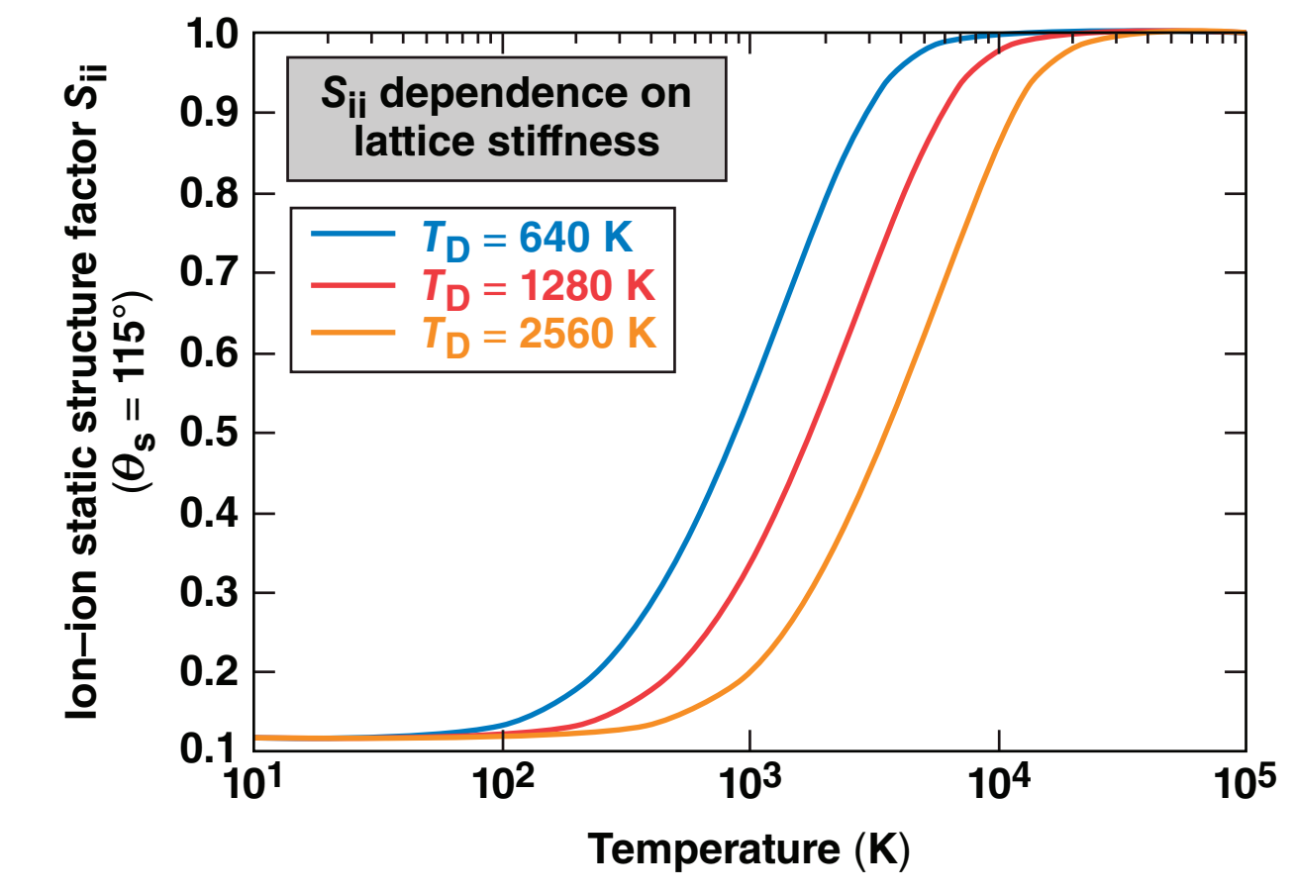
Structure factor (S_{ii}) and density estimate ⇒ **PXRDIP** Bragg diffraction measurements; **XRTS** Elastic feature measurements

Ionization estimate ⇒ **XRTS** Relative intensity of elastic and inelastic peaks; **VISAR** Reflectivity and conductivity measurements using the Drude model

Temperature estimate ⇒ **XRTS** Shift of the down-scattered inelastic structure; **PXRDIP** Debye–Waller measurements of scattering intensity data

Pressure estimate ⇒ **VISAR** Shock breakout data





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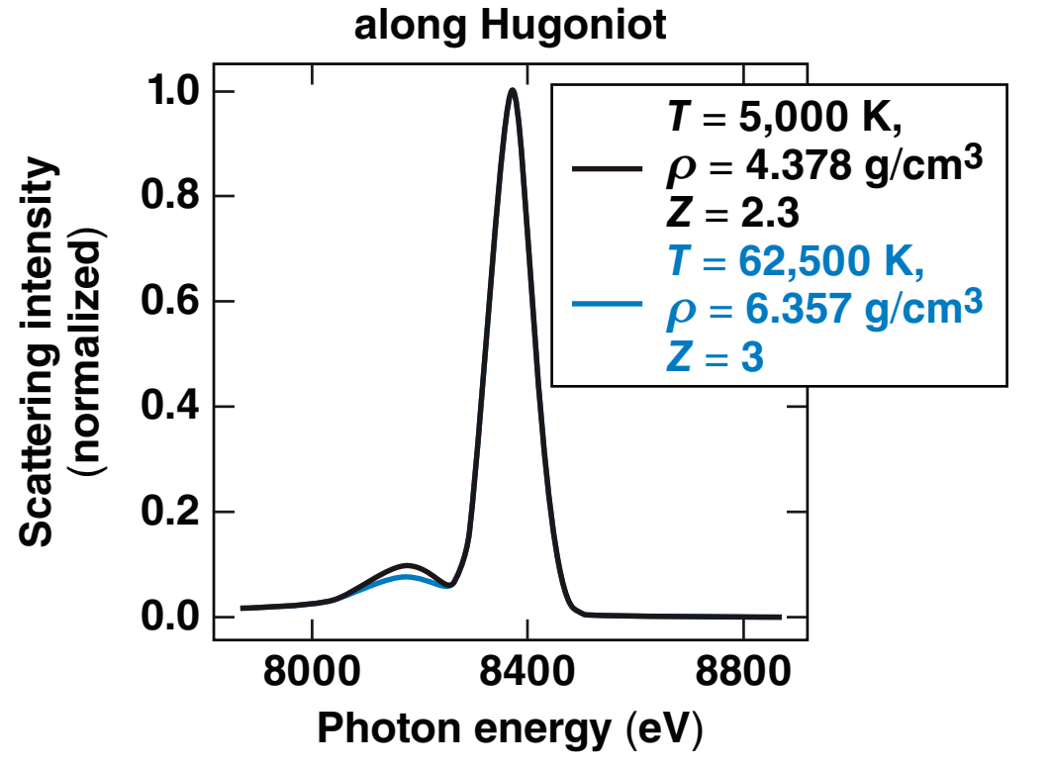

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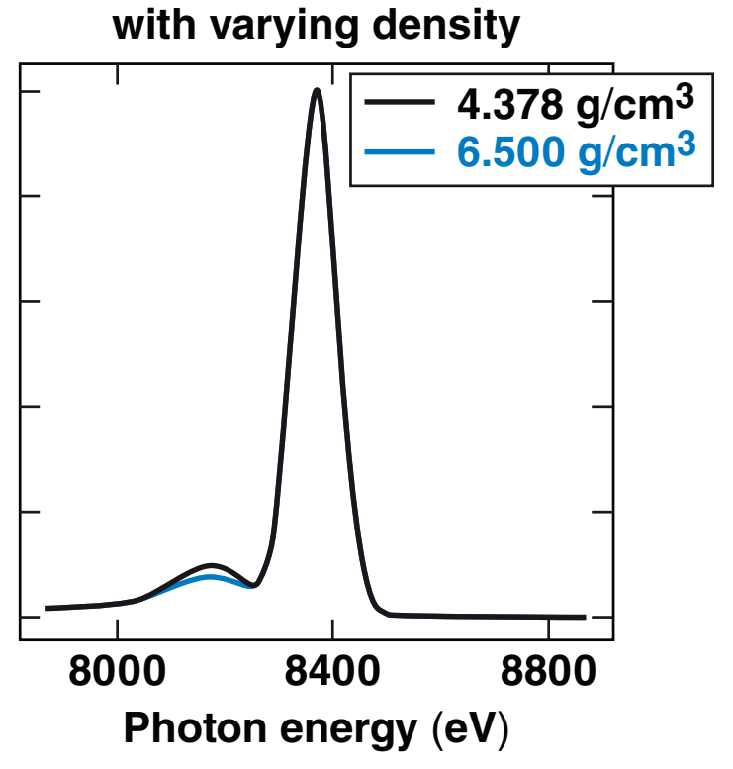

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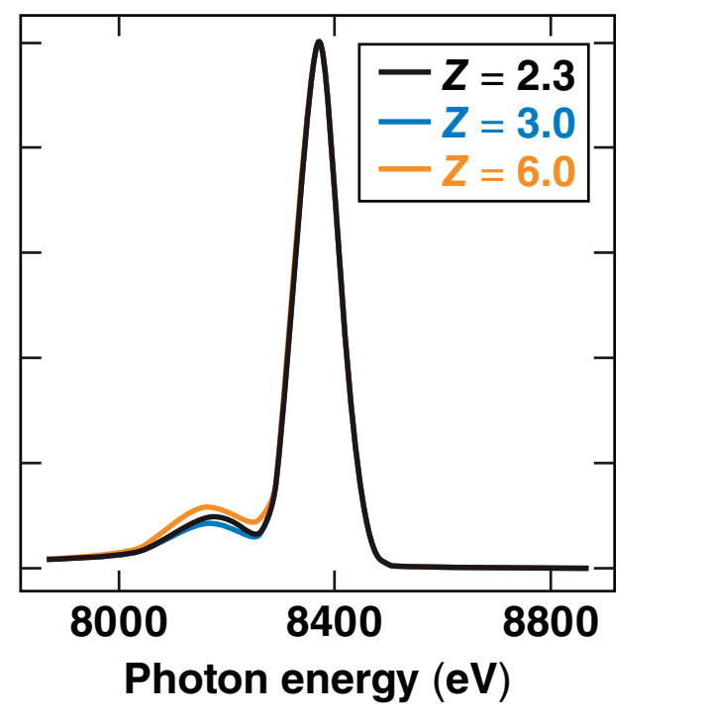

XRTS: x-ray Thomson scattering

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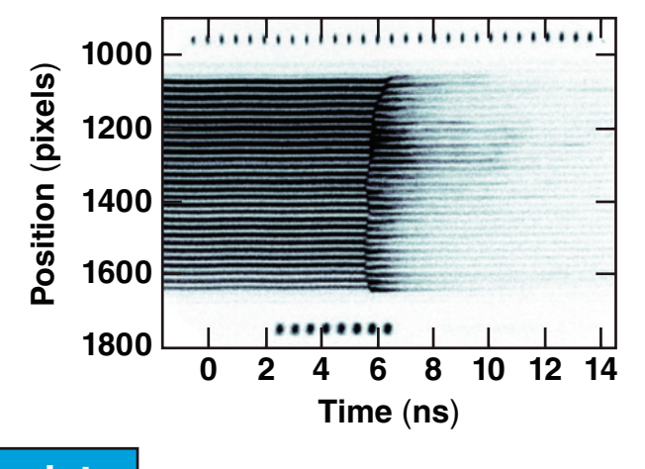
Preliminary Experimental Results

Shock breakout data (VISAR)



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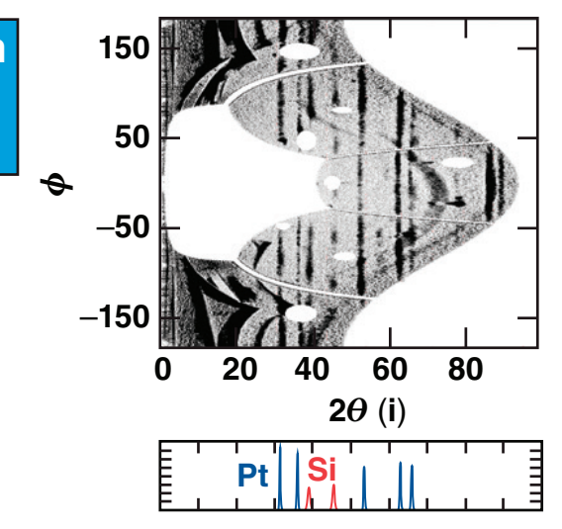

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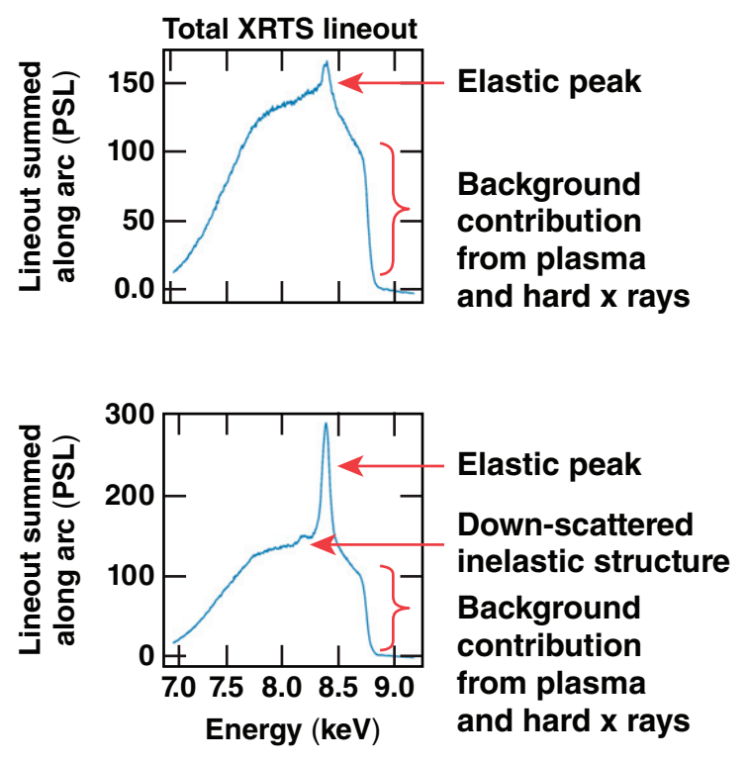
7.5-Mbar pressure



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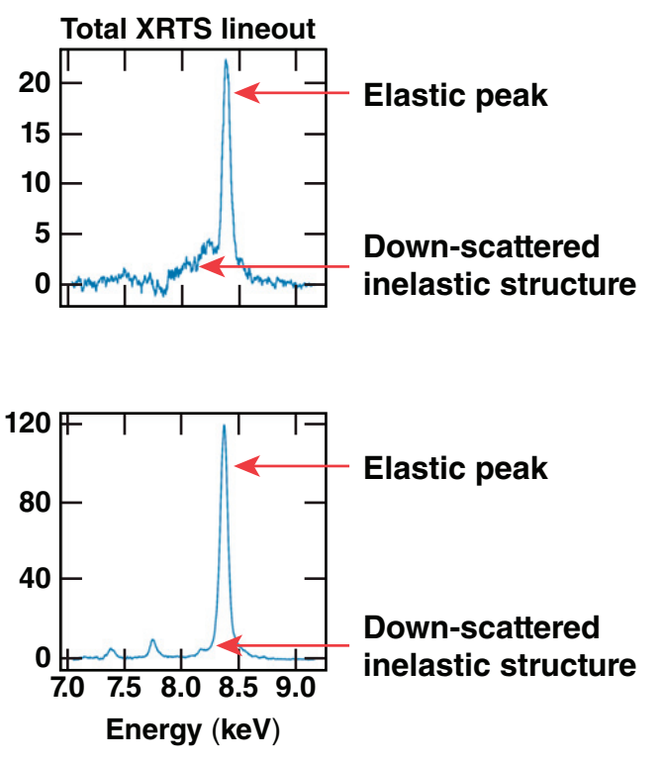

XRTS data at 95° scattering angle

XRTS data at 60° scattering angle

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References

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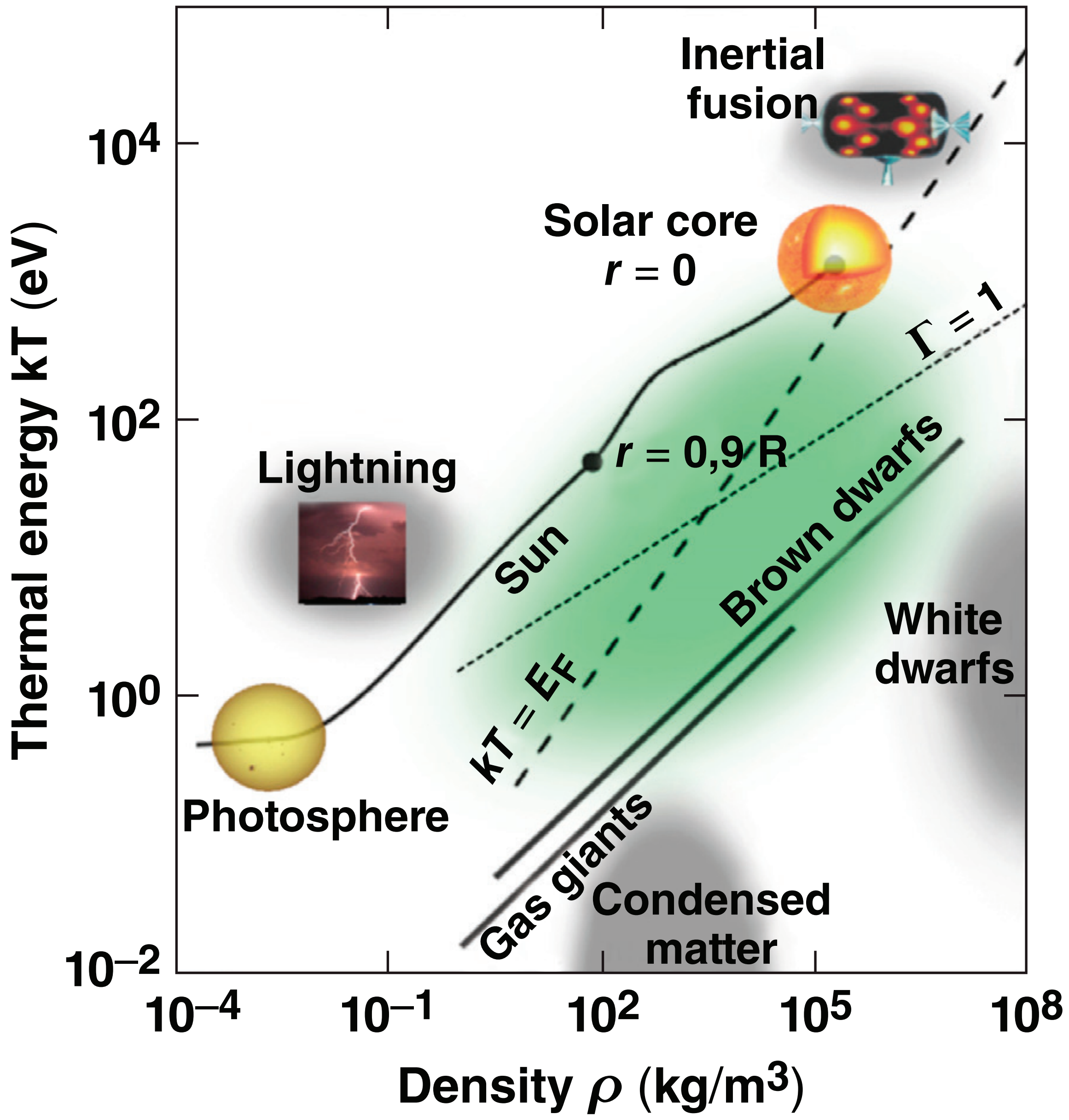

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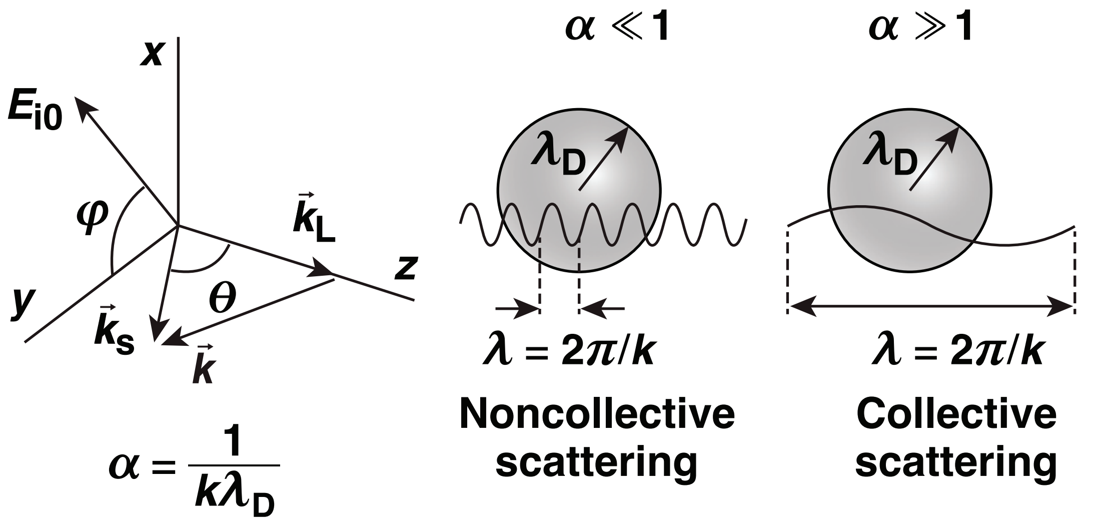

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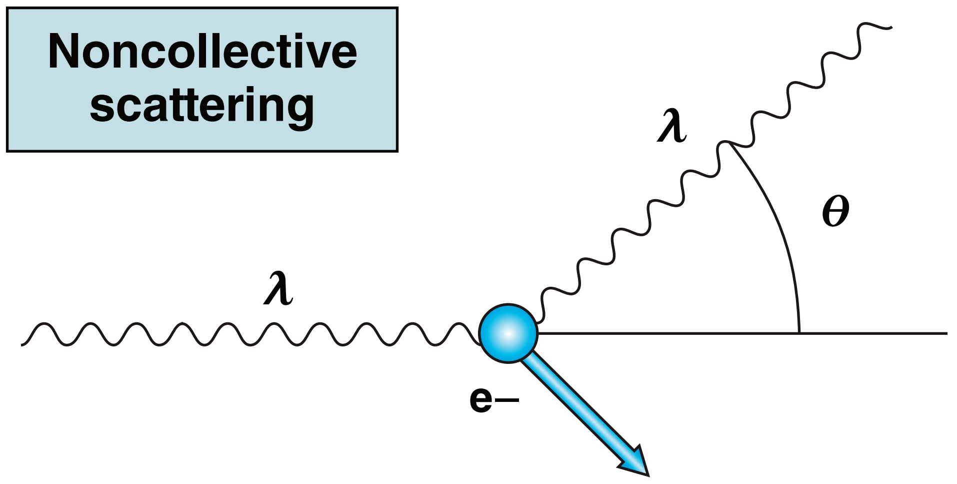

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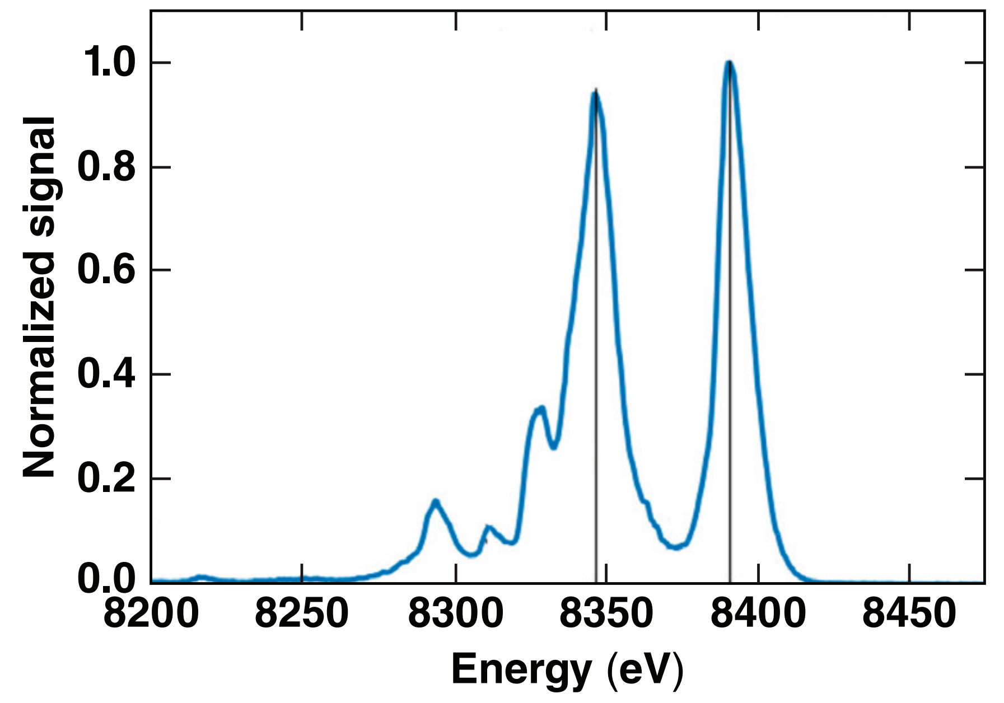

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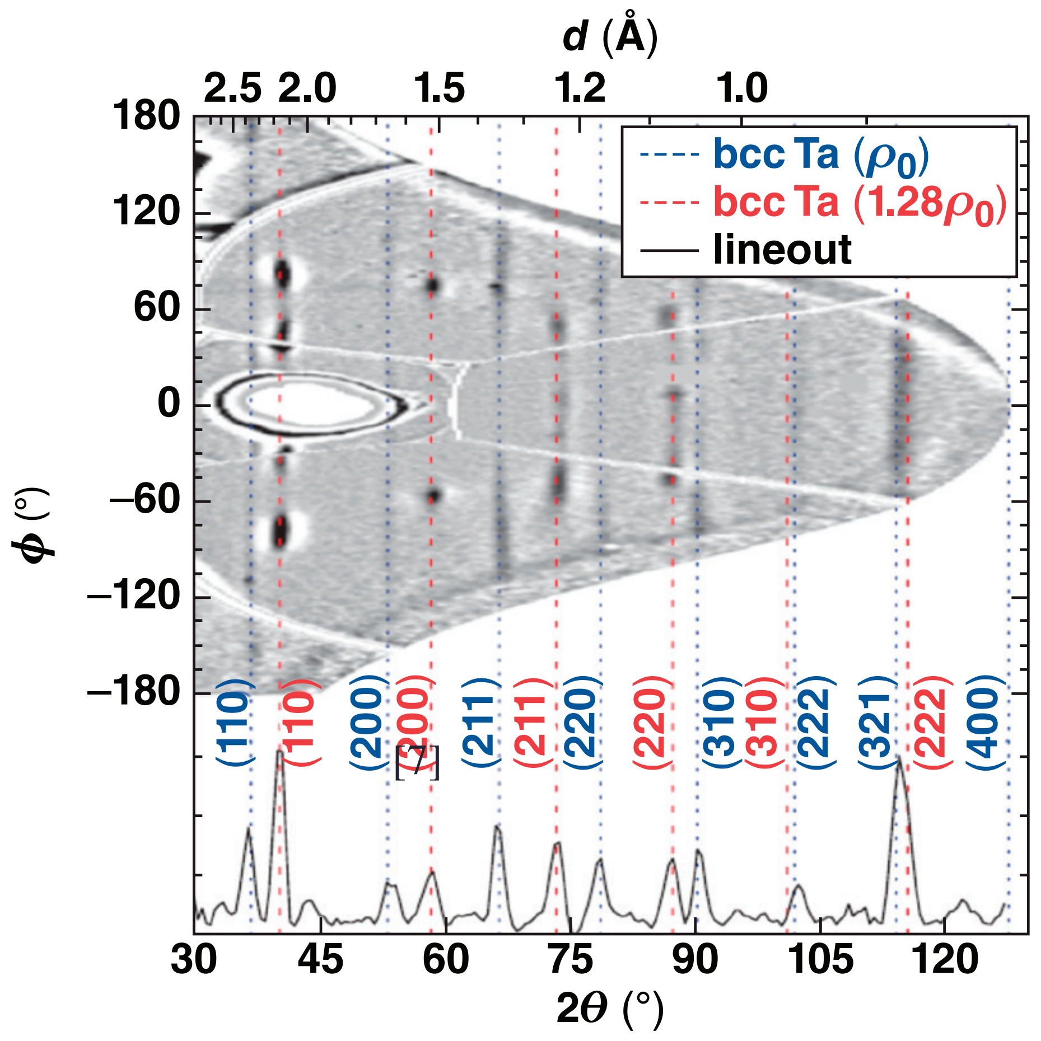

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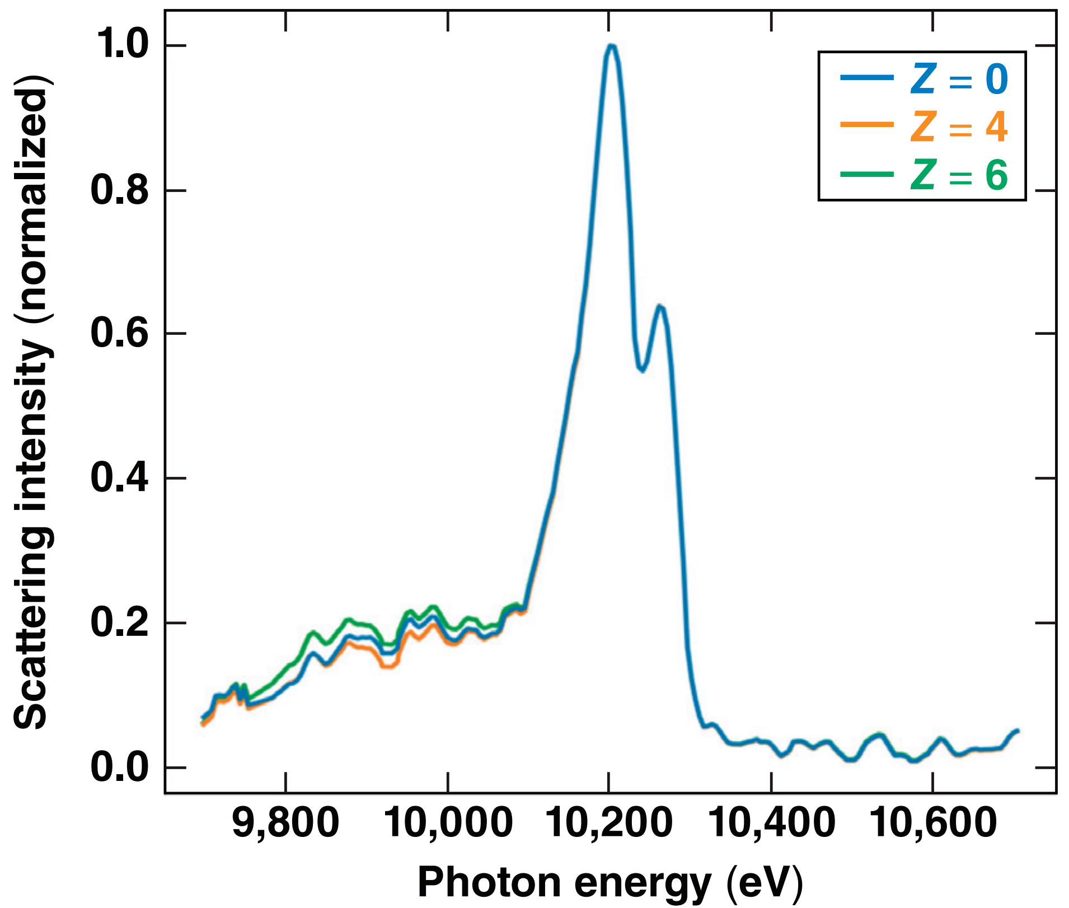

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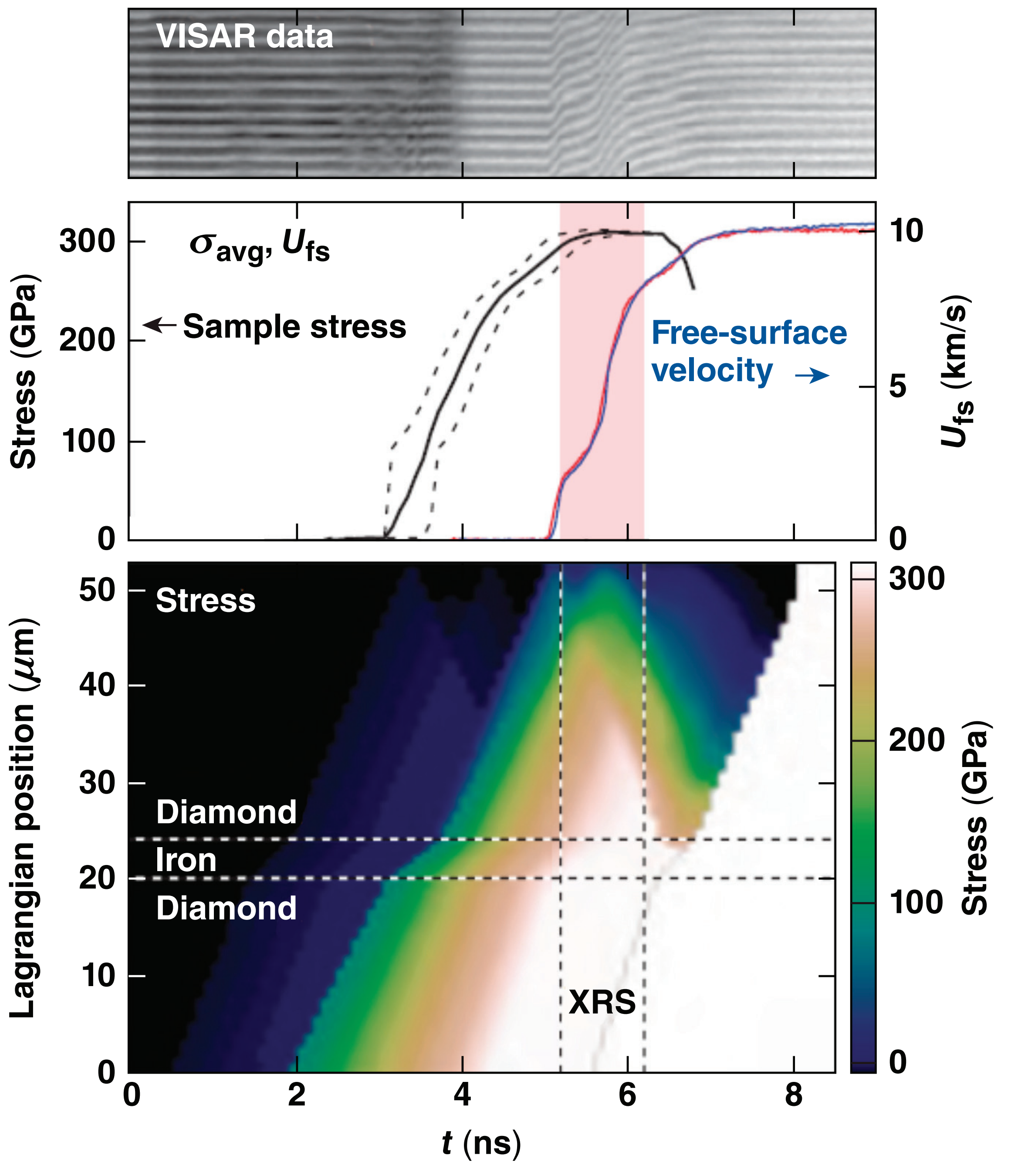

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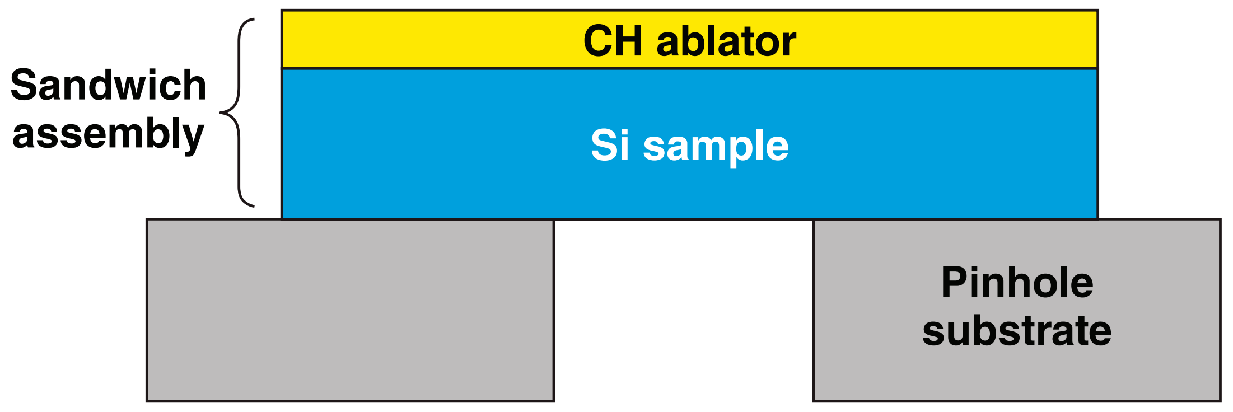

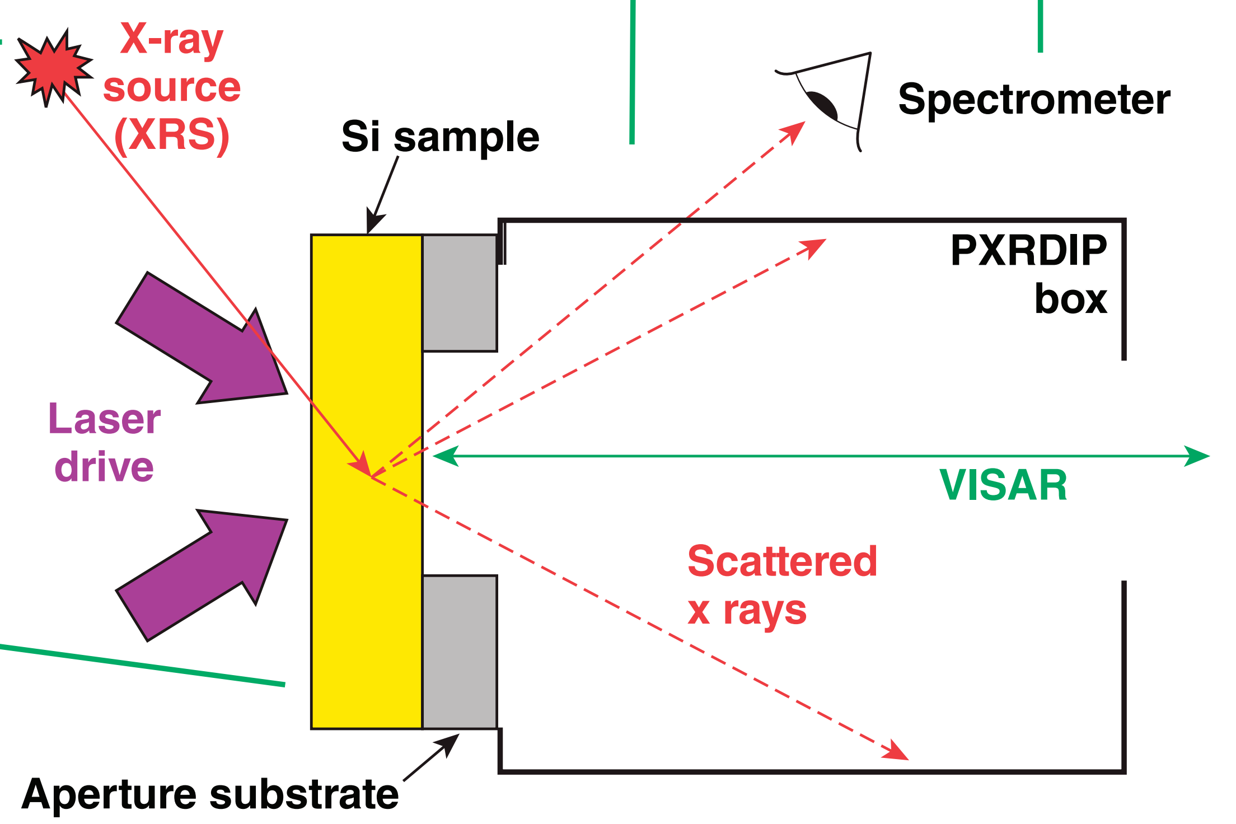

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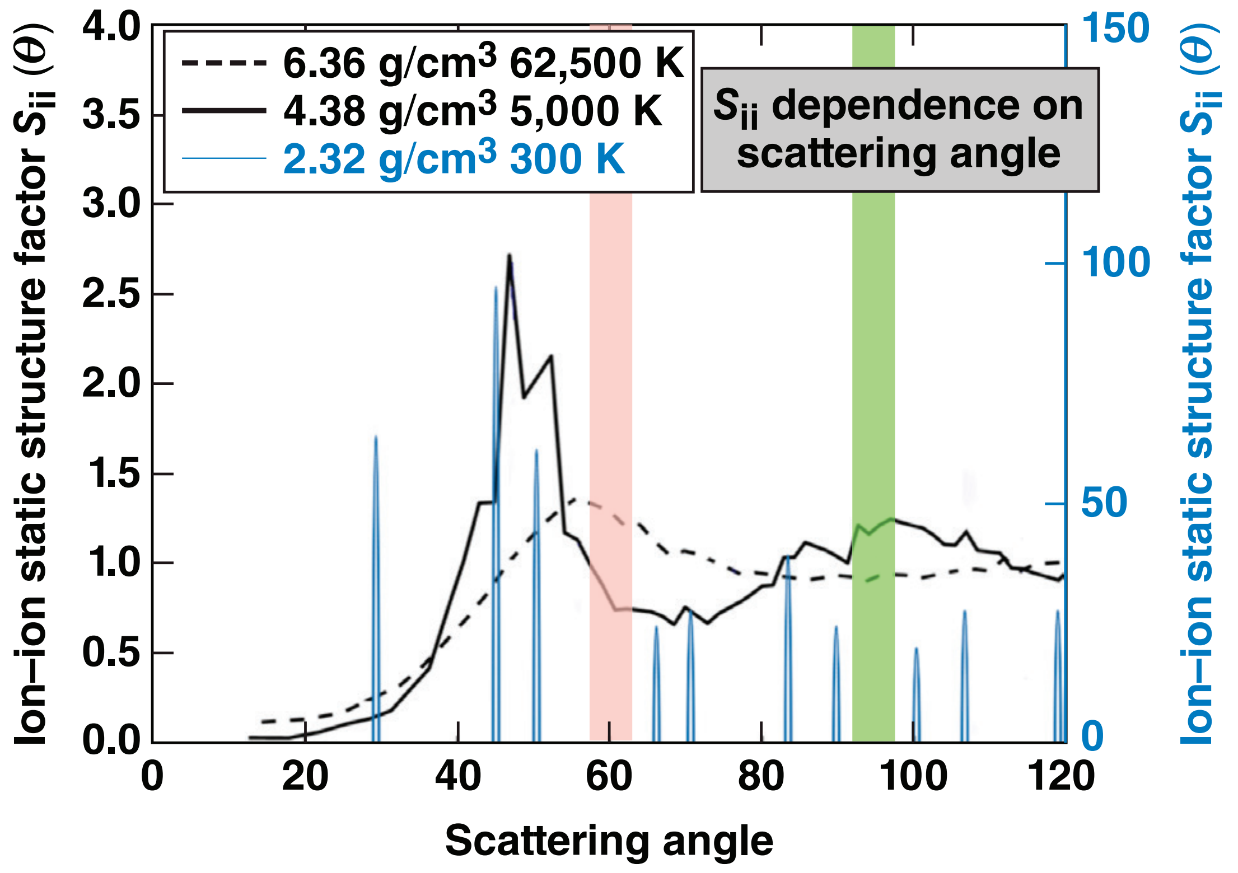

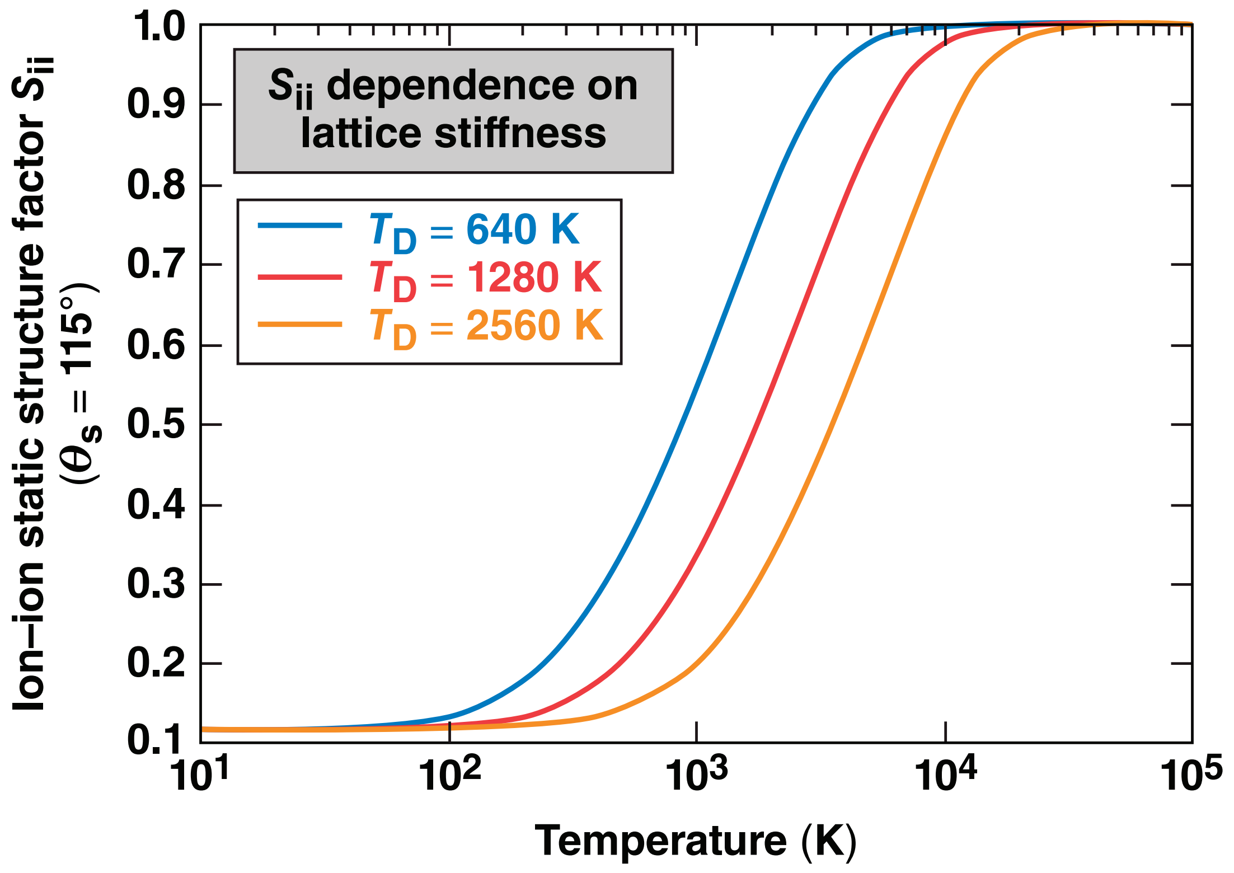

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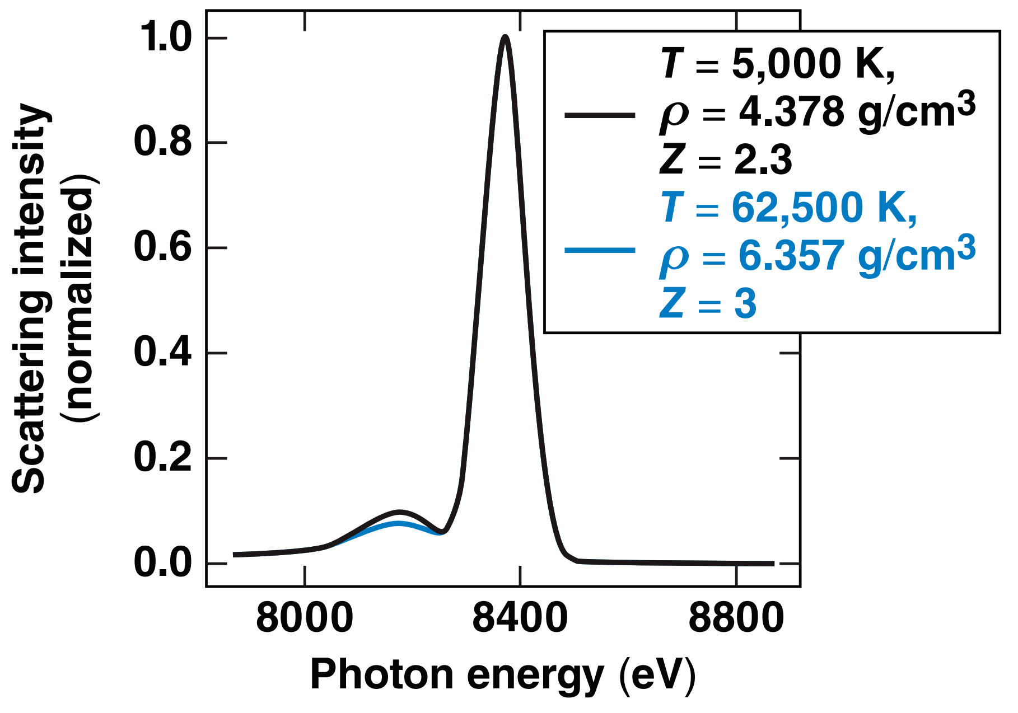

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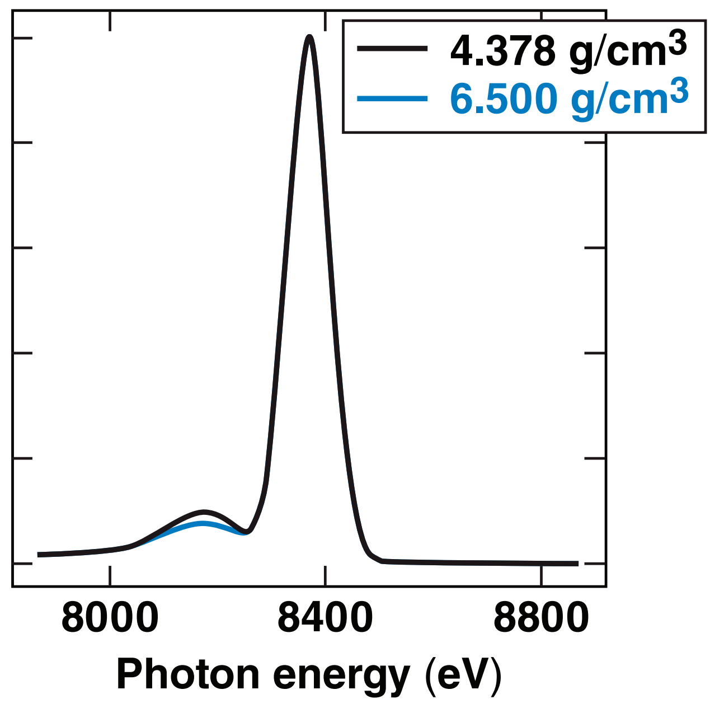

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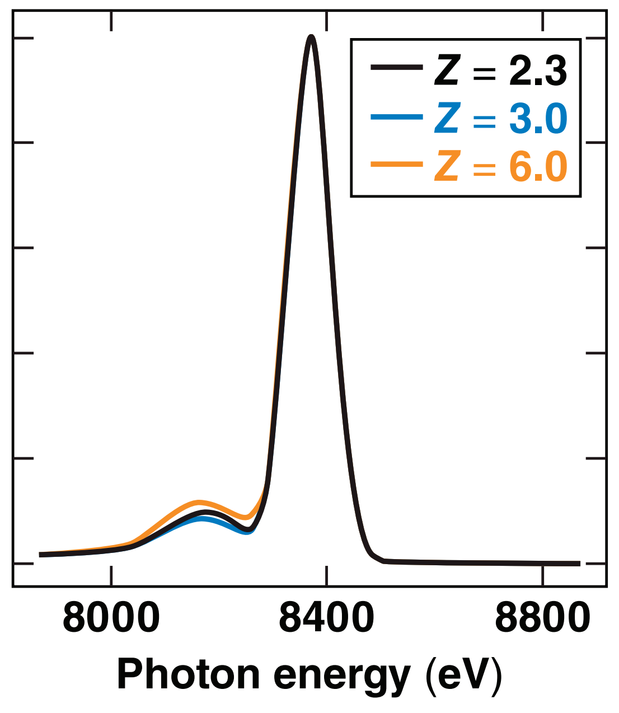

XRTS: x-ray Thomson scattering

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Preliminary Experimental Results

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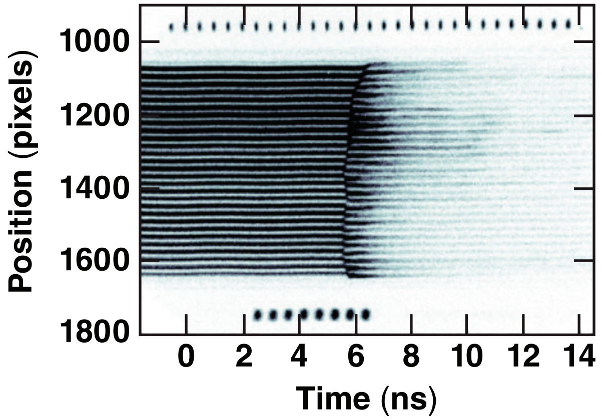

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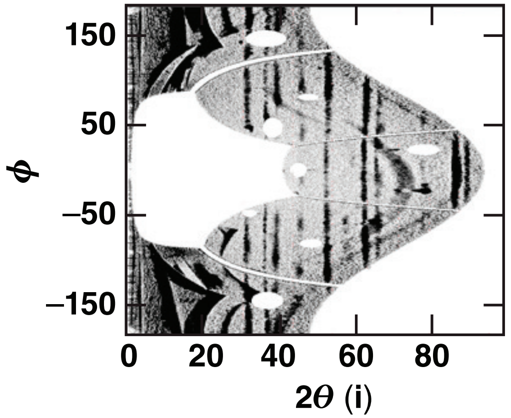

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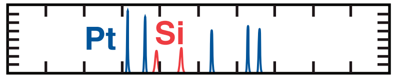


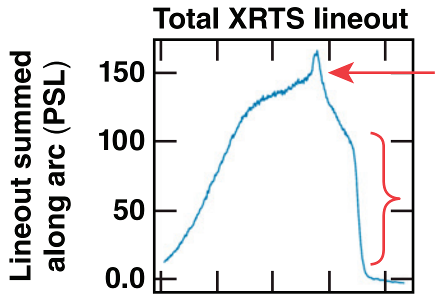
Diffraction pattern of Si at 2.1 Mbar

Scattering data (Z-spec)

7.5-Mbar pressure

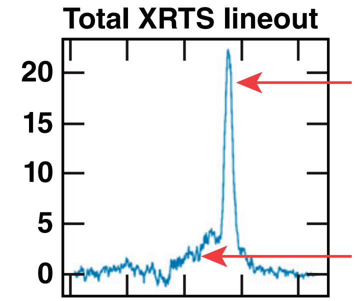
1.1-Mbar pressure

XRTS data at 95° scattering angle



Elastic peak

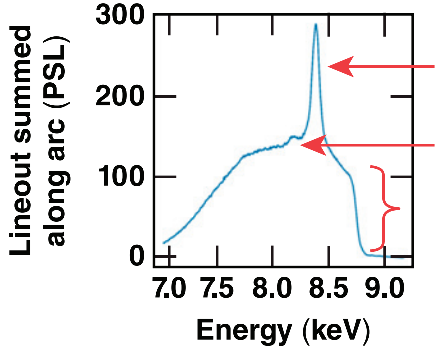
Background contribution from plasma and hard x rays



Elastic peak

Down-scattered inelastic structure

XRTS data at 60° scattering angle



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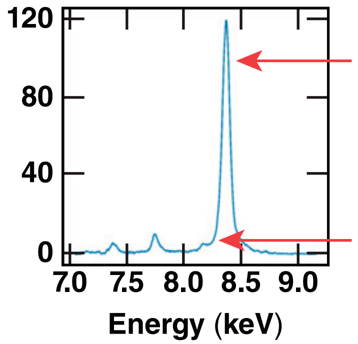

Elastic peak

Down-scattered inelastic structure

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