Time resolved x-ray scattering from warm materials

Roger Falcone

Physics Department, UC Berkeley
Advanced Light Source, LBNL

Warm materials are dynamic

- energy deposition of ~ one quanta per unit cell drives structural and other property changes that can be probed by time-resolved, x-ray scattering

- a variety of quanta can be utilized to “heat” materials
  - THz to far-IR directly drives phonon modes
  - near-IR excites electrons from valence to conduction bands
    - with adjustable excess-energy that rapidly couples to other modes
  - optical to uv excites electronic transitions and charge transfer states
  - soft x-rays couple core levels to valence states
  - hard x-rays penetrate and excite larger volumes

- coupling of excitation to various modes is defined by the time-scale
  - times << picoseconds can involve non-thermal processes
    - photochemistry, electron re-scattering
  - times >> picoseconds involve thermal processes
    - mode diffusion, ablation

- consider... relevant scale length divided by relevant velocity
Warm materials are studied by pump-probe techniques at a variety of facilities

- **Small-scale laboratories** provide intense, short-pulse lasers to create and probe warm (high-energy-density) materials
  - Probes include plasma x-ray sources, high-harmonic sources

- **Intermediate-scale facilities** include petawatt lasers, pulsed particle beams, x-ray synchrotrons, free-electron lasers, etc., and are widely accessible

- **Large-scale facilities** allow large volume studies to extreme high-energy-density conditions, but have limited access
  - NIF megajoule laser, Vulcan PW laser, OMEGA kJ laser, pulsed power
An example of HEDS science: liquid carbon

Molecular dynamics calculations predict:
- high density liquid: mainly $sp^3$ coordinated
- low density liquid: mainly $sp$ coordinated

**Phases of Carbon**

The goal is to study these phases under extreme conditions, liquid phases and melting lines.

**Diamond**

**BC8 (P > 1100 GPa)**
- Body Centered Cubic with 8 atom basis
- Theoretical phase proposed in analogy with Si
- Semi-metallic, not yet found experimentally

**Cubic**
- Metallic, not yet found experimentally
Liquid carbon

- Astrophysics and Planetary Science: State of Carbon in Giant Planets

- High pressure research: Theoretical limit of diamond based technology

- Technological: Capsules for *Inertially Confined Fusion* (Carbon as an ablator material)
Phase Diagram of Carbon

- Predicted maximums in melting lines
- Triple point
- Negative slope of Diamond-BC8 transition
- Experimentally verified negative melting slope
  - \(( P > 500 \text{ GPa})\) by Shock experiments (Eggert et al. 2007)
Melting lines obtained by the two-phase simulation method

- Even with the most realistic molecular dynamic simulation, melting lines are not trivial to obtain
- Density Functional Molecular Dynamics on 128 carbon atoms
- Quantum mechanical electrons and Classical Ions
- Ab initio (no fitted parameters)
- Solid and Liquid initially present in same simulation
- Interface evolves at a given $P$ and $T$
- Most stable phase grows
- Melting line is bracketed recursively

Correa et al. PNAS 103(5) (2006)
Quantum Molecular Dynamics does a great job in terms of predicting structural properties:

but we need to go further...
Can we reproduce experimental results and predict specific results for new conditions?
High-energy-density carbon has been probed by x-ray absorption (near and extended edge)

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

S. Johnson, et al
Predicting high-T absorption spectra

![Graph showing absorption spectra](image-url)
Perturbed liquid state structure and dynamics can be probed by x-ray scattering (small and wide angle).
Time-resolved structural changes in H₂O are seen upon charge injection. Difference signal at 100 ps following charge injection implies molecular re-orientation around injected charge with similarities to thermally induced changes. A. Lindenberg.
Laser-sliced x-ray pulses from synchrotrons are used as tunable probes of HED matter

X-Ray FELs produce x-ray pulses: eventually may be tunable for spectroscopy
High-order harmonic radiation from multi-TW lasers produces intense soft x-ray fluxes for pump-probe HED science.

- tunable soft x-ray peak power > 1 MW
- beam divergence < 1 mrad
- shot to shot fluctuations < 10%
- pulse length < 30 fs
- spatial and temporal coherence
- examine non-linear phenomena

Allison, Belkacem, Hertlein, VanTilborg
Ultrafast “x-ray streak cameras” enable high-speed recording of atomic dynamics

With space (1d) and time resolution, can record changing spectral response
Fastest streak cameras can resolve << picosecond

Dynamic mode
1000 shots

Static mode
1000 shots

Au photocathode

Jun Feng, Howard Padmore LBNL
Ultra-fast X-ray Streak Cameras at the ALS

Transmission Streak Camera

Reflection Streak Camera
Laser-generated strain, bond-breaking, and hot electron-phonon coupling can initiate a solid-to-liquid phase transition which can be probed by ultrafast x-ray scattering.

Disordering of a lattice through bond-breaking observed at even shorter times at the SPPS

- (111) and (220) reflections measured
- Non-thermal melting observed

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

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

SPPS Collaboration
High Energy Density Matter occurs widely

- **Hot Dense Matter (HDM)** occurs in:
  - Supernova, stellar interiors, accretion disks
  - Plasma devices: laser produced plasmas, Z-pinches
  - Directly and indirectly driven inertial fusion experiments

- **Warm Dense Matter (WDM)** occurs in:
  - Cores of large planets
  - X-ray driven inertial fusion experiments

[Hydrogen phase diagram image]
An example: Inertial Confinement Fusion experiments depend on behavior of WDM

- Laser is focused through hole in hohlraum
- Laser impinges on wall creating a plasma
- Wall plasma radiates x-rays
- X-ray are absorbed in the pusher part of sphere
- Heated material drives a shock
Designs for ICF involve “gentle” compression and the WDM regime is sampled

- The $\rho$-$T$ paths for Be and DT traverse the WDM regime

- Time history of Be pusher and DT fuel

- $\rho$-$T$ Track of Be pusher and DT fuel

- DT EOS may be modified by WDM effects
  - dissociation, pressure ionization
  - changes the detailed performance of ICF ignition capsules

- Be will have WDM effects: ionization
The defining concept of warm dense matter (WDM) is coupling weakly coupled plasmas.

- Plasma seen as separate point charges
- Plasma is a bath in which all particles are treated as points

When either $\rho$ increases or $T$ decreases, $\Gamma > 1$

- Particle correlations become important
- Energy levels shift and ionization potentials are depressed
WDM is defined by temperature relative to the Fermi energy

- Fermi energy, \( E_{\text{Fermi}} \), = maximum energy level of e\(^-\) in cold matter

- When \( T \ll E_{\text{Fermi}} = T_{\text{Fermi}} \) standard condensed matter methods work

- When \( T \sim T_{\text{Fermi}} \) one gets excitation of the core
  - Ion - e\(^-\) correlations change and ion-ion correlations give short and long range order

![Diagram showing energy levels and Fermi energy](image)
WDM, created by isochoric heating using short pulses, will isentropically expand sampling phase space.

- XFEL can heat matter rapidly and uniformly to create:
  - Isochores (constant $\rho$)
  - Isentropes (constant entropy)
  - Using underdense foams allows more complete sampling

- Isochores (constant $\rho$)
- Isentropes (constant entropy)
Ablation of a surface under high energy flux

G. Gilmer, B. Sadigh, LLNL
Thomson scattering enables direct determination of both material and plasma properties

• 25 eV, $4 \times 10^{23}$ cm$^{-3}$ plasma XFEL produces $10^4$ photons from the free electron scattering

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

By varying the scattering angle, collective modes of dense matter are probed
X-rays provide a unique probe of HED matter

- Due to absorption, refraction, & reflection, visible lasers cannot probe high density

- X-ray scattering from free electrons provides a measure of the $T_e$, $n_e$, $f(v)$, and plasma damping

- x-ray FEL scattering signals will be well above noise for HED matter
Scattering parameter
\[ \alpha = \frac{1}{(k\lambda_D) \sim \lambda_L / (4\pi \lambda_D \sin(\theta/2))} \]
\( \lambda_L \) (incident wavelength), \( \theta \) (scattering angle), \( \lambda_D \) (screening length)

\( \alpha > 1 \) : collective scattering regime
Scattering on density fluctuations outside Debye sphere
\[ \rightarrow \text{Sensitive to collective motion: Plasmon peak from electron plasma oscillation} \]

\( \alpha < 1 \) : non-collective scattering regime
Scattering on density fluctuations inside Debye sphere
\[ \rightarrow \text{Sensitive to random thermal electron motion: Compton peak.} \]
Compton/Thomson scattering with optical probe

Fermi degenerate plasma regime: \( T_e < T_F \)
Strongly coupled plasma regime: \( T_e > T_F, \Gamma_{ee} > 1 \)
\( \Gamma_{ee} \) = Coulomb potential energy/Kinetic energy of free electrons
Ideal plasma: \( \Gamma_{ee} < 1 \)

O. L. Landen et al. JQSRT (2001)

\[ \theta = 180^\circ, \alpha = 0.3 \]

Non-collective Thomson Scattering \((\lambda^* < \lambda_D)\)

Optical Laser

Conventional, optical wavelength
\[ \lambda = \lambda_0 [1 \pm (v/c)\sin(\theta/2)] \]

Boltzmann distribution

Intensity

Wavelength

\( \theta = 180^\circ, \alpha = 0.3 \)

\( \lambda = 2.4 \text{ Å} \)

\( \lambda = 2400 \text{ Å} \)

\( \alpha = 0.1 \)

\( \alpha = 0.3 \)

Fermi degenerate plasma regime

X-ray wavelength

Conventional, optical wavelength

Solid Be

\( n_e \text{ (cm}^{-3}\text{)} \)

\( T_e \text{ (eV)} \)
Compton/Thomson scattering with x-ray probe in dense matter

Fermi degenerate plasma regime: $T_e < T_F$

Strongly coupled plasma regime: $T_e > T_F$, $\Gamma_{ee} > 1$

$\Gamma_{ee} =$ Coulomb potential energy/Kinetic energy of free electrons

Ideal plasma: $\Gamma_{ee} < 1$

$\theta = 180^\circ$, $\alpha = 0.3$

X-ray wavelength

Conventional, optical wavelength

$\lambda = \lambda_0 [1 + 2(h\nu/mc^2)\sin^2(\theta/2) \pm (v/c)\sin\theta/2]$

Compton peak

Rayleigh peak

Fermi or Boltzmann distribution

$\Gamma_{ee} = \frac{\text{Coulomb potential energy}}{\text{Kinetic energy of free electrons}}$

$T_e (\text{eV})$

$n_e (\text{cm}^{-3})$

Strongly coupled plasma regime

Fermi degenerate plasma regime

$\lambda = 2.4 \text{ Å}$

$\lambda = 2400 \text{ Å}$

$\alpha = 0.1$

$\alpha = 0.3$

$\theta = 180^\circ$, $\alpha = 0.3$

O. L. Landen et al. JQSRT (2001)
Scattering regimes in the $\rho$-$T$ plane

In dense plasmas
- standard theoretical approaches fail
- theoretical uncertainties are large

Collective scattering in dense plasmas
- probes transition region
X-ray source
- penetrates dense plasmas
Forward scattering and plasmon in dense matter

Scattering parameter $\alpha$

$$\alpha = 1/(k\lambda_D) \sim \lambda_L/(4\pi \lambda_D \sin(\theta/2)) \rightarrow \lambda^*/\lambda_D$$

$\alpha > 1$ : Collective regime, $\lambda^* > \lambda_D$.

Orderly oscillatory behavior under the long-range Coulomb forces.

The density fluctuation in the plasma behave collectively and oscillate around $\omega_p$.

With x-ray probe for WDM, strong asymmetry or almost gone of blue-shifted peak.

From the plasmon peak, we can have better accurate information about $T_e$!!

From the fluctuation-dissipation theorem

$$S(k, \omega) = \frac{1}{2\pi N} \int e^{i\omega t} < \rho_e(k, t) \rho_e(-k, 0)> dt,$$

$$S(k, \omega) = -\frac{\varepsilon_0 \hbar k^2}{\pi e^2 n_c} \frac{1}{1 - e^{\hbar \omega/k_BT}} \text{Im} \varepsilon^{-1}(k, \omega),$$

$$\frac{S(k, \omega)}{S(-k, -\omega)} = \exp(-\frac{\hbar \omega}{k_BT})$$

Sensitive to $T_e$
Sensitivity of scattering spectra to plasma condition

**Forward scattering:**
Plasmon peak position provides the local electron density.

**Backward scattering:**
Compton peak intensity is sensitive to ionization state.
Sensitivity of scattering spectra to plasma condition

Forward scattering:
Plasmon peak position provides the local electron density.

Backward scattering:
Compton peak position is less sensitive to electron density.
Current Thomson scattering experiments are done at large laser facilities.
Omega laser = 17 beams, 480 J each, total energy ~8.7 kJ,

Target: Be-foil, thickness 0.24 mm
Laser intensity: $3 \times 10^{14}$ W/cm$^2$
Pulse duration: 3 ns

WDM with ~43 µm depth is generated over 500 ps with uniform condition.
We obtained plasmon scattering from shock compressed Be. The position of the plasmon resonance yields density $n_e = 1 \times 10^{23} \text{ cm}^{-3}$, $T_e = 10 \text{ eV}$ at 3 ns.

2 ns drive beams at $t = 0$; analyze plasma between 2.6-3.4 ns.
- for x-ray pulse backlighting of warm matter on high-energy laser systems, we use multiple laser beams with about 10,000 J in a few ns, for pumping a plasma on a surface that radiates K-\(\alpha\) and He-\(\alpha\) x-rays

- this converts to about 1 J of x-ray photons radiated into 4\(\pi\)

- there is then about 1 mJ for use in illuminating the sample, within the collected solid angle

- this probe x-ray beam compares well in energy per pulse with the LCLS per pulse energy, which has 1 mJ

- LCLS pulses will be more collimated, narrower BW, and shorter in duration (~ 200 fs)
Materials science and lattice dynamics at ultrahigh pressures and strain rates define a frontier of condensed matter science.

Unexplored regimes of solid-state dynamics at extremely high pressures and strain rates will be accessible on NIF.

[D.H. Kalantar et al., PRL 95,075502 (2005); J. Hawreliak et al., PRB, in press (2006)]
Intense x-ray fluxes from LCLS will enable real-time in situ measurements of microstructure evolution at high pressure.

**What is the timescale of the bcc-hcp phase transformation in Fe?**

**Current measurement limited to timescales >> psec**

**Simulations predict subpicosecond phenomena observable using LCLS**

[Kadal et al., *Science* (2002)].
X-Ray FELs will enable a range of HED experiments (talk by R.W. Lee)

• Creating Warm Dense Matter
  • Generate \(\sim 10\) eV solid density matter
  • Measure the equation of state

• Probing dense matter with Thomson Scattering
  • Perform scattering from solid density plasmas
  • Measure \(n_e, T_e, \langle Z\rangle, f(v)\)

• 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
Challenges to theorists: Absorption and scattering cross-sections

At the highest intensities (i.e., up to requirements for atomic-resolution single macromolecule imaging: $10^{22}$ W/cm$^2$):

- Does the ratio of absorption to scattering stay the same, affecting single macromolecular imaging studies (dependence of damage and signal)?

- Will Raman processes allow useful broadbanding of the LCLS pulse, for absorption spectroscopy (NEXAFS, etc.)?

- Will transparency or guiding effects be important, for deeper penetration in HEDS studies?
Challenges under "warm" conditions in condensed matter, materials physics, and plasma physics can be addressed:

- understand the dynamic interplay between electronic structure (energy levels, charge distributions, bonding, spin) and atomic structure (coordination, bond distances, arrangements)

Fundamental time scales range from picoseconds (conformational relaxations in molecular systems, and electron-lattice energy transfer times in solids), to ~100 fs (vibrational periods), to ~10 fs (electron-electron scattering), to <1 fs (electron-electron correlations)

X-rays are ideal probes of atomic structure, electronic structure, and plasma properties

New x-ray sources should enable the application of x-ray spectroscopic and scattering techniques (XANES, EXAFS, XMLD, XMCD, RIXS) on fundamental time-scales.