Isochoric Laser Heating for the study of Warm Dense Matter

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Outline

• Introduction
  – What is *Warm Dense Matter*?
  – *Idealized Slab Plasma & Isochoric Laser Heating*

• Physics under non-equilibrium, extreme conditions
  – Electrical conductivities
  – Lattice stability
  – Band structure and electron density of state

These are university-scale experiments
What is Warm Dense Matter?

- **WDM introduced in 2000**, characterized by
  - \( kT \sim E_{\text{Fermi}} \)
  - \( \Gamma_{ii} = \left[ \frac{\text{P.E.}}{\text{K.E.}} \right]_{\text{ions}} > 1 \)

- **Many-body, disordered system**
  - Partial electron degeneracy
  - Excited electronic states
  - Pressure ionization
  - Strong ion-ion correlation

- **High-pressure system**
  - WDM is also HED Matter (>1 Mbar or \(10^{11} \text{ J/m}^3\))
  - Inertial confinement only
  - Rapid expansion
Warm Dense Matter is both fundamentally important and of broad relevance

- As finite-temperature condensed matter or strongly-coupled degenerate plasma, WDM is the basis for understanding the convergence of condensed matter and plasma science

- WDM finds applications in many disciplines
  - High Energy Density physics
  - Inertial Confinement Fusion
  - Shock physics
  - Material science
  - Planetary science
WDM is an uncharted frontier as readily seen from the widely use EOS table - Sesame

Sesame EOS for copper [K. Trainor, JAP (1983)]

APW - Electron band theory at 0K

GRAY - Semi-empirical Gruneisen-Debye theory for solid-melt-liquid

MC - Soft Sphere (Expanded liquid, vapor)

OCCIPITAL - Saha ionization equilibrium

TFNUC - Thomas-Fermi-Kirzhnits theory with semi-empirical nuclear corrections

ACTEX - Perturbation theory for high temperature ionization equilibrium

A critical void appears in the Warm Dense Matter regime
A major hurdle in WDM studies is the lack of single-state data

- Laboratory WDM tends to be non-uniform due to hydrodynamic expansion at extreme pressure

- Properties measured on non-uniform or multi-state systems can only be compared with theory through code simulations that take into account gradient effects

Unambiguous tests of theory requires
- Single-state physical data
- Directly observed state parameters
The concept of an *Idealized Slab Plasma* offers a means to achieve single-state measurements

- An *Idealized Slab Plasma* is a planar plasma that can be considered as a single uniform state in which any residual non-uniformities will impose negligible impact on the measurement of its uniform properties.

- The state can be characterized from direct measurements such as mass density and energy density.
An approach to realize the ISP concept is Isochoric Laser Heating of a solid

- Laser heating in the $fs$ time scale mitigates hydro expansion to yield isochoric condition
- Matching sample thickness to range of laser deposition or conduction scale length yields isothermal condition

Forsman et al., PRB 58, R1248 (1998)

20nm Al heated with a 100fs, 400nm laser

Isochoric heating is scalable to X-rays, electrons, protons or ions
The first *ILH* experiment is the measurement of electrical conductivity of warm dense Au

- Isothermal heating produced by laser skin-depth deposition and ballistic electron transport
- Isochoric condition maintained by material strength & inertia

**WDM state characterized by** $\rho_0$ **and** $\Delta \varepsilon$

- $\Delta \varepsilon$ determined directly from $\{R, T\}$ of pump laser

**Probe** $\{R^*, T^*\}$ **yields single-state data on** $\sigma(\rho_0, \Delta \varepsilon)$
Measurements of S-pol \{R^*, T^*\} reveal an interesting temporal behavior

- Three distinct stages are observed
  - An initial transient
  - Quasi-steady state
  - Hydrodynamic expansion

\[ \Delta \varepsilon = (3.5 \pm 1.0) \times 10^6 \text{ J/kg} \]

Similar behavior seen with P-pol probe
Quasi-steady-state behavior is unexpected

- Hydrodynamic simulations suggest disassembly of the foil in \( \sim 1\) ps after heating when the lattice reaches melting temperature

  - Expansion gives rise to a plasma gradient on the surface of the foil; the gradient scale length will continue to increase with time

  - To maintain constant probe \( R^* \) and \( T^* \), it would require the dielectric properties of the non-uniform system to evolve in a manner that precisely mitigates gradient effects at all times

  This is improbable

The problem of hydro code is the lack of solid state effects
Quasi-steady-state behavior has important consequences

• It confirms the absence of significant hydrodynamic expansion, preserving the uniform, slab structure of the heated foil

• It yields an uniform state that is characterized by the direct observables of mass density $\rho_o$ and excitation energy density $\Delta\varepsilon$

This ensures realization of the Idealized Slab Plasma concept in isochoric heating of a solid by fs laser
The quasi-steady state validates single-state measurement of AC conductivity

- Probe \{R^*, T^*\} data for quasi-steady state used to solve Helmholtz eqs. for EM wave in a uniform dielectric slab
- This yields \( \sigma_\omega(\rho_o, \Delta\varepsilon) \) as direct benchmark for theory

Results obtained from 800nm, S-pol probe
We can learn more if we assume nearly free electron behavior

- Nearly free electron behavior is expected
  - Absence of interband transition at 800 nm
  - Conductivity effected by electrons near Fermi surface

- Drude model:
  \[
  \sigma(\omega) = \sigma_r + i\sigma_i = \frac{\sigma_o}{1 + \omega^2 \tau^2} (1 + i\omega\tau),
  \]
  \[
  \tau = \frac{\sigma_i}{\sigma_r} \frac{1}{\omega}, \quad \sigma_o = \sigma_r (1 + \omega^2 \tau^2), \quad n_e = \frac{m_e \sigma_o}{e^2 \tau}
  \]
This extends our single-state data to include $\tau$, $\sigma_0$ and $<Z>$.

At normal conditions:

$\sigma_0 = 4.1 \times 10^{17} \text{ s}^{-1}$

$n_e = 3.8 \times 10^{22} \text{ cm}^{-3}$

Widmann et al.,
PRL 92, 125002 (2004)

Drude behavior of $\sigma$ at 800nm is subsequently confirmed.
The data provided the first benchmark of Purgatorio in the WDM regime

S. Hansen, B. Isaacs, V. Sonnad, P. Sterne, B. Wilson

- **Purgatorio Code**
  - Neutral-pseudo atom model
  - Dirac equ. for bound wave functions
  - Phase shifts by matching numerical wave functions to analytical forms at ion sphere radius
  - Bound & continuum electron density from Fermi distribution
  - Inelastic crystal structure factor [Baiko et al., PRL 81, 5556 (1998)]
  - Electrical resistivity from extended Ziman formulation

- Agreement in $\sigma_0$ for $\Delta \varepsilon < 10^7$ J/kg
- Discrepancy in $\tau$, $n_e$
- Need for multi-parameter tests
What is the phase of the quasi-steady state?

- Calculations of equation of state and transport properties require phase information, solid versus liquid, to determine the structure factor of the state.

- The identity of the quasi-steady state is also key to understanding non-equilibrium phase transitions induced by ultrafast excitation.

The immediate questions are

- If the lifetime of quasi-steady state is governed by stability of the lattice, is the limit set by a critical value of lattice energy density and can it be determined?

- Does the quasi-steady state retain any long or short range order?
To determine lifetime of quasi-steady state, we probe hydro expansion with FDI.

**Diagram:**
- **Pump** (150fs, 400nm)
- **CCD**
- **PD**
- **Au 30nm**
- **Spectrometer**
- **Michelson Interferometer**
- **Probe** (150fs, 800nm)
- **R**
- **Δt**
- **T**

**Text:**

To determine lifetime of quasi-steady state, we probe hydro expansion with FDI.
Quasi-steady state is confirmed in six different measurements.

S-pol \{R^*, T^*\}

P-pol \{R^*, T^*\}

S/P-pol \Delta\phi

\[3.5 \times 10^6 \text{J/kg}\]

\[3.5 \times 10^6 \text{J/kg}\]

\[3.8 \times 10^6 \text{J/kg}\]

\[4.0 \times 10^6 \text{J/kg}\]
To quantify quasi-steady state duration, we use an extensive set of S-pol FDI data.
What are the processes governing solid-plasma transition in the heated foil?

- Laser heating of $s/p$ electrons and photo excitation of $d$-electrons
- Electron-hole recombination
- Electron-electron thermalization
- Escape of heated electrons forming a surface sheath; sheath thickness is limited by space charge field
- Lattice heating effected by electron-phonon coupling
- Melting of the lattice
  - Ultrafast, non-thermal melting?
  - Thermal melting to meta-stable superheated liquid?
  - Superheated solid?
- Disassembly of the superheated state into a plasma
To describe lattice heating, we use a modified Two-Temperature Model

TTM:

\[
C_e(T_e) \frac{dT_e(t)}{dt} = -g \left[ T_e(t) - \varepsilon_l(t) \frac{\rho_{Au}}{C_l} \right] + S(t)
\]

\[
\rho_{Au} \frac{d(\varepsilon_l(t))}{dt} = g \left[ T_e(t) - \varepsilon_l(t) \frac{\rho_{Au}}{C_l} \right], \quad \varepsilon_l(t) = \frac{C_l T_l(t)}{\rho_{Au}}
\]

Electron-phonon coupling: \( g = (2.2 \pm 0.3) \times 10^{16} \text{ W/m}^3\text{K}^* \)

Heat capacities: \( C_e(T_e) = \frac{\partial U_e(T_e)}{\partial T_e}, \quad C_l = 2.5 \times 10^6 \text{ J/m}^3\text{K}^\dagger \)

Laser energy deposition: \( S(t) = \frac{\Delta \varepsilon \rho_{Au}}{\tau_P \sqrt{\pi}} \exp \left( -\frac{t^2}{\tau_P^2} \right) \)

†Maxmillian's Chemical and Physical Data, Maxmillian Press, London, 1992
We postulate that disassembly is a rate-independent critical phenomenon.

- Quasi-steady-state duration $\Delta \tau$ is determined by a critical value $\varepsilon_D$ independent of heating rate (or $\Delta \varepsilon$)

$$\Delta \varepsilon = 4.2 \times 10^6 \text{ J/kg}$$
The heating-disassembly model shows good agreement with observation

- This yielded the first measurement of the critical lattice energy $\epsilon_D=(3.3\pm0.3)\times10^5$ J/kg for solid-plasma transition under ultraviolet laser excitation

\[ \Delta\Phi \text{ S-pol data} \]
\[ g=1.9\times10^{16} \text{ W/m.K, } \epsilon_D=3.0\times10^5 \text{ J/kg} \]
\[ g=2.5\times10^{16} \text{ W/m.K, } \epsilon_D=3.6\times10^5 \text{ J/kg} \]

To probe long/short range order in quasi-steady state, we use broadband dielectric function

- For Au, intra & inter-band transitions in 450-800nm of $\varepsilon(h\nu)$

- $\varepsilon(h\nu)$ determined from \{R*, T*\} of supercontinuum probe
180fs, 800nm laser is focused onto CaF$_2$ to generate a 450-800nm supercontinuum probe

Probe illuminates nanofoil at 45°-incidence in 30µmx600µm line focus, covering both heated and unheated regions

In-situ calibration eliminates the need for
- Absolute intensity calibration
- Measurement of shot-to-shot variation in probe intensity
Frequency chirp in supercontinuum is measured using Kerr optical gate

- Supercontinuum provides spectral measurements from 450-800 nm
- Frequency chirp gives rise to time-encoded spectrum
  - To remove effect of chirp in measurements
    - Bin spectral data in 10nm intervals
    - Apply temporal shifts using chirp data

![Graph showing frequency chirp in supercontinuum](image-url)
Temporal evolution of $\varepsilon(h\nu)$ of Au at $2.9 \times 10^6$ J/kg

Data corrected for frequency chirp

- Quasi-steady-state behavior seen in 1.2-4 ps consistent with earlier finding [Ao et al., PRL 2006]

- $\varepsilon_1(h\nu)$ relatively featureless

- $\varepsilon_2(h\nu)$ shows distinct components
  - Intraband transitions below 2.3 eV
    - Enhancement in transitions
    - Overshoot at 1.55 eV similar to previous observation
  - Interband transitions above 2.3 eV
    - Enhancement in transitions
Dependence of $\varepsilon(h\nu)$ of Au on excitation energy density $\Delta\varepsilon$

- **For** $\Delta\varepsilon$ of $2.6 \times 10^6$, $4.7 \times 10^6$ J/kg
  - The 1.4-2 ps probe delay falls within the quasi-steady-state

- **For** $\Delta\varepsilon$ of $1.7 \times 10^7$ J/kg
  - Disassembly occurs at 2.38 eV for a probe delay of 1.9 ps, consistent with previous data

- Intra band transitions
  - Enhancement with $\Delta\varepsilon$
  - Drude behavior

- Inter band transitions
  - Enhancement with $\Delta\varepsilon$
  - Increasing red shift with $\Delta\varepsilon$
Drude behavior in intra band transitions points to discrepancy in $\sigma(h\nu)$ calculation

- Spectral structures in $\sigma(\omega)$ above 1.3 eV were reported [Mazevet et al., PRL 2005]
  - Sampling of Brillouin Zones over only $\sim 8^3$ $k$-points

- Limited BZ sampling can lead to spurious spectral structures [T. Ogitsu & E. Schwegler]
  - fcc Au at 0 K
  - Convergence is reached with $128^3$ $k$-points

$\varepsilon_2$ data in disagreement with calculations lacking treatment of non-adiabatic effects of electron-phonon coupling
The prominence of inter band transitions raises many interesting questions

- Persistence of $d$-band in the quasi-steady state
  - If $d$-band is the result of long range order, this would be first evidence of the quasi-steady state being a superheated solid

- Red shift can be due to temperature-induced changes in the energy distribution of the electrons

- Enhancement is likely a non-equilibrium effect
  - Equilibrium calculations for Al shows disappearance of interband transitions at melting (Benedict et al., PRB 2005)
  - Photoemission spectroscopy on fs-laser excited Au at 300$\mu$J/cm$^2$ shows residual non-thermal electron distribution after 670fs (Fann et al., PRB 1992)

Summary

- The *Idealized Slab Plasma* Concept has been realized in *Isochoric Laser Heating*.

- This has become a unique platform for the study of non-equilibrium, high-energy-density Warm Dense Matter free from gradient effects:
  - Electrical conductivity
  - Lattice energy density for solid-plasma transition
  - Persistence of band structure in quasi-steady state with non-equilibrium electron DOS

Warm Dense Matter an emerging frontier in plasma & CM science
- 2002 LLNL Workshop on Extreme States of Material: WDM to NIF
- 2002 US-France Workshop on WDM
- 2003 CECAM Workshop on QMD Approaches of WDM
- 2006 Accelerator-Driven WDM Workshop
- 2006 Lansce Dynamic Experiment Facility Workshop