Study of Warm Dense Matter Physics with Ultra-Short Pulse Lasers

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Outline

Ellipsometric pump-probe measurements of WDM

+ Diffuse scattering of probe pulse
+ White-light femtosecond probe beam
  for frequency dependence of AC conductivity
+ Data reconstruction technique to identify surface expansion
  and detect sharp interface inside plasma
+ New EOS calculation and proposal to obtain critical point of metal

*Electron localization in WDM (low conductivity, metal-insulator transition)
*Positive-negative ion plasma in WDM
*Two phase fluid region (gas & liquid)( droplet formation, EOS in 2 phase, .. )
*....
Purpose of this study

Warm dense matter:
Material properties between solids(liquid) and plasmas

* Chemical force (condensed matter) ~ Coulomb force (ideal plasma)

* Electron degenerated plasma (Giant planet interior material)

* Strongly coupled plasma

* Metal-insulator transition (minimum conductivity, similar to Anderson transition?)

* Two phase region [gas and liquid] (droplet or debris formation)

There are a lot of new physics and many uncertain phenomena.
We need heating faster than expansion, and measurements with high resolution.
Ellipsometric pump-probe measurements in WDM

- X-Y plot (Rp/Rs vs. F(Rp, Rs, δp-s))
- Reconstruction method with Fresnel’s law
- Diffuse scattering measurements
Measurements of ellipsometric parameter and diffuse scattering

\[ I_1 \approx |R_s|^2 \]
\[ I_2 \approx |R_p|^2 \]
\[ I_3 \approx \frac{1}{2} \left( |R_p|^2 + |R_s|^2 - 2|R_p||R_s|\sin(\delta) \right) \]
\[ I_4 \approx \frac{1}{2} \left( |R_p|^2 + |R_s|^2 + 2|R_p||R_s|\sin(\delta) \right) \]
What optical electromagnetic waves see

Reflection at sharp boundary
Impedance miss-matching

\[ r = \frac{(n + ik) - 1}{(n + ik) + 1} = \frac{\sqrt{\varepsilon_1 + i\varepsilon_2} - 1}{\sqrt{\varepsilon_1 + i\varepsilon_2} + 1} \]

Propagation in the expanded matter
n(x), k(x)

AC conductivity
electron mobility + atomic polarizability
3 Stokes’ Parameters or
Rp, Rs, δp-s

Droplet formation when? how?
Critical point, two fluid region
Conductivity of fluid with droplet
To check starting time when expansion component cannot be neglected.

### Measured Parameters

Stokes’ parameters

- $s_1(t)$, $s_2(t)$, $s_3(t)$,  
- $s_0$

$F=3$

### Plasma Parameters

- Single interface: $F=2$
- Expanded plasma: $F>3$

\[
\rho = \tan(\varphi) \exp(i\Delta)
\]

- Fresnel’s law available
- No

#### Graphs

- Au shot 021114-4-1 Rp signal
- Onset time of expansion [fs]

#### Axes

- Pumping intensity [W/cm²]
- Time (fs)
- Reflectivity, measured reflectivity
- Cold parameter $t_0$
- Onset time $t=t_s$ from Fresnel’s law
Change of polarization state of probe beam with target heating

\[ Y = \frac{I_3 - I_4}{I_3 + I_4} = \frac{2|R_s||R_p|\sin(\delta)}{|R_s|^2 + |R_p|^2} \]

Very sensitive to optical constant and thickness of plasma

X = \frac{I_2}{I_1} = \frac{|R_p|^2}{|R_s|^2}
Strong reduction of AC conductivity in Gold
We have measured Au, Cu, Al, W, Mo, Sn, Fe, SS304, SiO₂.

For $\lambda=745\text{nm}$
Experimental results (Gold targets)

Higher intensity
Higher peak temperature
AC conductivity in Au

\[ \varepsilon = 1 + \left[ \varepsilon_{r}^{\text{atom}} + i \varepsilon_{i}^{\text{atom}} \right] + \left[ -\frac{\omega_p^2}{\omega^2} \frac{\left(\omega \tau\right)^2}{1 + \left(\omega \tau\right)^2} + i \frac{\omega_p^2}{\omega^2} \frac{\omega \tau}{1 + \left(\omega \tau\right)^2} \right] \]

experiments

simulations

Full Drude

0.1 Drude

0.01 Drude
Multi frequency probe of warm dense Au plasma

expansion $\Rightarrow r_p$ decreases, $\Rightarrow$ both of $X, Y$ decrease
Y increase at 850nm $\Rightarrow$ change of $\delta$ should be large

For 745nm

$Y = \frac{I_3 - I_4}{I_3 + I_4}$

$X = \frac{R_p}{R_s}$

$\Gamma_{\text{pump}} > 10^{14}$W/cm$^2$
Reconstruction method shows surface expansion

Probe laser of longer wavelength looks at deeper region?
Different frequency probe observes different plasma?

Each probe frequency may see different region.

Gold rarefaction with recombination (t=1ps)

atom density for Au solid = \(5.9 \times 10^{22} \text{cm}^{-3}\)

Here long wavelength penetrates more deeply,
Contrary to usual plasma behavior.
Electron localization in WDM
M-NM transition(?) may be faster process in USP plasma.

\[ n^2 - k^2 \] (real part of \( \varepsilon \))

- **W**
- **Au**
- **Cu**

Gold rarefaction with recombination (t=1ps)

850nm
745nm
780nm

maybe insulator
maybe metal

Two phase region
Droplet formation
Time resolved diffuse scattering by Au targets

lower pump intensity

higher pump intensity
Turn-on time of W scattering is shorter than that of Au.

onset time of diffuse scattering

Au  0.8–2.6 ps
W   0.75–1 ps
(with 0.3 ps heating duration)

$I=10^{13}–10^{14} \text{W/cm}^2$
Summary

1. Strong reduction of AC conductivity is observed in ellipsometric measurements.
2. Frequency dependence also denotes small contribution of free-electron.
3. We observed some evidence of Au$^+$-Au$^-$ plasmas.
4. These results give us the evidence of localization of electron in WDM.
5. Diffused scattering signal due to reach the two phase boundary can be detected.
6. Droplet formation looks very fast.
7. New EOS for Tin. We propose new measured method to detect critical point for metals.