Optical Constants of Ultra-Short-Pulse Laser Heated Metal

YONEDA Hitoki, MORIKAMI Hidetoshi, UEDA Ken-ichi and MORE Richard M.¹⁾

Institute for Laser Science, University of Electro-communications, Chofu, Tokyo 182-8585, Japan

¹⁾ *National Institute for Fusion Science, Toki, Gifu 509-5292, Japan*

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The complex refractive index ($n + ik$) of ultra-short-pulse laser heated gold metal was measured with a new ellipsometric pump-probe technique. Two ratios of four different probe-beam polarizations were used to determine s- and p-reflectivity and their phase difference. In the early stage of heating, only the imaginary part of the dielectric constant $\epsilon(\text{Im}[\epsilon] = 2nk)$ increased while real part of ϵ ($\text{Re}[\epsilon] = n^2 - k^2$) was almost constant. This agrees with the Drude model. We observe a pause in the rise of $\text{Im}[\epsilon]$ at the boiling temperature. Beyond this point, $\text{Re}[\epsilon]$ started to change and the observed parameters disagree with the Drude theory. It was found that the parameters of the expanding plasma follow a unique trajectory in n, k space, even though time variation of $\text{Re}[\epsilon]$ and $\text{Im}[\epsilon]$ are different for various pump intensities. This means the gold remains in a unique state such as the neutral groundstate in this intensity range.

Keywords:

ultra-short-pulse laser, high density plasma, complex refractive index, drude model

Pump-probe reflectometry with ultra-short-pulse laser is a powerful tool for observation of laser heated plasmas. Though at least two parameters have to be measured for obtaining complex optical constant of absorbing materials, only reflectivity was measured in many previous experiments [1]. This situation required some models for understanding material properties of heated materials. To improve this, Blanc [2] proposed detection of phase change with a frequency domain interferometer. This method had enough accuracy and potential for space- and time-resolved measurements but there was complex post processing of measured data. Recently, we proposed a different way to obtain s- and p-polarized reflectivity (r_s, r_p) and the phase difference (δ) with a single-shot technique [3]. Figure 1 shows a schematic drawing of the experimental setup. The probe beam (120 fs, 745 nm) illuminated a $5 \mu\text{m}\phi$ area in center of $10 \mu\text{m}\phi$ spot of the pump beam (300 fs, 248 nm). We measured four probe-beam intensities having different Stokes parameters. The observed optical system is similar to a division-of-wavefront photopolarimeter [4]. Intensity of pump beam was

varied in range of $4 \times 10^{12} \sim 2 \times 10^{13} \text{ W/cm}^2$ while probe beam intensity was about 10^{10} W/cm^2 and there was no observable nonlinear effect. The value of r_s, r_p ,

Fig. 1 Schematic drawing of experimental setup of ellipsometric pump-probe measurements.

author's e-mail: yoneda@ils.uec.ac.jp

and δ were calculated from $I_1 \sim I_4$ signals with following relations.

$$\frac{I_2}{I_1} = \frac{|r_p|^2}{|r_s|^2}, \quad \frac{I_3 - I_4}{I_3 + I_4} = \frac{2|r_s||r_p|\sin(\delta)}{|r_s|^2 + |r_p|^2}.$$

At early time of heating, expansion plasma in front of solid surface could be neglected. In this case, the complex refractive index ($n + ik$) was calculated with Fresnel's law. The complex dielectric constant ($\epsilon = \epsilon_1 + i\epsilon_2$) was also calculated with the basic equations $\epsilon_1 = n^2 - k^2$ and $\epsilon_2 = 2nk$. Both the solid metal surface and expanded plasma must be considered at late times. In the following discussion, we use the data obtained with assumption of a single interface. The limitations of this assumption will be clarified.

Figure 2 shows temporal waveforms of imaginary part of ϵ ($\text{Im}[\epsilon] = 2nk$) and real part of ϵ ($\text{Re}[\epsilon] = n^2 - k^2$). We plotted those for five different pump intensities from 4×10^{12} to 2×10^{13} W/cm². We observed flat points in $\text{Im}[\epsilon]$. According to the Drude model, the dielectric constant was expressed with following equations.

$$\text{Re}[\epsilon] = n^2 - k^2 = 1 - \left(\frac{\omega_{pe}}{\omega_0} \right)^2 \frac{(\omega_0 \tau)^2}{(\omega_0 \tau)^2 + 1},$$

$$\text{Im}[\epsilon] = 2nk = \left(\frac{\omega_{pe}}{\omega_0} \right)^2 \frac{(\omega_0 \tau)^2}{(\omega_0 \tau)^2 + 1},$$

where ω_{pe} , ω_0 , and τ are plasma frequency, laser frequency, and damping time. The value of τ should linearly decrease with increase of temperature. Since the growth of $\text{Im}[\epsilon]$ between room temperature and the flat point was about 8, this point is probably the boiling temperature of gold. The pause duration due to boiling was also estimated with latent heat of evaporation (3.5 eV / atm) and heating rate due to pump laser (8 ~ 40 eV / atom/300 fs). The calculated pause duration were 130 ~ 26 fs for $I = 0.4 \sim 2 \times 10^{13}$ W/cm². Therefore, in the highest intensity case, we couldn't see the flat point, while in the lower intensity case, temporal resolution was enough high to observe it.

After boiling, $\text{Re}[\epsilon]$ also changed. While $\text{Im}[\epsilon]$ increased monotonically during heating, slope of $\text{Re}[\epsilon]$ changes from negative (for lower intensity case) to positive (higher intensity case).

In Fig.3, we indicate data on X-Y plot in space of n and k . In this figure, we plot iso-contour lines of measured two pairs of signals (I_1/I_2 , $(I_3 - I_4)/(I_3 + I_4)$). It is important that these contour lines cross orthogonally

for minimizing determination error from measured ratios to n and k values. Even though temporal waveforms of each intensity are different, the trajectories at any

Fig. 2 Typical waveform of real and imaginary part of dielectric constant. Pump laser Intensity was varied from 4×10^{12} to 2×10^{13} W/cm². Arrows denoted intensity increasing direction.

Fig. 3 X-Y plot (n and k space) of trajectories of transient state from solid to plasma. Iso contour lines for ratios of measured signals were also shown in this figure.

intensity pass along an almost unique route. The reason for this is probably that the observed gold properties reflect a constant material condition like a neutral gas. If we considered small fraction of free electrons, their effect is so large that trajectories change with small difference in electron density. (The expanded plasma density was still higher than the critical density.) Since the laser provides large enough absorbed energy on atoms for ionization, this uniqueness is surprising. However, there is strong cooling due to adiabatic expansion and fast recombination occurs.

Some trajectories pass through a line of $n = k$ at higher intensity case. The $\text{Re}[\varepsilon]$ ($= n^2 - k^2$) changed from negative to positive. This change is one point of evidence for a transition from metal to insulator. In addition, the estimated τ became negative after passing through this line. Therefore, the Drude model is entirely

invalid. More detailed analysis based on solution of the Maxwell wave equations with expanding plasma gives us the parameters of such high density plasma. This analysis will be presented elsewhere.

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