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Detection of hydrodynamic expansion in ultrashort pulse laser ellipsometric pump-probe experiments

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In ultrashort-pulse laser interaction with solid target materials, the target rapidly heats, melts, evaporates, and begins to expand as a vapor or plasma. The onset of hydrodynamic expansion following surface evaporation is a switching point, where the dominant physics changes from temperature dependence of the solid dielectric function to refraction by the dense vapor cloud. We propose and demonstrate a method to analyze reflection data to identify this onset of target expansion. We use two of the Stokes parameters obtained from ellipsometric pump-probe measurements to determine a dielectric function with an assumption of no expansion. We use this dielectric function to predict the full set of reflectivity measurements. If there is a sharply defined target interface, this method reproduces the experimental data. When the plasma expansion is no longer negligible, the prediction deviates from the experimental measurements. This comparison shows when the plasma expansion is no longer negligible.

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Today, the analysis of the physical properties of warm dense matter is a frontier research area. Pump-probe reflection experiments with ultrashort-pulse (USP) lasers are very convenient for this research because they permit the controlled creation of well-defined high-density plasmas. Price *et al.* measured the absorption of USP laser light by several materials [1]. In the warm dense plasma region they found a wide variety of absorption coefficients for different materials, while the hot plasma absorption was similar for all materials. Grimes *et al.* measured the femtosecond time-resolved reflectivity of heated material by using pump-probe experiments with USP lasers, which could determine the complex dielectric constant of Fe and C [2]. The hydrodynamics of a laser-heated target makes it more difficult to uniquely determine plasma properties from the observed data. Although some previous experiments found evidence for plasmas with a scale length shorter than the laser wavelength [3,4], the analysis required an assumption for the plasma dielectric function. At present there is no general theory for the electrical properties of such warm dense plasmas, although the Drude theory is often used. Recently, we developed a technique to measure the complete set of Stokes parameters with an ellipsometric pump-probe method. Combining a computer simulation with the solution of the Maxwell equations, we found the reflection by warm dense gold is dominated by the atomic polarizability [5]. The inferred dielectric constant was quite different from the Drude model. Theoretical quantum molecular dynamical simulations also found cases that disagreed with the Drude theory [6]. Because of the great fundamental interest of localization or non-Drude conductivity phenomena, a more detailed measurement and analysis is now in order.

In principle, Fourier interferometry can be used to measure the expansion length of plasmas [2]. Even with these difficult measurements, the expansion is not precisely determined without accurate information about the dielectric function of the expanded plasma. However, at early times after short-pulse heating, the target expansion is negligible, and the normal Fresnel law can be used to determine the complex dielectric function of the heated solid target directly from the experimental data. After sufficient heating, the target surface expands, and a low-density, high-gradient plasma is created. From that time, the reflection of the probe laser light depends on both the surface plasma and the hot liquid target material. A difficulty in the interpretation of these experiments is clear from the simulations: the reflected or transmitted probe light interacts with both the expansion plasma and the high-density plasma, and the observed parameters are "integrated" along this path.

It is important to know when the effect of plasma expansion starts to be dominant. To determine this time we can use a comparison between the measured reflectivity and a reflectivity estimated with an assumption of a single interface. In our ellipsometric experiments we measure four reflected waves, which have different Stokes parameters. The details of the experimental conditions were explained in a previous paper [5]. In brief, we measured $I_1 \propto |r_s|^2$, $I_2 \propto |r_p|^2$, $I_3 \propto \frac{1}{2}(|r_s|^2+|r_p|^2-2|r_s||r_p|\sin\delta)$, and $I_3 \propto \frac{1}{2}(|r_s|^2+|r_p|^2+2|r_s||r_p|\sin\delta)$, respectively. In these formulas, r_p and r_s are the amplitude of the *s*- and *p*-polarized probe beam components, δ is the phase difference between the *s*- and *p*-polarized components, and I_1, I_2, I_3, and I_4 are the measured signals. This means we can determine a complete set of Stokes parameters, three independent parameters that characterize the reflected probe light. However, under the assumption of a single sharply defined interface, two Stokes parameters are sufficient to determine the complex dielectric function. Therefore, we can check the assumption of a sharp interface by calculating the remaining Stokes parameter. Strictly speaking, in this analysis we also assume that thermal conduction is fast enough to create a uniform temperature in the skin depth area of the probe laser. Normally, this is true because the thermal conduction is much faster than the thermal expansion of target surface.

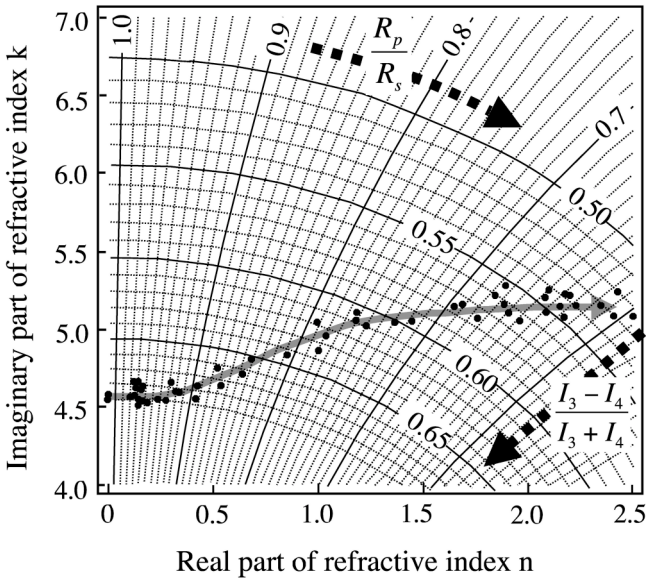

FIG. 1. Typical trajectory of the *X*-*Y* plot for gold using ellipsometry. The vertical axis is the real part of the complex refractive index and the horizontal axis is the imaginary part of the complex refractive index. The curves in this figure correspond to constant *X* and *Y*.

The actual procedure is the following. We form ratios of parameters $X=I_2/I_1=|r_p|^2/|r_s|^2$ and $Y=(I_3-I_4)/(I_3+I_4)=[2|r_p||r_s|\sin(\delta)]/(|r_p|^2+|r_s|^2)$ from the four measured reflectivity signals. These ratios are used because they have less sensitivity to fluctuations of the probe beam intensity. The relations between *X* and *Y* and optical refractive index *n* and *k* are shown in Fig. 1. These variables are often used in ellipsometry measurements [7]. The almost orthogonal set of curves for *X* and *Y* means that *n* and *k* are well determined by measurements of *X* and *Y*. For gold targets, we draw a typical trajectory of the apparent optical refractive index for the gold targets. In the case of gold, there is a unique trajectory in Fig. 1, for a range of pump laser intensities ($I=4\times10^{12}-2\times10^{13}$ W/cm^2). For higher intensity illumination or for other materials, the trajectories depend on intensity.

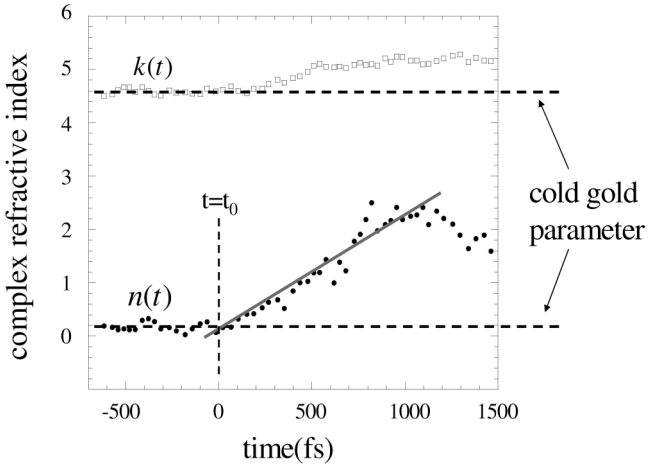

FIG. 2. Typical example of time-resolved optical refractive index $n(t)$ and $k(t)$ for gold.

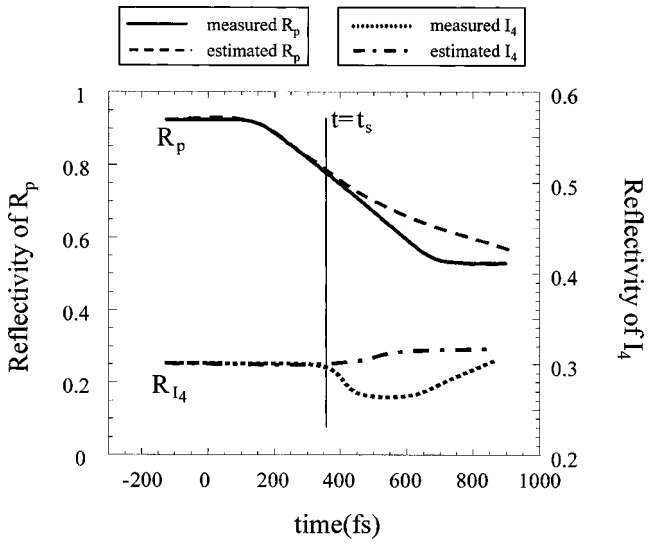

FIG. 3. Comparison of reflectivity changes of *p*-polarized (R_p) and circular polarized (R_{14}) probe beam. In the case of low pump intensity (about 4×10^{12} W/cm^2), the measured and reconstructed curves agree until about 380 fs. Each reflectivity changes at $t=t_s$. In the region of $t>t_s$, we cannot neglect the expansion component at the target surface.

Even with such a situation, we can decide the apparent optical refractive index.

In our experiments, the probe beam has a wavelength of 745 nm and pulse duration of $\tau=120$ fs. The angle of incidence is 64° measured from the normal to the target surface. The pump duration is 300 fs and wavelength 248 nm, respectively. The other parameters of this measured system are described in Ref. [5]. The center time of the probe beam can be changed with a time step of 13 fs.

Typical histories of the estimated $n(t)$ and $k(t)$ are shown in Fig. 2. Apparently, before the arrival of the pump beam, the probe beam looks at the cold gold target surface so that the $n(t)$ and $k(t)$ values agree with the reference data [8].

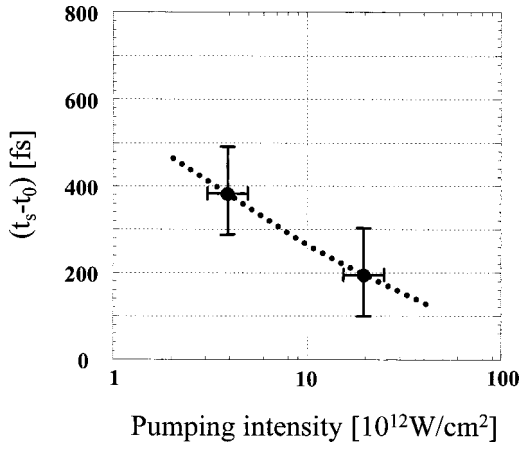

FIG. 4. Pumping intensity dependence of the single interface region (t_s-t_0). In our measurement the time resolution is about 100 fs. However, the onset of hydrodynamic motion, inferred from the disagreement of measured and reconstructed reflectivities, changes by 180 fs.

After the pump beam arrives, n and k both change. In Fig. 2 the real part of the optical index $n(t)$ has more change than the imaginary index $k(t)$. We decide on the time zero for the horizontal axis by the start time of the changes of $n(t)$. Although there was no direct measurement of the temperature, the plasma is thought to be heated to a few tens of eV after the end of the pump beam. This temperature was estimated from the absorbed laser energy, the specific heat of gold, and typical thermal conduction depth. At late time the probe beam interacts with an expanding plasma so that these $n(t)$ and $k(t)$ at later times are artificial values.

From these estimated $n(t)$ and $k(t)$, we calculate the expected reflectivity signals for four detector channels. Each channel has different Stokes parameters. Figure 3 shows the comparison of the measured and reconstructed reflectivities for p-polarized reflectivity (channel I_2) and also for the left-circular polarized beam. The calculation agrees with the experiment for early times during the heating. However, the two curves begin to disagree at time $t_s=380$ fs. This separation time is the same for all four channels.

This behavior is reasonable because we can get three independent Stokes parameters from the four signals. We use two degrees of freedom for the calculation of the apparent $n(t)$ and $k(t)$ and so only one degree of freedom remains. If there is a discrepancy, it means the target violates the assumption of a single sharply defined interface. In practice this means we cannot neglect the target expansion after this time ($t=t_s$). Before t_s, the estimated numbers of Fig. 3 correctly give the optical properties of the solid-density plasma. For obtaining the plasma scale length of plasma, strictly speaking, we would need to know the plasma profile and dielectric constant of the expanding material. With an assumption of adiabatic expansion and simple equilibrium condition for atom and ion abundance, we can estimate that the scale length is about 3 nm when $t_s=300$ fs.

Figure 4 shows the intensity dependence of the waiting time for significant hydrodynamic motion. This waiting time decreases with the increase of pumping intensity. This temperature dependence is easy to understand because the temperature of the target surface increases with pumping intensity and the expansion velocity should also be also be higher at higher temperature.

In summary we have proposed a method of determining the starting time when the hydrodynamic expansion cannot be neglected for the interaction of an USP laser and a solid target. In this method only experimental data is used without any kind of expansion model or theory of the dielectric response. In the case of our gold targets, even at relatively low intensity and for a heavy target material, the effect of plasma expansion cannot be neglected at a few hundred femtoseconds after the heating pulse.

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