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Real-time ultrafast dynamics of dense, hot matter measured by pump-probe Doppler spectrometry

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Abstract. A detailed understanding of the critical surface motion of high intensity laser produced plasma is very crucial for understanding the interaction. We employ the two colour pump-probe technique to report the first ever femtosecond scale ultrafast dynamics measurement of the critical surface of a solid plasma produced by a relativistically intense, femtosecond pump laser beam (10¹⁸ W/cm², 30 fs, 800 nm) on an aluminium target. We observe the Doppler shift of a time delayed probe laser beam (10¹² W/cm², 80 fs, 400 nm) up to delays of 30 ps. Such unravelling of dynamics has not been possible in earlier measurements, which typically used the self reflection of a powerful pump pulse. We observe time dependent red and blue shifts and measure their magnitudes to infer plasma expansion velocity and acceleration and thereby the plasma profile. Our results are very well reproduced by 1D hydrodynamic simulation (HYADES code).

1. Introduction
Intense, femtosecond laser pulses explosively ionize solids and produce some of the highest energy densities achievable in the laboratory [1]. The resulting plasma not only offers great opportunities for studying extreme states of matter, but can also provide a source for high brilliance, high energy, tuneable, ultrashort sources of x-rays, electrons, ions and positrons [1]. The physics of such plasmas has deep implications for the fast ignition scheme of laser fusion [2]. At ultrahigh intensities the physics is challenging, given the variety of absorption mechanisms that can operate, the relativistic nature of the particles and the complex nature of their transport [1]. The instantaneous profile of the plasma density is a crucial parameter that controls much of the light coupling physics [3], generation of plasma instabilities [4] etc. The light coupling in steep and high density plasmas occurs essentially at the critical surface where for example, strong plasma oscillations can be excited (resonance absorption) [5]. In the four decades of investigation of laser produced plasmas, the dynamics of coupling of long (nanosecond) pulses have been investigated very thoroughly in experiments, analytical theory and simulations [6], but the actual motion of the plasma near to the critical surface was integrated out during the long pulse durations. In the ultrashort regime, there have been far fewer studies of the dynamics at extremely high intensities, though there are studies at moderate intensities.

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Liu and Umstadter [8] reported that the ponderomotive pressure at even as low an intensity as 10^{15} \text{ W/cm}^2 is enough for a 400 fs laser pulse to steepen the electron density profile. Kalashnikov et al. [9], reported red and blue shifts as a function of the laser pulse shape for a 2 ps pulse at 10^{17} \text{ W/cm}^2, measuring the self reflection of the incident pulse. Sauerbrey et al. [10] reported detailed and interesting measurements of reflectivity, x-ray generation and spectral shifts at 10^{17} \text{ W/cm}^2 for 350 fs laser pulses. They reported red and blue shifting and dependence of the spectral shifts on intensity and chirp of the input laser pulse. Sauerbrey [11] also presented a detailed model of the laser–plasma interaction and demonstrated the conditions of the laser pulse and target parameters that could produce either red-shift or blue-shift. In many of these studies [7-13], the effects that were considered were essentially ‘instantaneous’ as the measurements relied on reflection of the irradiating pulse itself.

Here we create plasma on a metal target at 10^{18} \text{ W/cm}^2 with an 800 nm, 30 fs laser pulse and interrogate the plasma at different later ‘instants’ with a 400 nm, 80 fs laser pulse, tracking the plasma all the way up to 30 ps. We present clear evidence for the initial inward motion of the critical surface which bounces back at later time. We are able to derive ‘instantaneous’ velocity and acceleration profiles of the initially kicked plasma. Our results are well reproduced by 1D hydrodynamic simulation (HYADES code) [14]. We believe our results are important for studies that seek to understand energy and mass transport in the plasma at relativistic intensities.

2. Experimental

![Diagram](image)

**Figure 1.** (a) Schematic of the experimental setup: Avantes (SP1) and Ocean Optics HR-2000 (SP2) UV-visible spectrometers; (b) Normalized reflected spectra of the probe beam from the plasma surface at several time delays.

We use the two colour pump-probe technique for measuring the dynamics of the critical layer. (figure 1(a)). The pulses are derived from a 20 TW, 30 fs Ti-Sapphire chirped pulse amplification laser. A p-polarized pump pulse at 800 nm (having 37 nm band-width), pulse at repetition rate of 10 Hz, is incident at 40° to the target normal. Its nanosecond prepulse contrast is 3 \times 10^{-6}. A small portion of the pump pulse is extracted and frequency doubled in a 1 mm thick BBO crystal to generate a probe pulse at 400 nm (having 3 nm band-width), which has duration of 80 fs. The probe pulse is delayed; with a resolution of 1 \mu m using a PLX retroreflector mounted on a linear translation stage and is passed through a BG-39 filter to remove residual 800 nm light. The pump pulse is focused with an f/4 gold coated off axis parabolic mirror to a diameter of 20 \mu m on optically polished aluminium target. The probe is focused to 60 \mu m with an f/30 lens so as to be collimated on the plasma created by the pump. Typical pump intensities generated are about 5 \times 10^{18} \text{ W/cm}^2, while the probe is at \sim 10^{15} \text{ W/cm}^2. The pump-probe simultaneity at the target (i.e. zero time delay) is obtained by monitoring the reflectivity of the probe beam as a function of the delay; at zero time, there is a sharp transition in reflectivity from a high value to a lower value. The probe pulse reflected from the target is divided by a beam splitter and fed to two different spectrometers (SP1 and SP2) (figure 1). An ‘Avantes’ UV-visible spectrometer (SP1) operates in the range of 245 to 505 nm having spectral resolution of 1.3 Å, while
another (SP2, ‘Ocean Optics HR-2000’) works in the range of 350 to 445 nm, having a spectral resolution of 0.5 Å. Here we present only the spectra measured by SP2 because of its higher resolution. 50 spectra are collected and averaged at each time delay. The narrow spectrum of probe pulse facilitates the observation of small wavelength changes, as evident in our data.

3. Results and Discussion

Figure 2. Doppler shift in reflected probe spectra (a) experimental and (b) 1D HYADES simulations. Calculated instantaneous (c) expansion velocity and acceleration from Doppler shift.

Figure 1(b) illustrates the normalized reflected spectra of the probe beam from the plasma surface. The black curve is the probe spectrum at zero time delay. The figure 2(a) shows that for aluminium, the reflected probe pulse is red-shifted at early times (up to 19 ps) while the shift reverses and becomes increasingly blue for larger delays. The normally incident 400 nm probe pulse reflects from its critical surface located at electron density of \(6.3 \times 10^{21}\) cm\(^{-3}\). Our data indicate that critical surface initially recedes from the probe, pushing into target. At later times, this surface moves back towards vacuum, getting progressively higher in speed. Our data clearly indicate a ‘turn around’ time of around 19 ps.

The temporal resolution in our experiment enables a deduction of ‘instantaneous’ velocity and acceleration of the critical surface, shown in figure 2(c). The shifts are consistent with velocities of the order of \(10^7\) cm/s expected from simple analytical estimates based on \(0.5c \left(\frac{\Delta \lambda}{\lambda}\right)\) (cos \(\theta\)) \[8\], while the acceleration is found to be of the order of \(10^{18}\) cm/sec\(^2\) \[11\]. The velocities are similar to those found by Kalashnikov et al. \[9\] which were derived from the ‘instantaneous’ second harmonic reflection from the plasma.

The modelling of the target hydrodynamics under the influence of the laser was performed using the 1D radiation hydrodynamics code HYADES \[14\]. HYADES is a 1D Lagrangian radiation hydrodynamics simulation code with a flux limited diffusion model for electron conduction and a multi-group diffusion model of thermal radiation flow within the target. The results of HYADES modelling as pertains to the interaction of the main pulse with the target cannot be expected to accurately reflect the interaction since a PIC or PIC-Hybrid code would be necessary for this purpose. It should be borne in mind, therefore, that the location and quantity of energy deposited during the main pulse will not be accurately represented by HYADES. The prepulse interaction should, however, be well represented by the models incorporated in the code.

In the modelling, a 500 µm thick target is divided into 100 Lagrangian cells, which are feathered toward the driver facing surface. Two different laser sources are incorporated, one at 800 nm which represents the pump laser, and the other the probe at 400 nm. The probe is modelled so as to determine whether the interaction of the probe with the plasma is affecting the result of the experiment. A flux limiter of 0.06 is employed, which is appropriate for the prepulse interaction of the pump beam. The calculations described in the previous subsection were analyzed by estimating the leading order wavelength shift that the moving critical surface would induce, i.e. \(\Delta \lambda = (2 \chi_{\text{crit}} \lambda_{\text{probe}}) / c\), where, the velocity of the critical surface was approximated to the fluid velocity at the critical surface. Post-processing the above calculation yields the wavelength shift as a function of probe time delay, and this is shown in figure 2(b). Although there is not exact quantitative agreement, the qualitative behaviour of the wavelength shift with probe delay is reproduced and the absolute wavelength shifts are within a
factor of 2 of the experimental measurements (figure 2(a)). This discrepancy is likely explained by the fact that HYADES poorly represent the energy deposition of the main pulse.

By examining the simulation output in light of figure 2(b), the experimental measurements can now by physically interpreted as follows. The pump laser launches a compression wave into the front surface plasma which has been formed by the laser prepulse. At early times the position of the critical surface for the shorter wavelength probe beam tracks this compression wave as it moves into the target. Thus at early times a red shift is measured. At later times the compression wave has propagated into a region of overdense plasma, and so the critical surface for the probe now sits in a region that is undergoing rarefaction and thus the critical surface is now moving into the vacuum and towards the laser. Therefore a blue shift is measured.

From the analysis of the simulation results the qualitative behaviour of the spectral shift can be understood. The simulations are within a factor of two of an accurate quantitative description with an experiment. This means that the experimental approach used in here can provide detailed insights into the plasma dynamics in the crucial energy deposition region, with sufficient time resolution that it can be readily compared to a variety of different simulation types. This can consequently allow for a better theoretical understanding of the interaction of the main pulse with the solid target.

4. Conclusions

We report the first ever pump-probe dynamics of the critical surface of solid density plasma produced by high intensity, femtosecond laser. Time-resolved ultrafast Doppler spectrometry is a powerful technique to infer fast dynamics in dense plasma systems. The variation in the peak shift and modified spectral-width of the probe beam is observed as a function of probe delay. At intensities we use in the experiment here, the plasma motion is mostly driven by hydrodynamics and hence occurs in picoseconds timescales. Ponderomotive pressure becomes more significant at higher intensities, and critical surface oscillations can be as fast as the speed of light. The oscillating surfaces can up-shift the driving pulse to high harmonic attosecond pulses. Doppler spectrometry with few-cycle pulses can provide invaluable information regarding these otherwise obscure ultrafast dynamics.

5. References

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