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Mondal, S., Lad, Amit D., Ahmed, Saima et al. (5 more authors) (2010) Doppler Spectrometry for Ultrafast Temporal Mapping of Density Dynamics in Laser-Induced Plasmas. Physical Review Letters. 105002. pp. 1-4. ISSN 1079-7114

https://doi.org/10.1103/PhysRevLett.105.105002

Doppler Spectrometry for Ultrafast Temporal Mapping of Density Dynamics in Laser-Induced Plasmas

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(Received 19 October 2009; revised manuscript received 25 May 2010; published 30 August 2010)

We present high resolution measurements of the ultrafast temporal dynamics of the critical surface in moderately overdense, hot plasma by using two-color, pump-probe Doppler spectrometry. Our measurements clearly capture the initial inward motion of the plasma inside the critical surface of the pump laser which is followed by outward expansion. The measured instantaneous velocity and acceleration profiles are very well reproduced by a hybrid simulation that uses a 1D electromagnetic particle-in-cell simulation for the initial evolution and a hydrodynamics simulation for the later times. The combination of high temporal resolution and dynamic range in our measurements clearly provides quantitative unraveling of the dynamics in this important region, enabling this as a powerful technique to obtain ultrafast snapshots of plasma density and temperature profiles for providing benchmarks for simulations.

DOI: 10.1103/PhysRevLett.105.105002 PACS numbers: 52.38.−r, 52.50.Jm, 52.65.−y

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 also serves as a tunable high energy, ultrashort pulse, and high brilliance source of x rays, electrons, ions, and positrons [1]. Understanding the physics of such plasma is critical for the fast ignition scheme of laser fusion [2] as well. 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]. Most high power lasers have levels of prepulses and pedestals that will drive significant plasma formation and hydrodynamics, making the interaction extremely complex. The instantaneous profile of the plasma density is a crucial parameter that controls much of the light coupling physics [3], and hence the nature of the plasma at longer time scales, and is therefore a crucial input for any simulation of the coupling of laser or particle energy to laser-driven fusion targets. However, the plasma density profile continuously changes throughout the interaction. The plasma in the interaction region moves on ultrafast time scales and, hence, is difficult to track. Here, we present the first high resolution temporal mapping of these ultrafast dynamics using femtosecond pump-probe Doppler spectroscopy. Monitoring the time-dependent Doppler shifts in a femtosecond probe beam, we are able to infer ultrafast evolution of plasma density profiles.

Most of the light coupling in femtosecond laser interactions occurs essentially at the critical surface where strong plasma oscillations can be excited [4,5]. Giant magnetic fields are found around this surface [6], and relativistic oscillations of this surface can up-shift incident light to higher-order harmonic, attosecond pulses [7]. In the four decades of investigation of laser-produced plasmas, the dynamics of coupling of long (nanosecond) pulses have been investigated thoroughly in experiments, analytical theory, and computer simulations [8], but the long pulses integrated out the rapid motion of the critical surface, and thus its motion was never probed in real time. In the ultrashort regime, previous literature have reported similar studies at moderate intensity levels [9–13] ranging from the presence of ponderomotive pressure causing spectral redshift even at low intensities (10^{15} W/cm^2) [10] to the dependence of spectral shifts on intensity and chirp of the input laser pulse [12,13]. However, all these studies [9–15] present a convoluted picture of the "instantaneous" profile as the measurements relied on reflection of the irradiating pulse itself. An independent ultrashort probe pulse is required to understand the complex plasma dynamics unambiguously.

Here, we report the first attempt—to the best of our knowledge—to unravel the ultrafast dynamics of high density plasma layers at relativistic intensities. Previous studies were limited to modest intensities [9,10], and there does not appear to be a clear picture of the crucial, ultrashort time-period evolution of the plasma density, which is extremely important for useful discrimination of the energy and mass transport within the target. In this Letter, we provide this crucial, missing perspective.

We create plasma from a metal target by using an 800 nm laser pulse of 30 fs duration focused to an intensity of 5×10^{18} W/cm^2 and interrogate the plasma at different later "instants" with a 400 nm, 80 fs laser pulse, tracking the plasma evolution at the position of the probe critical surface throughout the 40 ps after the interaction with the pump pulse. We present clear evidence for the initial inward motion of the plasma in this region, which is reversed at later time, and derive instantaneous velocity and acceleration profiles. We show that the experimental results are consistent with the results of a hybrid simulation that uses a 1D electromagnetic particle-in-cell (PIC) simulation for the initial evolution and a hydrodynamics simulation for the later times [16,17]. We believe our results are very important for studies that seek to understand energy and mass transport in the plasma at relativistic intensities.

Figure 1 shows the experimental setup for the pump-probe Doppler spectrometry that we adopted. A p-polarized pulse from the tabletop Ti:sapphire chirped pulse amplification laser (30 fs, 800 nm, 20 TW maximum) at Tata Institute of Fundamental Research is used as a pump beam and is focused at a 40° angle of incidence onto the target, by using an off-axis parabola. The measured focal spot diameter of 16 μm gives peak intensities of 5×10^{18} W/cm^2. The pump pulse has a nanosecond contrast of 3×10^{-6}. A small fraction of the pump pulse is extracted by a beam splitter and frequency doubled in a 1 mm thick β-barium borate crystal to generate a probe pulse at 400 nm (Fig. 1, inset), with a duration of 80 fs. A BG-39 filter is used to remove the residual 800 nm (Fig. 1, inset) present in the probe beam. The probe beam is delayed with respect to the pump by using a delay stage of 1 μm precision. The probe beam incident nearly normal to the target is focused to a diameter of 60 μm (intensity of 3×10^{12} W/cm^2) and spatially overlapped with the pump focal spot. Temporal overlap between pump and probe 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 delay, there is a sharp transition in reflectivity from a high value to a lower value [18]. The reflected probe pulse is divided by a beam splitter and fed to two different spectrometers (Fig. 1): a UV-visible spectrometer, SP1 (Ocean Optics HR-2000, 350–445 nm, resolution 0.5 Å), and another, SP2 (Avantes, 245–505 nm, resolution 1.3 Å). The two spectrometers are used for *in situ* normalization and to establish the repeatability and consistency of the measurements. We present only the spectra measured by the spectrometer SP1, averaged over 50 shots at each time delay. We wish to point out a very interesting advantage of using 400 nm pulses for probing the plasma, apart from the usual one of elimination of pump noise. This is the narrow and smooth spectrum at 400 nm under our experimental conditions, in contrast to the highly modulated and broad spectrum at 800 nm (which is a result of the pulse shaping by the "Dazzler" acousto-optic modulator and modifications in the different amplifiers). The narrow spectrum facilitates the observation of small wavelength changes, as shown later.

The quantitative shift or change in the normalized reflected spectra of the probe beam from the plasma surface illustrates the overall features of the Doppler spectrometry

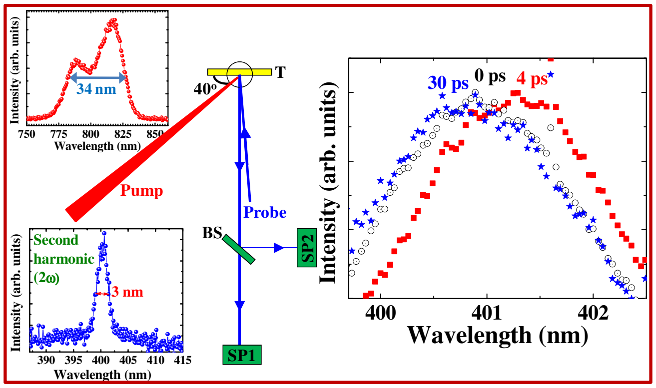

FIG. 1 (color online). Schematic of the experimental setup: target (T), beam splitter (BS), Ocean Optics HR-2000 (SP1), and Avantes (SP2) UV-visible spectrometers. The inset at top left shows fundamental (800 nm, pump) and that at bottom left shows second harmonic (400 nm, probe) pulses. The inset at right shows normalized reflected spectra of the probe beam from the plasma surface at time delays of 0 (black, hollow circle), 4 (red, filled square), and 30 ps (blue, filled star).

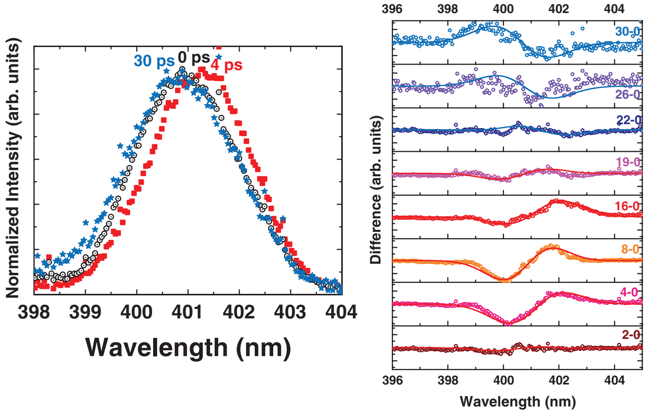

FIG. 2 (color). Right: Time-delayed spectra derived after subtracting spectrum at zero time delay. Left: Normalized spectra of the reflected probe beam for the aluminum target are shown for a comparison.

experiment. We have recorded the spectra for different solid targets and point out interesting features in the observations. The inset in Fig. 1 (right) shows the typical behavior of the normalized spectra for optically polished aluminum targets. The black curve is the spectrum at zero time delay, while the red and the blue curves are at 4 and 30 ps delay, respectively. It is clear that the spectrum at the earlier delay is redshifted, while that at later time is blueshifted.

While the shifts are evident, to clearly portray the differences a reference (zero time delay) spectrum is subtracted from all time-delayed spectra. The subtracted spectra are shown in Fig. 2. The right panel shows that for aluminum at a pump intensity 5×10^{18} W/cm^2, the reflected probe pulse is redshifted 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 an electron density of 6.3×10^{21} cm^{-3}. Our data thus indicate that this plasma layer initially recedes from the probe, pushing into the target. At later times, this surface moves back and accelerates towards vacuum. These data show a "turn around" time of around 19 ps. We comment on the magnitude of the shifts a little later.

Figure 3(a) shows the measured shift in the wavelength for the aluminum target at various delays and Fig. 3(b) the results of the 1D HYADES code along with 1D electromagnetic PIC simulations. The temporal resolution in our experiment enables a deduction of instantaneous velocity [Fig. 3(c)] and acceleration [Fig. 3(d)] of the critical surface. The shifts are consistent with velocities of the order of 10^7 cm/s expected from simple analytical estimates [$0.5c\Delta\lambda/\lambda(1/\cos\theta)$] [10], while the acceleration is found to be of the order of 10^{18} cm/sec^2 [13]. The velocities are similar to those found by Kalashnikov *et al.* [11], which were derived from instantaneous second harmonic reflection from the plasma in a single pulse (2 ps duration) experiment.

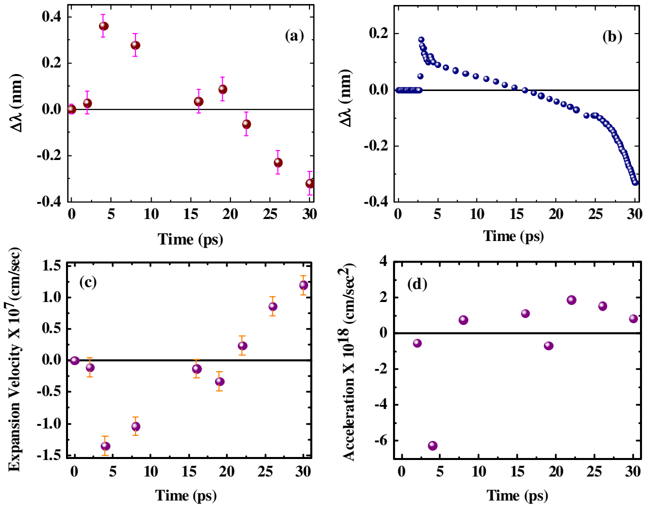

FIG. 3 (color online). Doppler shift in reflected probe spectra (a) experimental and (b) hybrid simulation that uses a 1D electromagnetic PIC simulation for the initial evolution and a hydrodynamic code for the later times. Calculated (c) expansion velocity and (d) acceleration from Doppler shift.

The modeling of the target hydrodynamics under the influence of the laser prepulse was performed by using the HYADES code [16]. HYADES is a 1D Lagrangian radiation hydrodynamics simulation code with a flux-limited diffusion model for electron conduction and a multigroup diffusion model of thermal radiation flow within the target.

In the modeling, a 500 μm thick target is divided into 100 Lagrangian cells, which are feathered toward the driver-facing surface. A flux limiter of 0.06 is employed, which is appropriate for the prepulse interaction of the pump beam. The results of HYADES modeling as pertains to the interaction of the main pulse with the target cannot be expected to accurately reflect the interaction, so a PIC code is employed 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 radiation hydrodynamics code.

The next stage in the numerical modeling was to determine the heating of the plasma by the main pulse. This was done by using a 1D electromagnetic PIC code including electron-ion collisions. This code is a modified version of the one used in Ref. [17]. A laser pulse that matches the experimental parameters is normally incident on fully ionized aluminum plasma. The density profile of this plasma matches that from the HYADES calculations up until about $10n_c$. The plasma is initially at 300 eV (which is not far from the temperatures predicted by HYADES over this electron density range). It models the region around the 800 and 400 nm critical densities fairly well in terms of initial temperature and density. It is not a good model of the denser and colder regions of the target; however, they are not so relevant to the experimental observations.

In the PIC simulation, the 30 fs main pulse creates a burst of fast electrons that propagate deep into the target (beyond $10n_c$). The propagation of these fast electrons strongly heats the plasma beyond the critical surface. The simulation was run up to 350 fs. A background temperature profile was obtained at this point, and this was used, along with the density profile from HYADES, in a 1D hydrodynamic simulation. The simulation was done by using an Eulerian code that assumes an ideal gas and does not contain thermal conduction or radiation transport. The motion of the 400 nm critical surface is tracked in this simulation, and from this an observable wavelength shift is determined.

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.,

$$\Delta\lambda = \frac{2\dot{x}_{\mathrm{crit}}\lambda_{\mathrm{probe}}}{c},$$

where the velocity of the critical surface ($\dot{x}_{\mathrm{crit}}$) was derived by differentiating the position of the 400 nm critical surface at a certain instant and c is the speed of light. Postprocessing the above calculation yields the wavelength shift as a function of probe time delay, and this is shown in Fig. 3(b). It is interesting to see the good quantitative agreement in the amount of spectral shift.

By examining the simulation output in light of Fig. 3(b), the experimental measurements can now be physically interpreted as follows. The prepulse of the pump laser creates plasma and launches a compression wave into the front surface of this plasma. At early times the position of the critical surface for the shorter wavelength probe beam sits on this compression wave and tracks it as it moves into the target. Thus, at early times the probe is tracking the compression wave into the front surface plasma and a redshift is measured. In addition to the compression wave, there is also the plasma rarefaction that is happening in the front surface plasma (i.e., the expansion of the plasma into the vacuum). At later times the compression wave has propagated into a region of overdense plasma, 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 blueshift is measured.

The simulation and analysis of the simulation results show that the qualitative and quantitative behavior of the spectral shift can be understood and that the simulations are within a factor of 2 of an accurate quantitative description. This means that the experimental approach used in this study can provide detailed insights into the dynamics of the plasma 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 target.

In conclusion, we report the first ever measurement of plasma dynamics with a relativistic intensity femtosecond laser over an extended time period. Our study provides unprecedented and vital data for improving the modeling of ultrahigh intensity femtosecond laser interaction with overdense plasma. Time-resolved ultrafast Doppler spectrometry is a powerful technique to infer fast dynamics in dense plasma systems. At the intensities we use in the present experiment, the plasma motion is mostly driven by hydrodynamics and occurs on a time scale of a few picoseconds. At higher intensities, ponderomotive pressure becomes more significant and critical surface oscillations can approach the speed of light. These oscillating surfaces can up-shift the driving pulse to high harmonic attosecond pulses. Doppler spectrometry with few-cycle pulses can gather invaluable information regarding these otherwise obscure ultrafast dynamics.

G. R. K. acknowledges a DAE-SRC-ORI grant.

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