Studying the Dynamics of Relativistic Laser-Plasma Interaction on Thin Foils by Means of Fourier-Transform Spectral Interferometry

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We apply Fourier-transform spectral interferometry (FTSI) to study the interaction of intense laser pulses with ultrathin targets. Ultrathin submicrometer-thick solid CH targets were shot at the PHELIX laser facility with an intensity in the mid to upper 10^{19} W/cm² range using an innovative double-pulse structure. The transmitted pulse structure was analyzed by FTSI and shows a transition from a relativistic transparency-dominated regime for targets thinner than 500 nm to a hole-boring-dominated laser-plasma interaction for thicker targets. The results also confirm that the inevitable preplasma expansion happening during the rising slope of the pulse, a few picoseconds before the maximum of the pulse is reached, cannot be neglected and plays a dominant role in laser-plasma interaction with ultrathin solid targets.

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Studying the interaction of relativistic laser pulses with solid targets is a very rich field because of the many effects that occur simultaneously at the laser-solid interface. In this picture, the laser interacts with electrons that reach relativistic velocities in the laser field and collective effects can appear like strong charge displacements (currents, electrostatic fields). These in turn can accelerate the background ions of the target either from the surface, because of the generation of a dense electron sheath [1], or from the bulk [2]. The potential applications of such particle beams are numerous, from fast ignition [3,4] and proton imaging [5] to basic science in laboratory astrophysics [6] and nuclear physics [7].

In the laboratory, a much more complicated picture emerges from experiments aiming at studying laser-driven ion acceleration, because additional phenomena like the hydrodynamical preexpansion of the target during the rising slope of the laser pulse is a major issue. This preplasma strongly favors additional effects like self-focusing and filamentation, self-phase modulation, and laser absorption, which could supposedly change the interaction outcome [8]. In addition, the thin targets proposed in most of the advanced schemes [2,9,10] are very fragile, and they can undergo an uncontrolled massive hydrodynamical expansion in the last instants before the interaction with the high-intensity part of the pulse begins, yielding classically or relativistically

transparent propagation conditions, far from the conditions required by the acceleration models.

Tremendous work has been done to attempt to experimentally demonstrate laser-driven acceleration to high particle energies, but the experimental conditions and limitations have nearly always limited the success of such efforts. One of the underlying reasons for that is a poor quantitative understanding of the real plasma conditions at the beginning and during the interaction. Therefore, it is essential to have a much more precise basic knowledge in laser-solid interactions by developing models and evaluating them with experiments.

In order to quantify the laser-plasma interaction precisely, a suite of diagnostics has been traditionally employed. They include mostly electron and x-ray spectrometers to study the plasma self-emission and/or shadowgraphy techniques to characterize the plasma conditions. More recently, time-resolved optical diagnostics based on frequency-resolved optical gating (FROG) have proven to be useful tools to study the position and velocity of the laser ablation front [11] or the breakout of a light pulse through a thin target as the result of both laser hole boring and relativistic transparency effects [12].

In this Letter, we report on the first use of Fourier-transform spectral interferometry (FTSI) [13] as optical diagnostic for the characterization of the laser plasma interaction under realistic conditions at the PHELIX laser facility. To the lowest order, FTSI enables measuring precisely the time delay between two laser pulses at different relativistic intensities. Compared to previous works, FTSI gives access to some information on the transit time of a pulse through a thin target with high accuracy; we show in

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the following that this can be used to characterize the dominating effect in the interaction, namely, laser hole boring or relativistic transparency. For instance, when the intensity ratio between the two pulses is set such that one pulse experiences a relativistically transparent plasma and the second is applying hole boring, the pulse-to-pulse delay is maximized. In particular, we find that, although using state-of-the-art temporal contrast optimization equipment, thin submicrometer foils irradiated with PHELIX undergo a significant preplasma expansion happening in the last tens of picoseconds before the maximum of the pulse is reached. This evidence is supported by numerical hydrodynamical simulations of the recently upgraded RALEF-2D code [14].

To understand how the potential of FTSI can be exploited, let us at first consider the propagation of a laser pulse in a plasma that is near or overcritical. A laser pulse propagating in an underdense plasma experiences an index of refraction that is mostly lower than unity. When the light intensity is such that the free electrons of the plasma are accelerated to relativistic speeds, the optical properties of the plasma are influenced by the relativistic Lorentz factor γ of the electrons. For instance, the group velocity of a light pulse propagating in a plasma depends on its intensity. This propagation speed is the relativistically corrected group velocity of the pulse, given by

$$v_g = c \sqrt{1 - \frac{n_e}{\gamma n_c}}, \quad (1)$$

where c is the light speed in vacuum, n_e is the electron density, and n_c is the critical plasma density. Although very low group velocities can be expected when the electron density approaches the relativistically corrected critical plasma density, it is experimentally not realistic to observe this effect with state-of-the-art machines. This is due to the submicrometer propagation length at stake combined with the narrow width of this type of resonance.

When the plasma is overcritical, the laser pulse pushes the electrons forward because of its ponderomotive force and makes its way inside the plasma at the hole-boring velocity v_h [15],

$$v_h = c \sqrt{\frac{\Delta p n_c Z m_e I \lambda^2}{p n_e M 2.7}}, \quad (2)$$

where the ratio $\Delta p/p$ represents the fraction of the pulse momentum transfer, Z and M are the ionization degree and atomic mass of the plasma, respectively, m_e is the electron mass, and the product $I \lambda^2$ is expressed in the units of $10^{18} \text{ W} \mu\text{m}^2/\text{cm}^2$. For intensities in the range of 10^{19} to 10^{20} W/cm^2 , hole-boring velocities of the order of a few percent of c are to be expected, which has been also verified by experimental data [16]. In the hole-boring picture, the laser pulse pushes a plasma layer as long as the target can last. When the pulse is sufficiently long and the target thin enough, the pulse may emerge from the rear side of the target after a hole-boring time τ ,

$$\tau = \frac{dz}{c} \sqrt{\frac{p}{\Delta p} \frac{2.7}{I \lambda^2} \frac{M}{Z m_e} \frac{n_e}{n_c}}, \quad (3)$$

where dz is the plasma length. Given the hole-boring velocities that can be expected experimentally, it is foreseeable that, for relatively thin targets below $1 \mu\text{m}$, a measurable delay of the laser pulse should occur.

To measure this quantity, we realized the experiment described in Fig. 1 at the PHELIX facility [17] using the high temporal contrast option of the laser [18]. In such conditions, the nanosecond pulse pedestal generated by amplified spontaneous emission stays below the ionization threshold of the most sensitive materials, and the plasma generation happens during the rising slope of the pulse. A double-pulse structure is created in the front end of the laser using a Mach-Zehnder arrangement with an adjustable delay and energy ratio. Both pulses are perfectly collinear and exhibit an energy ratio of 1 : 10, a pulse duration of about 500 fs and a pulse to pulse delay of 3 ps. The total energy available during the experiment was between $94(\pm 4) \text{ J}$ on the target (85 J for the main pulse). This corresponds to a maximum laser intensity of $6(\pm 2) 10^{19} \text{ W/cm}^2$. The light leaking through the target is projected onto a screen for energy transmission measurement and the central portion of the beam is attenuated and transported to a fiber-coupled optical spectrometer (Ocean Optics HR2000). In addition, a second spectrometer located at the end of chain sensor of the laser records reference spectra used later on in the analysis.

We performed a series of shots on submicrometer-thick CH_2 targets fabricated at the target laboratory of the Ludwig Maximilian University in Munich. Although the amount of light transmitted through the target dramatically decreases with increasing target thickness, it is possible to record spectra that are modulated for all targets below $1 \mu\text{m}$ when the right neutral density filters are employed (see Table I). All spectra are three to five times broader than the initial laser pulse spectrum because of the relativistic self-phase-modulation (RSPM) happening during the propagation

FIG. 1. Experimental setup. OAP denotes an off-axis parabola.

TABLE I. Spectral interferometry signals for significant shots. The shot number and target thickness are indicated above the spectrum.

raw experimental data in the range [1035 nm - 1075 nm]	14022 (200 nm)	14104 (400 nm)
14108 (500 nm)	14106 (700 nm)	14107 (900 nm)

through the target. RSPM happens when the pulse propagates in an underdense plasma and the index of refraction of the plasma is influenced by the Lorentz factor of the electrons such that a rapid change of the intensity during the rising and falling slopes of the pulse results in the creation of new frequencies. Here, one notices that the spectra corresponding to the thicker targets are mostly enlarged on their blue side, indicating a dynamical shuttering effect as reported in [12].

We analyzed these experimental spectra using the FTSI [13] method to measure the spectral phase difference between the two pulses and retrieve the pulse-to-pulse delay. Here, the spectral interference, obtained by analyzing the double-pulse structure with a spectrometer, yields the phase difference between these two pulses with high precision. As an illustration, when both pulses are identical with spectral intensity $I(\omega)$, pulse-to-pulse delay τ , and intensity ratio $1:\eta$, the spectral signal S recorded by the spectrometer is given by

$$S(\omega) = I(\omega)[1 + \eta + 2\sqrt{\eta}\cos(\omega\tau)], \quad (4)$$

which includes both constant and modulation parts. The retrieval of the delay is obtained by taking the Fourier transform of the spectrum, isolating the modulation frequency using a numerical filter, and taking the argument of the inverse Fourier-transformed filtered signal. In such a case, the resulting function φ becomes

$$\varphi(\omega) = \omega\tau \quad (5)$$

and the delay between the two pulses equals the slope of φ . It is possible to use this property to precisely measure the delay between the two pulses, even for intensity ratios as high as ten or more, as the fringes are visible over a large intensity ratio range. In the experiment described above, the delay between two pulses of different intensities propagating through a thin target is compared to the pulse delay before the interaction point.

Figure 2 illustrates the precision of the measurement by showing the initial reference spectrum measured before the

FIG. 2. Measured (blue) and reconstructed (green) spectra based of the spectrum of shot 13992 (red) and a modulation given by Eq. (4). The data used are $\tau = 3.0896$ ps and $\eta = 0.090$.

target (blue) compared to a reconstructed spectrum using a single-pulse spectrum from another shot (red) and two fit parameters, η and τ . A rough value for τ is found at first by Fourier-transform analysis of the experimental curve with a precision of a few femtoseconds. Then, a fine-tuning is performed to get rid of the horizontal offset between the fringes, yielding an enhanced precision of 0.2 fs for τ . To perform the measurement for the instants when the pulse intensity is maximum, we restrict the analysis to the spectral range of the incident pulse in the range [1050–1060 nm]. Indeed, during the rising and falling edges of the pulse, the rapid change in intensity is responsible for RSPM and spectrally shifts the information related to these instants. For this reason, restricting the analysis to the initial laser spectral interval is equivalent to time gating the measurement to the time when the intensity is close to its maximum. In other words, the measurement is done for a narrow intensity range approaching the maximum intensity of the pulse, while the effects related to the rising and falling edges of the pulse are spectrally shifted out of the considered spectral window.

Figure 3 shows the result of the phase analysis for the PHELIX shot 14022 (200-nm foil) as an example. Here, the stronger phase slope indicates a longer delay between the two pulses induced by the propagation through the plasma. Figure 4 shows the results of the analysis for targets with a thickness varying between 200 and 900 nm, irradiated

FIG. 3. Phase measurement before and after the target.

FIG. 4. Delay change between the main and post pulse as a function of the target thickness.

with two pulses having an intensity ratio η of 10 and an initial delay of 3 ps. The measurement shows a delay that monotonically decreases with increasing target thickness. For the thinnest target shot with the double-pulse structure, an increase of the pulse-to-pulse delay by 505 fs is observed. This indicates that the second pulse is crossing the plasma at a much lower speed than the first one. This can be explained if the target has undergone enough preexpansion to become relativistically transparent for the main pulse but is still opaque for the second one. For targets thicker than 400 nm, the pulse-to-pulse delay drops rapidly to values at or below 150 fs and stabilizes around 0 for the thickest target shot in this experiment.

An illustration of the experimental results can be obtained when a simulation of the hydrodynamical preplasma expansion is conducted. For relativistic intensities that are confined to the vicinity of pulse maximum, particle-in-cell (PIC) codes are well suited. But the preplasma expansion, which starts as soon as the ionization threshold is reached, takes place on many tens of picoseconds, a time delay that is out of reach for PIC simulations. The corresponding plasma motion was modeled with help of the hydrodynamical simulation code RALEF-2D. This is an arbitrary Lagrangian-Eulerian fluid code with multigroup radiation transport, which solves one-fluid one-temperature hydrodynamic equations in two spatial dimensions on a quadrilateral mesh of multiblock structure using a second-order Godunov-type numerical scheme. The transfer equation for radiation is integrated with the method of short characteristics in the quasistatic approximation. The code has been recently upgraded to include laser propagation following the wave equation (diffraction, reflection, and absorption). In our previous work [19], we validated a previous version of RALEF-2D against the preplasma expansion driven by the nanosecond amplified spontaneous emission pedestal of short light pulses amplified in chirped pulse amplifiers. Now we apply RALEF-2D to the simulation of the target expansion over the last 100 ps before the pulse maximum is reached from the ionization threshold to 2×10^{17} W/cm². The pulse profile of the laser has been measured at the laser front end by a scanning third-order cross correlator (Sequoia). Since the laser amplifier is not saturated and exhibits a low B integral, we believe that this profile is representative of the fully

FIG. 5. Simulation of the target hydrodynamical preexpansion during the rising slope of the pulse (until 3 ps before the maximum intensity is reached), supposing various initial target thicknesses. The electron density profiles are given on the laser axis at the end of the hydrodynamical simulation. The dotted horizontal lines at n_c and n_0 indicate the plasma critical density and the density at roughly solid-state density, respectively.

amplified pulse profile. The result, discussed in detail elsewhere [18], shows that the ionization threshold of CH₂ is reached about 100 ps before the maximum of the pulse occurs. In the simulation, a scaled profile reaching 7×10^{19} W/cm² at maximum has been used with equations of state and opacities were provided by the Keldysh Institute of Applied Mathematics (Moscow) [20].

The simulations were performed in axial symmetry for various targets in the submicrometer range, with a focal spot of 4 μ m in accordance with the experiment. The corresponding on-axis spatial distributions of the electron density are shown in Fig. 5. For targets thicker than 500 nm, the preplasma profile is mostly identical with a scale length of roughly 3 μ m. However, thinner targets are heated significantly during the rising slope of the pulse and they have started to expand. Targets of 100-nm initial thickness and lower have expanded to densities near the critical plasma density, and these targets are expected to become transparent. Finally, for targets in between, an interesting regime can be found where the target appears to be relativistically transparent but nonrelativistically opaque. Using Eq. (3) and substituting I for the estimated post-pulse intensity and the measured hole-boring time for the 200-nm target (expanded to 1.25 μ m at full width half maximum), one finds a corresponding electron density of $9.9(\pm 2) \times 10^{21}$ cm⁻³. This value matches the predicted maximum electron density by RALEF-2D for this target thickness. Although this result is remarkable, one should stress that it should only be regarded as a qualitative validation of the model. The measurement with 200-nm target thickness illustrates an interesting and important case. While the main pulse can transit due to relativistic transparency, the weaker second pulse must bore through.

In conclusion, employing FTSI to intense laser pulses transmitted through targets in the submicrometer thickness range offers unique insights into the highly dynamic

processes in plasmas irradiated at relativistic light intensity. In particular, we found that targets below 400 nm pre-expand sufficiently such that the electron density drops to a point where relativistic transparency can happen. This experimental observation has the potential to explain why experimental data on particle acceleration at very small target thickness has so far failed to meet the expectations derived from theoretical models and numerical simulations such as PIC codes, which do not take into account the picosecond plasma dynamics prior to the peak intensity. Other parameters could be extracted with FTSI from the interference pattern, like the pulse-to-pulse energy ratio, to get additional information on absorption processes. This could be of particular interest for experiments aimed at optimizing the coupling of light into secondary radiation like laser-driven gamma ray generation. Finally, coupling the FTSI setup presented in this article with spectral phase measurement devices like FROG will enable providing a full reconstruction of the probe-pulse temporal structure, which will help to gain even more insight into the rich physics of laser-plasma interaction at ultrahigh intensities.

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