Subpicosecond dynamics of pre-plasma on a solid, formed by a ultra-high contrast, relativistic intensity pulse

Ankit Dulat1,*, C. Aparajit1, Anandam Choudhary1,*, Amit D. Lad1, Yash M. Ved1, and G. Ravindra Kumar1,*

1 Tata Institute of Fundamental Research, 1 Homi Bhabha Road, Colaba, Mumbai 400 005, India
*Corresponding author: ankit_090@tifr.res.in; and grk@tifr.res.in

Abstract

Using spectral interferometry technique, we measured subpicosecond time-resolved pre-plasma scale lengths and early expansion (< 12 ps) of the plasma produced by a high intensity (2 × 10^{18} W/cm^2) pulse with ultra-high contrast (10^{-9}). We measured pre-plasma scale lengths in the range of 3-15 nm. This measurement plays a crucial role in understanding the mechanism of laser coupling its energy to hot electrons and hence important for laser-driven ion acceleration and fast ignition approach to fusion.

1 Introduction

With recent advances in lasers, the interaction of high-power femtosecond pulses of ultrahigh contrast (UHC), with solid density plasma is now accessible, facilitating high energy density science. Unlike pulses having intense picosecond wing and nanosecond pedestal where interaction occurs in expanding, lower than solid density plasma, UHC pulses cause ‘instantaneous’ excitation of solid density matter [1].

The interaction mechanism of such clean pulses depends on well-defined initial conditions, especially on the scale length of the steep density (l_s << λ) pre-plasma [2, 3] formed before the arrival of the main pulse. Earlier studies have shown, the critical dependence of pre-plasma scale length on the laser absorption [4], the energy distribution of heated electrons [5, 6], angular distribution [7], and divergence of fast electron beam [8]. High harmonic generation from the interaction of ultra-intense pulses with solid density plasma also depends on the pre-plasma scale length [9]. Various other applications such as the fast ignition approach to fusion [5] and laser-driven ion acceleration [10] depend on the efficiency of coupling laser energy into fast electrons, which is sensitive to the scale length of preformed plasma. Therefore it becomes important to accurately measure the scale-length of the pre-plasma, which is extremely hard to measure and remains mostly unknown for high contrast relativistic pulses.

Until now, different techniques have been used to study the early evolution (after the arrival of the main pulse) of plasma with sub-picosecond time scale, but only a few have been used to study the pre-plasma conditions, which have their own limitations. Schlieren imaging technique [11] and shadowgraphy technique [12] have been widely used to locate the critical density layer, however diffraction effects limit the spatial resolution to the order of incident wavelength. Spatial interferometry [13] has been used earlier, to measure the time evolution of density profiles but again it suffers from limited spatial resolution. Other techniques like time-resolved spectroscopy of back-reflected light and Doppler spectrometry [14] are limited to observation of the dynamics at later delays because there is not much expansion/motion before the arrival of main pulse. Resonance absorption spectroscopy (RAS) [3, 15] has been used earlier, to measure the pre-plasma scale length by measuring plasma reflectivity differences between S and P-polarized light as a function of the angle of incidence, but it depends critically on the intensity and accurate modeling of the preformed plasma conditions, especially at high intensities. Unlike RAS, which is an indirect way of measuring pre-plasma evolution, frequency domain interferometry (FDI or spectral interferometry) technique has an advantage of directly inferring the plasma dynamics from the measured phase shifts. Also, it can measure pre-plasma length scales with nanometer spatial resolution. It has been used earlier to measure the dynamics at positive delays of the plasma at relatively low intensities (<10^{16} W/cm^2) [2, 16].

In this paper, using frequency domain interferometry, we measured the scale length of the pre-plasma with nanometer spatial resolution and early expansion of the plasma at positive delays, created by 800 nm, high contrast (10^{-9}), 27 femtosecond (fs) pulses at a peak intensity of 2 × 10^{18} W/cm^2.

2 Experiment

The experiment (Fig. 1a) was performed with a 150-terawatt Ti:sapphire laser, which can deliver 800 nm, 27...
fig. 1. (a) Schematic of experimental set-up for spectral interferometry. M1-M7: Mirrors, BBO: β-barium borate, BG: Blue-green (BG-39) filter, DS: Delay stage, BS: Beam splitter, L1-L3: Lenses, OAP: Off-axis parabola, SP: Spectrometer (b) Intensity contrast of 800 nm, measured using SEQUIOA [17, 18]. (c) Pulse-width of 800 nm (in red), measured using SPIDER [19] and pulse-width of 400 nm (in blue), measured using SD-FROG [20]. FWHM of the two pulses is labeled by $t_{1/2}$.

Femtosecond pulses. The measured picosecond intensity contrast and pulse width of the laser are shown in Fig. 1(b) and 1(c) respectively.

P-polarized laser pulses (100 mJ pulse energy) were focused with an f/3 off-axis parabolic mirror (OAP) to a 7 µm spot at an incidence angle of 45°, creating a peak intensity of $2 \times 10^{18}$ W/cm². We used transparent dielectric target (BK7 glass) with Al coating of 200-nm thickness at the front. The target was moved between laser shots to provide a fresh surface to each laser shot. A small fraction (5%) of the main laser beam was extracted and up-converted to its second harmonic (400 nm) by a β-barium borate (BBO) crystal (2 mm thick). The measured temporal profile of this second harmonic (SH) pulse is shown in Fig. 1(c). SH pulse was split into two pulses with a Michelson interferometer (MI): a reference pulse (arriving before the pump) and a probe pulse (reaching after the pump). A BG-39 filter was used after the BBO crystal to remove residual 800 nm light. One of the mirrors (M4) in MI is mounted on a high-precision motor stage which can be used to change the delay between the probe and reference pulses. Since the fringe spacing in FDI is inversely proportional to the delay $t$ between these pulses, we have kept a fixed optimum delay (T) of 5.3 ps in our experiment (depending on the resolution of our spectrometer). The probe pulse was time-delayed with respect to the pump pulse using a high precision motorized retro-reflector delay stage.

The pair of two pulses (reference and probe) were focused onto the target at near-normal incidence ($\sim 5^\circ$) with a convex lens (L1) to a spot of $\sim 120$ µm (measured by knife-edge method). The focal spot size of the probe and reference beams was several times larger than the pump focal spot size to permit uniform illumination of the region under test. Spatial overlap between the pump and the probe beams was ensured by high-resolution imaging. The reflected probe and reference pulses were collected by an f/2 plano-convex achromat lens and focused on single-mode fiber (Thorlabs, SM-300), which couples the light to a high resolution (0.3 Å) spectrometer (OII, HR-4000). The temporal overlap between the pump and the probe pulses was monitored by observing the probe reflectivity as a function of probe delay. The experiment was performed in a vacuum chamber at $10^{-5}$ Torr.

3 Results and Discussion

Spectral Interferometry relies upon measuring quantitative phase changes in probe pulse that is reflected from the plasma created by the pump laser. Fig. 2 shows three spectral interferograms, each measured for a different delay ($\tau$) with respect to the pump. For each delay, the interferogram in ’red’ is without any plasma on the target, while that in ’blue’ is the phase-shifted one due to the interaction of probe pulse with the plasma. As observed, the interferogram gets more phase-shifted and
its amplitude decreases as plasma evolves. The phase shift in the probe pulse due to plasma has been calculated from the measured modulated spectrum using the Fast-Fourier-Transform (FFT) based numerical algorithm [22]. Details of the phase extraction procedure are given in the Appendix.

The two main possible contributions to this phase change are (1) changes in the refractive index of the material between the reflecting surface (critical density layer) and the observer, due to rapid ionization of the material and subsequent change in the electron density distribution. (2) due to the rapid expansion of the plasma and motion of the reflecting surface (Doppler phase shift).

The extracted total phase is shown in Fig. (3). The error bars are due to shot-to-shot fluctuations of input laser energy and background vibrations which perturbs the interferogram. We observed a non-zero but small phase change at negative delays of -6.6 ps (before the main fs pump pulse hit the target) which is indicative of presence of significant plasma. Since our fs pulse doesn’t have any nanosecond prepulse, we expect this phase change to be mainly due to the ionization of the material by picosecond (ps) pedestal, which results in a very steep density profile. Subsequently, there is a tiny bit of gradual increase in this phase shift as we approach time-zero. Around time-zero \( \tau = 0 \), there is a rapid fluctuation in the phase. This phase shift is mainly due to the intense ionization of the target around the peak of the pulse and subsequent explosive expansion of low-density fast electron cloud (accelerated by various mechanisms) escaping the target. The oscillation in the phase around \( \tau = 0 \) could be related to the ionization by intense \( (10^{15} \text{ W/cm}^2) \) pre-pulse (at \( \sim -1 \text{ ps} \)) and post-pulse (at \( \tau < 5 \text{ ps} \)) as shown in Fig. 1(b).

\[ \phi(t) = 2 \int_{\infty}^{z=z_c(t)} \frac{\omega}{c} \sqrt{\epsilon(s)} ds \approx 2\omega \left( \int_{\infty}^{z=z_c(t)} \frac{1}{2n_e} \right) \frac{1}{n_c(s,t)} ds \]  

where \( s \) is the coordinate along beam path, \( \omega \) is the angular frequency, \( c \) is the speed of light in vacuum, \( \epsilon \) is the permittivity that can be approximated as \( \epsilon = 1 - (n_e/n_c) \) with \( n_e, n_c \) is the local electron density and critical density respectively. The imaginary part in \( \epsilon \) (due to ion-electron collisions) has been neglected since it mainly causes absorption of the probe pulse, contributing weakly to the phase change. The measured phase shift is \( \Delta \phi = \phi - \phi_0 \), where

\[ \phi(t) = 2 \int_{\infty}^{z=z_c(t)} \frac{\omega}{c} \sqrt{\epsilon(s)} ds \]

Fig. 2. Measured interferogram at different time delays \( \tau \). Interferogram in red (Ref) is without plasma, in blue is with plasma on target. Insets show the zoomed central region of the interferogram.

Fig. 3. Time-resolved total phase shift calculated from FDI data, is compared with Doppler phase shift, calculated using the earlier Doppler spectrometry data (Jana et al. [14]).

The phase shift at later time delays is mainly due to the rapid expansion of background plasma which has been heated through energy transfer from the fast electrons [14]. This phase shift can not be fitted with a linear curve, indicating that at early times (< 10 ps) plasma is not expanding with a constant velocity, which is consistent with earlier measurements (at similar experimental conditions) using Doppler spectrometry.

The total phase shift experienced by the probe beam can be expressed as the path integral through the plasma up to point where the beam is reflected \( (z_c) \) and back \( (z = 0 \) being the target surface intial position) [23].

\[ \phi(t) = 2 \int_{\infty}^{z=z_c(t)} \frac{\omega}{c} \sqrt{\epsilon(s)} ds \approx 2\omega \left( \int_{\infty}^{z=z_c(t)} \frac{1}{2n_e} \right) \frac{1}{n_c(s,t)} ds \]  

[14]
\[ \phi_0(t) = 2 \int_0^\infty \left( \frac{\omega}{c} \right) ds \]  

(2)

is the phase experienced by the reference pulse. The first term in total phase shift is only due to the motion (Doppler phase shift) of the reflecting surface and can be re-expressed as:

\[ \frac{d\phi}{dt} = 2\omega_0 \left( \frac{nu}{c} \right) \cos(\theta) \]  

(3)

where \( u \) is the velocity of the critical surface and \( n \) is the refractive index. Using the velocity data from earlier measurements of the Doppler spectrometry [14] at similar experimental conditions (\( I = 4 \times 10^{18} \text{ W/cm}^2 \)), we have calculated the contribution of Doppler phase in total phase shift (Fig. 5). As observed, for time-delays less than 4 ps, the phase shift is mainly due to the ionization effects and motion of fast electron cloud. It is only after 4 ps, contribution of doppler phase starts increasing and then becomes dominant at larger time delays.

Assuming an exponential density profile \( n_e = \frac{n_0}{1+\exp(z/l_s)} \), we calculated the length scale \( (l_s) \) of the plasma (along target normal) from the measured phase shifts using Eq. (1).

4 Conclusions

We have investigated the dynamics of solid density pre-plasma created by ultra high-contrast intense laser pulses using spectral interferometry. We measured pre-plasma scale length with nanometer spatial resolution and on the sub-ps time scale. This technique is very sensitive and can measure even the tiniest phase changes occurring due to the ionization. So, by appropriate modeling/simulations of pre-plasma conditions, it, therefore, is possible to map out the picosecond pedestal of ultra-high contrast, high-intensity pulses by measuring the phase changes at times before the main pulse hits (negative delays). This method may offer an advantage over other non-linear techniques [17, 18] used to measure ps-contrast, which are sensitive to the wavelength and struggle to measure contrast better than \( 10^{-10} \).

Funding. GRK acknowledges partial support from J.C. Bose Fellowship grant (JBR/2020/000039) from the Science and Engineering Board (SERB), Government of India.

Acknowledgement. We thank Sunil B Shetye for his help in setting up vibration-free interferometer.

References

[1] M. M. Murnane, H. C. Kapteyn, M. D. Rosen, and R. W. Falcone, “Ultrafast x-ray pulses from laser-produced plasmas,” Science, vol. 251, no. 4993, pp. 531–536, 1991.

[2] P. Blanc, P. Audebert, F. Falliès, J. P. Geindre, J. C. Gauthier, A. Dos Santos, A. Mysyrowicz, and A. Antonetti, “Phase dynamics of reflected probe pulses from sub-100-fs laser-produced plasmas,” Journal of the Optical Society of America B, vol. 13, no. 1, p. 118, 1996.

[3] O. L. Landen, D. G. Stearns, and E. M. Campbell, “Measurement of the expansion of picosecond laser-produced plasmas using resonance absorption profile spectroscopy,” Physical Review Letters, vol. 63, no. 14, pp. 1475–1478, 1989.
[4] Y. Ping, R. Shepherd, B. F. Lasinski, M. Tabak, H. Chen, H. K. Chung, K. B. Fournier, S. B. Hansen, A. Kemp, D. A. Liedahl, K. Widmann, S. C. Wilks, W. Rozmus, and M. Sherlock, “Absorption of Short Laser Pulses on Solid Targets in the Ultrarelativistic Regime,” Physical Review Letters, vol. 100, p. 085004, Feb. 2008.

[5] A. G. MacPhee, L. Divol, A. J. Kemp, K. U. Akli, F. N. Beg, C. D. Chen, H. Chen, D. S. Hey, R. J. Fedosejevs, R. R. Freeman, M. Henesian, M. H. Key, S. Le Pape, A. Link, T. Ma, A. J. Mackinnon, V. M. Ovchinnikov, P. K. Patel, T. W. Phillips, R. B. Stephens, M. Tabak, R. Town, Y. Y. Tsui, L. D. Van Woerkom, M. S. Wei, and S. C. Wilks, “Limitation on Prepulse Level for Cone-Guided Fast-Ignition Inertial Confinement Fusion,” Physical Review Letters, vol. 104, p. 055002, Feb. 2010.

[6] J. Peebles, M. S. Wei, A. V. Arekiev, C. McGuffey, R. B. Stephens, W. Theobald, D. Haberberger, L. C. Jarrott, A. Link, H. Chen, H. S. McLean, A. Sorokovikova, S. Krasheninnikov, and F. N. Beg, “Investigation of laser pulse length and pre-plasma scale length impact on hot electron generation on OMEGA-EP,” New Journal of Physics, vol. 19, no. 2, 2017.

[7] M. I. Santala, M. Zepf, I. Watts, F. N. Beg, E. Clark, M. Tatarakis, K. Krushelnick, A. E. Dangor, T. McCanny, I. Spencer, R. P. Singhal, K. W. Ledingham, S. C. Wilks, A. C. Machacek, J. S. Wark, R. Allott, R. J. Clarke, and P. A. Norreys, “Effect of the plasma density scale length on the direction of fast electrons in relativistic laser-solid interactions,” Physical Review Letters, vol. 84, no. 7, pp. 1459–1462, 2000.

[8] V. M. Ovchinnikov, D. W. Schumacher, M. McMahon, E. A. Chowdhury, C. D. Chen, A. Morace, and R. R. Freeman, “Effects of preplasma scale length and laser intensity on the divergence of laser-generated hot electrons,” Physical Review Letters, vol. 110, no. 6, pp. 1–4, 2013.

[9] S. Kahaly, S. Monchocé, H. Vincenti, T. Dzelzainsis, B. Dromey, M. Zepf, P. Martin, and F. Quéré, “Direct Observation of Density-Gradient Effects in Harmonic Generation from Plasma Mirrors,” Physical Review Letters, vol. 110, p. 175001, Apr. 2013.

[10] L. A. Gizzi, E. Boella, L. Labate, F. Baffigi, P. J. Billo, F. Brandi, G. Cristoforetti, A. Fazzi, L. Fulgentini, D. Gove, P. Koester, D. Palla, and P. Tomassini, “Enhanced laser-driven proton acceleration via improved fast electron heating in a controlled preplasma,” Scientific Reports, vol. 11, Jun. 2021.

[11] R. Benattar, J. P. Geindre, P. Audebert, and J. C. Gauthier, “Optical probing of a plasma created by a 100 femtosecond laser,” Optics Communications, vol. 88, no. 4–6, pp. 376–380, 1992.

[12] I. Dey, A. Adak, P. K. Singh, M. Shaikh, G. Chatterjee, D. Sarkar, A. D. Lad, and G. R. Kumar, “Intense femtosecond laser driven collimated fast electron transport in a dielectric medium—role of intensity contrast,” Optics Express, vol. 24, no. 25, p. 28419, 2016.

[13] K. Adumi, K. A. Tanaka, T. Matsuoka, T. Kurahashi, T. Yabuuchi, Y. Kitagawa, R. Kodama, K. Sawai, K. Suzuki, K. Okabe, T. Sera, T. Norimatsu, and Y. Izawa, “Characterization of preplasma produced by an ultrahigh intensity laser system,” Physics of Plasmas, vol. 11, no. 8, pp. 3721–3725, 2004.

[14] K. Jana, A. D. Lad, M. Shaikh, V. R. Kumar, D. Sarkar, Y. M. Ved, J. Pasley, A. P. Robinson, and G. R. Kumar, “Generation of a strong reverse shock wave in the interaction of a high-contrast high-intensity femtosecond laser pulse with a silicon target,” Applied Physics Letters, vol. 114, no. 25, 2019.

[15] J. C. Kieffer, P. Audebert, M. Chaker, J. P. Matte, H. Pépin, T. W. Johnston, P. Maine, D. Meyerhofer, J. Delettrez, D. Strickland, P. Bado, and G. Mourou, “Short-pulse laser absorption in very steep plasma density gradients,” Physical Review Letters, vol. 62, no. 7, pp. 760–763, 1989.

[16] J.-P. Geindre, P. Audebert, S. Rebibo, and J.-C. Gauthier, “Single-shot spectral interferometry with chirped pulses,” Optics Communications, vol. 26, no. 20, p. 1612, 2001.

[17] R. C. Eckardt and C. H. Lee, “Optical third harmonic measurements of subpicosecond light pulses,” Applied Physics Letters, vol. 15, no. 12, pp. 425–427, 1969.

[18] G. Albrecht, A. Antonetti, and G. Mourou, “Temporal shape analysis of Nd<sup>3+</sup>: YAG active passive modelocked pulses,” Optics Communications, vol. 40, no. 1, pp. 59 – 62, 1981.

[19] C. Iaconis and I. A. Walmsley, “Spectral phase interferometry for direct electric-field reconstruction of ultrashort optical pulses,” Opt. Lett., vol. 23, pp. 792–794, May 1998.

[20] D. J. Kane and R. Trebino, “Characterization of arbitrary femtosecond pulses using frequency-resolved optical gating,” IEEE Journal of Quantum Electronics, vol. 29, no. 2, pp. 571–579, 1993.

[21] E. Tokunaga, A. Terasakiy, and T. Kobayashi, “Femtosecond phase spectroscopy by use of frequency-domain interference,” Journal of the Optical Society of America B, vol. 12, no. 5, p. 753, 1995.

[22] M. Takeda, H. Ina, and S. Kobayashi, “Computer-Based Topography and Interferometry,” J. Opt. Soc. Am. A, vol. 72, no. 1, pp. 156–160, 1982.

[23] P. Antici, J. Fuchs, M. Borghesi, L. Gremillet, T. Grismayer, Y. Sentoku, E. D’Humières, C. A. Cecchetti, A. Mančić, A. C. Pipahl, T. Toncian, O. Willi, P. Mora, and P. Audebert, “Hot and Cold Electron Dynamics Following High-Intensity Laser Matter Interaction,” Physical Review Letters, vol. 101, p. 105004, Sep 2008.
The interference signal with pump excitation for each pump-probe delay ($\tau$) can be expressed as $[21]$:

$$I'(\omega) = \left|E_{pr}(\omega)\right|^2 \left(1 + R + 2\sqrt{R} \cos(\omega T - \Delta\phi(\omega))\right)$$

(4)

where $\Delta\phi = n_r \omega L / c$ is the phase change due to the plasma, $R = \exp\left(-2n_{img}\omega L / c\right)$ is the reflection coefficient, $L$ is the length scale of the plasma and $n_r, n_{img}$ are real and imaginary part of refractive index of plasma respectively.

To extract the phase change ($\Delta\phi$), we take the Fast-Fourier transform (FFT) of the measured spectrum (4), which gives the peaks centred at $0, \pm T$ which is given by expression

$$F[I'(\omega)](t') = (1 + R) G(t') + \sqrt{R} \exp(i\Delta\phi) G(t' - T) + \sqrt{R} \exp(-i\Delta\phi) G(t' + T)$$

(5)

where $G(t')$ is the Fourier transform of $\left|E_{pr}(\omega)\right|^2$. Each of the two peaks at $t' = \pm T$ contains the information about phase change ($\Delta\phi$) as well as reflection coefficient ($R$). So we filter out right side peak ($t' = T$) and do the inverse FFT to get

$$I'(\omega) = \left|E_{pr}(\omega)\right|^2 \sqrt{R} \exp(i(\omega T + \Delta\phi))$$

(6)

A similar analysis of reference interferogram (without pump excitation) will give

$$I(\omega) = \left|E_{pr}(\omega)\right|^2 \exp(i\omega T)$$

(7)

Now by just dividing equation (6) by equation (7), we get: $P(\omega) = \sqrt{R} \exp(i\Delta\phi)$.

The imaginary part of logarithm of $P(\omega)$ gives the required phase shift ($\Delta\phi$). The phase so obtained is indeterminate to a factor of $2\pi$. Also, a computer-generated function subroutine gives a principal value ranging from $-\pi$ to $\pi$. These discontinuities in the phase can be corrected by a simple algorithm of phase unwrapping $[22]$. 

---

[21] M. Cerchez, M. Swantusch, M. Toncian, X. M. Zhu, R. Prasad, T. Toncian, C. Rödel, O. Jäckel, G. G. Paulus, A. A. Andreev, and O. Willi, “Enhanced energy absorption of high intensity laser pulses by targets of modulated surface,” Applied Physics Letters, vol. 112, may 2018.