

Impulsive excitation of a solid by extreme contrast, high-intensity femtosecond laser pulses

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(Received 18 August 2025; accepted 31 January 2026; published 6 April 2026)

We present the ultrafast dynamics of the interaction between a high-intensity extreme contrast (expected to be around $\sim 10^{-18}$ at hundreds of picoseconds timescale) femtosecond laser and a solid. Simultaneous measurements of probe Doppler spectrometry and reflectivity in pump-probe experiments reveal the presence of extreme pressure in the solid density region, which triggers a long-lived (~ 15 ps) strong inward shock. Hydrodynamic simulations accurately replicate these observations, providing a detailed explanation of the underlying physics.

DOI: [10.1103/dtc5-ftnd](https://doi.org/10.1103/dtc5-ftnd)

I. INTRODUCTION

Studies of high-intensity, ultrahigh contrast [1–3], ultrashort laser interactions with solid targets offer exciting fundamental science and promise applications in particle acceleration, hard x-ray sources, nuclear fusion, and laboratory astrophysics [4–16]. The extreme rapidity of such interactions poses a challenge for the spatiotemporal study of the interaction region. Experimental probing of highly excited matter has utilized various techniques, using the pump-probe method in plasma reflectometry and Doppler spectrometry [17–22], polarimetry [23,24], and spectral interferometry [25,26]. These techniques have been employed from the femtosecond scale to tens of picoseconds, concomitant with particle-in-cell and hydrodynamic simulations. These studies have revealed shock waves induced by the intense pressure ($> 10^{12}$ Pa) generated during the interaction [19].

A major concern in the experiments has been the intensity contrast, which refers to the intensity in the low-intensity wings of the amplified laser pulses in relation to the peak intensity. For a decade or so, high-intensity lasers have provided contrast levels of 10^{-10} . More recently, the use of plasma mirrors and pulse cleaning techniques has improved the contrast by another two to three orders of magnitude [27–31].

It is essential to further improve the contrast, as the presently achievable and projected peak intensities are reaching highly superrelativistic levels. Second-harmonic generation (SHG) is a straightforward and efficient method to achieve the square of the contrast of the fundamental pulse (subject to practical limitations in implementation and performance), as suggested long ago [32] and utilized in recent studies [33–36]. However, it has been discovered that the pulse width of a second-harmonic beam strongly depends on the input energy on the SHG crystal and reduces as the energy of the fundamental wavelength increases [37]. In our case, the pulse duration is in the 100–200 fs range, depending on the intensity.

The interaction of extreme contrast pulses with solids, that is, the impulsive excitation of a high-density surface, is a highly intriguing phenomenon and key fundamental issues surrounding this interaction still remain unknown and unexplored. In this article, we delve into the dynamics of this interaction at femtosecond resolution using pump-probe reflectometry and Doppler spectrometry. We track the plasma dynamics to tens of picoseconds, demonstrating strong inward motion of the plasma density. We also observe an intriguing anticorrelation between the time variation of plasma reflectivity and the Doppler shift. Hydrodynamic simulations explain the observations and offer insights into the dynamics.

II. EXPERIMENTAL METHODS

A. Setup

The Tata Institute of Fundamental Research in Mumbai houses a Ti:sapphire laser with a maximum peak power of 150 TW (Amplitude, France). This laser has a pulse width of 27 fs and a central wavelength of 800 nm. It exhibits an intensity contrast of 10^{-9} at 100 ps, measured using a third-order

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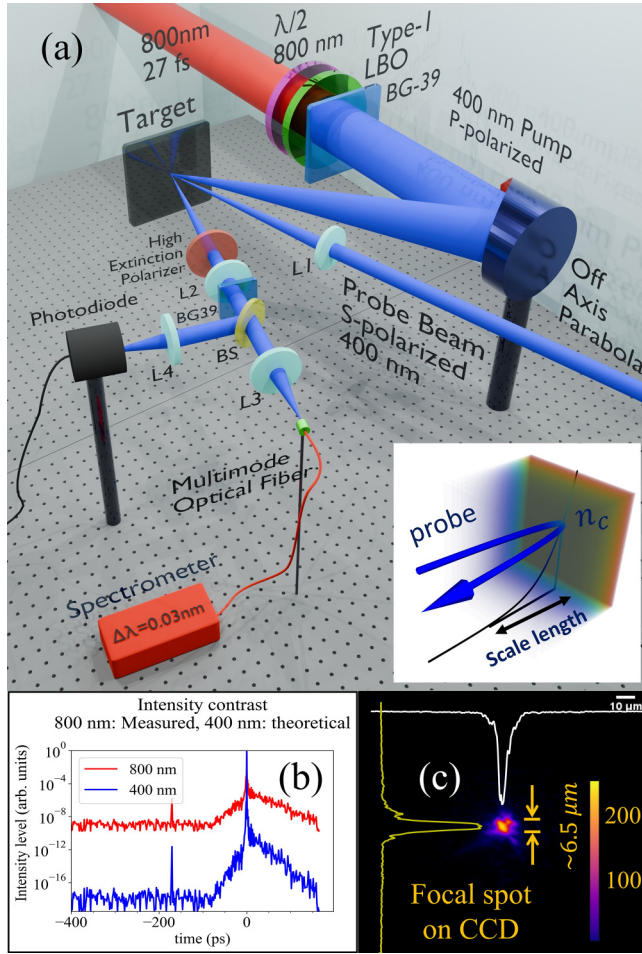


FIG. 1. (a) Schematic of the experimental setup. In the actual setup, there are three 400 nm dichroic mirrors used (along with the BG-39 filter) after the LBO to remove unconverted 800 nm. A high-extinction polarizer (Nicol prism) is placed before the diagnostics to pass only the reflected probe. (b) Measured contrast profile for the 800 nm fundamental beam and the theoretical contrast of 400 nm SHG beam. (c) The optimized focal spot of 400 nm pump beam with full width at half maximum (FWHM) of $\sim 6.5 \mu\text{m}$ as captured by charge-coupled device (CCD) camera.

cross-correlation device (SEQUOIA, Amplitude). In the experiment depicted in Fig. 1, the laser pulse is split into a strong pump pulse and a weak probe pulse. The pump path utilizes an antireflection coated half-wave plate followed by a 2 mm thick type-1 lithium triborate (LBO) for conversion of the 800 nm fundamental beam to a 400 nm *p*-polarized SHG [36].

To optimize the conversion and polarization of the pump within the vacuum, the LBO is mounted on a dual angle control stage, which allows for adjustment of the incident angle θ and the transverse plane rotation angle ϕ . The LBO output is reflected by three 400 nm high-reflectivity mirrors, which efficiently attenuate the unconverted 800 nm beam with an efficiency of $\frac{I_{800\text{ on-target}}}{I_{800\text{ unconverted}}} < 10^{-5}$. An additional thin BG-39 filter is employed to block the remaining unconverted 800 nm beam. An off-axis $f/3$ dielectric parabolic focusing mirror ($R > 99\%$) is used to focus the beam on a BK7 glass target ($50\text{ mm} \times 50\text{ mm} \times 3\text{ mm}$). A vacuum-compatible energy meter, mounted on a translational stage, can move in and out

of the beam path for measuring the energy on target. This energy meter has also been used to optimize the compressor grating position (to get shortest 800 nm near infrared (NIR) pulse) by maximizing the 400 nm second-harmonic energy. This ensures maximum SHG energy on the target but the SHG pulse width is subject to dispersion in the 2 mm thick LBO crystal and the BG-39 filter used to eliminate the unconverted 800 nm beam. The measured on-target energy (for maximized SHG) exhibits a fluctuation of $\sim 5\%$. The measured contrast of 800 nm and the theoretically expected contrast of 400 nm are illustrated in Fig. 1(b). The pump focal spot is measured to have a diameter of $\sim 6.5 \mu\text{m}$ [Fig. 1(c)] at the target position.

The probe is converted to the second harmonic (400 nm *s*-polarized) using a 2 mm thick β -barium borate crystal and reflected from a retroreflector mounted on a linear stage to control the delay between the pump and probe pulses. The unconverted 800 nm beam in the probe arm is blocked by a BG-39 bandpass filter. The probe is focused on the target using a 15 cm planoconvex lens. The spatial and temporal matching of the two pulses is verified using standard procedures [18,19,22]. The details of the setup are given in the Supplemental Material [38]. Due to dispersion in the BG-39 filter, focusing lens (and also the chamber window) the probe pulse width can increase from 170 fs (assuming it to be transform limited during SHG process) to around 171 fs. This dispersion is not significant and has minimal effect on the measurements.

We may note here that the high intensity and femtosecond duration of the laser ionize the target very rapidly on the rising edge of the laser pulse, essentially turning it into a dense plasma. Nonlinear optical effects that arise at much lower intensities are overwhelmed right on the very early rising edge of the high-intensity pulse and do not affect our measurements. After each shot, the focal spot position on the target is damaged, and hence the target is moved (rastered) to a new position to take the next shot.

B. Methodology

The probe reflects from the evolving critical surface or a high-density hump on a lower-density base of plasma for 400 nm wavelength ($n_{\text{cr}} = \omega_L^2 m_e \epsilon_0 c^2 / e^2 \approx 6.97 \times 10^{21} \text{ cm}^{-3}$) [17,22,39]. The probe is collected with an achromatic lens of focal length 15 cm, divided with a $200 \mu\text{m}$ thick fused silica beam splitter, and one part is sent to a photodiode connected to an oscilloscope and the other to a high-resolution triggerable spectrometer (HR-4000, resolution of 0.03 nm). A high-extinction (extinction ratio $\sim 10^{-3}$) Nicol prism polarizer and a BG-39 filter are placed before the beam splitter to transmit only *s*-polarized beam and minimize noise from 800 nm fundamental beam and 400 nm LBO converted *p*-polarized pump beam.

The reflected probe suffers a Doppler shift due to the motion of the pump excited surface. The shift is given by [40]

$$\nu_{\text{observed}} = \nu_{\text{source}} \frac{1 + \frac{2v}{c} \cos \alpha + \frac{v^2}{c^2}}{1 - \frac{v^2}{c^2}}, \quad (1)$$

where α is the incident angle of the probe on the target. As the probe incidence angle is very small, we can assume $\cos \alpha \approx 1$

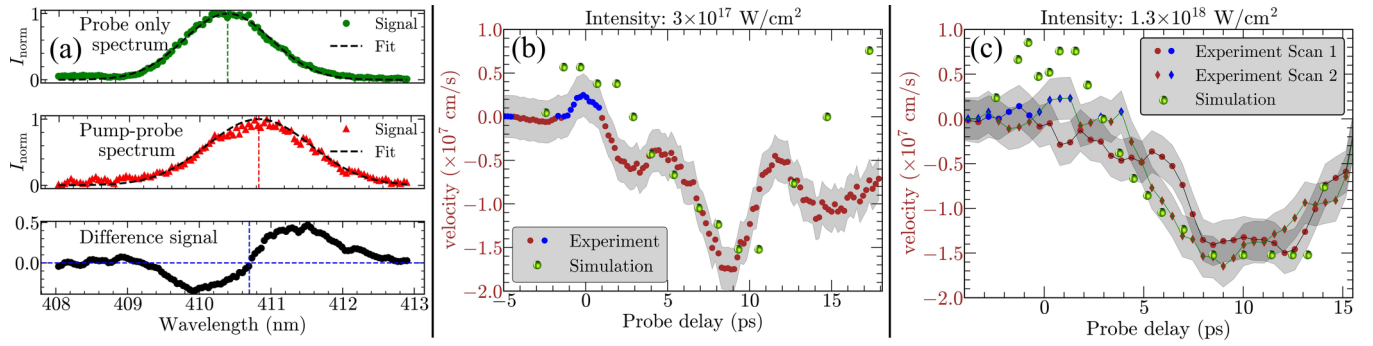


FIG. 2. (a) The measured raw (normalized with respect to peak) spectrum for probe only signal and pump-probe signal (for $I_{\text{peak}} = 1.3 \times 10^{18}$ W cm⁻² and delay of 12 ps) and the difference of pump-probe signal to probe only signal. The measured velocity of 400 nm critical surface ($n_{\text{cr}} \sim 6.97 \times 10^{21}$ cm⁻³ for $\lambda = 400$ nm) has been shown for different intensities: (b) 3×10^{17} W cm⁻² and (c) 1.3×10^{18} W cm⁻².

in Eq. (1) and approximate to

$$v = -0.5c \frac{\Delta\lambda}{\lambda}, \quad (2)$$

with $\Delta\lambda = (\lambda - \lambda_0)$ and $\lambda_0 = c/v_{\text{source}}$ [41]. Here, the negative sign indicates that the plasma is moving into the target. We determine λ_0 as the central wavelength of the Gaussian-fitted input probe spectrum (averaged over 40–50 shots). For a given delay, λ represents the central wavelength of the Gaussian-fitted reflected probe signal [Fig. 2(a)]. The standard deviation of the input probe signal is ≈ 0.064 nm, which is used to calculate the error bar for the pump-probe shots.

III. RESULTS AND DISCUSSION

A. Time-resolved Doppler spectrometry

Figures 2(b) and 2(c) show the temporal evolution of the critical surface motion for intensities of 3×10^{17} and 1.3×10^{18} W cm⁻², with pulse widths of 186 and 100 fs, respectively. Note that the converted SHG pulse duration depends on the input energy on the LBO, as established by Aparajit *et al.* [37]. In the intensity calculation, we have taken the fundamental input energy-dependent SHG pulse width.

The red and blue colors indicate the critical surface moving inward (redshift) and outward (blueshift) from the target surface, respectively. The error bars represent the shot-to-shot fluctuations in the central wavelength, as indicated by the gray bands in the plots. A small outward motion can be observed for each intensity after the interaction with the main pulse, followed by a long inward motion. The reflected probe signal beyond 15 ps is weak for detection; however, the trend of reducing redshift is consistent with the prediction from simulations, as discussed later.

These experimental measurements show that the near-solid density matter receives an extreme impulse in a very short time due to the ultrahigh contrast of the pulse that pushes matter inside for a long time. Earlier wave-front measurements at shorter timescales show a similar inward motion of the plasma critical surface [42,43].

It is interesting to see that the Doppler shift for 3×10^{17} W cm⁻² [Fig. 2(b)] shows an oscillatory behavior. Fourier analysis of the Doppler shift signal shows a peak at around $\nu = 180$ GHz in the frequency domain. The corresponding temporal periodicity comes out to be 5.6 ps. The

physical reasons for these oscillations need further thought and study and we propose to investigate them in the near future.

B. Simultaneous measurement of plasma reflectivity

We can also estimate the velocity of the critical surface by analyzing the simultaneously measured reflectivity of the plasma. As shown in Fig. 3(a), the reflectivity (at $I_{\text{peak}} \approx 3 \times 10^{17}$ W cm⁻²) increases when the pump interacts with the solid and creates plasma. However, at later delays, the plasma expands significantly with increased energy absorption, leading to a reduction in reflectivity. The drop in reflectivity can be fitted with an exponential curve:

$$R_{\text{norm}}(t) = \exp\left(-\frac{t - t_{\text{peak}}}{\tau}\right),$$

where t_{peak} is the delay at which plasma reflectivity reaches its maximum. The exponential decay timescale is measured to be $\tau = 2.04 \pm 0.34$ ps.

The plasma absorption depends on the scale length of the underdense preplasma, which in turn depends on the velocity of the critical surface, i.e., $L(t) \sim \langle v_{\text{crit}}(t') \rangle_{t'} \times t$, where $\langle v_{\text{crit}}(t') \rangle_{t'}$ is the time-averaged velocity for the critical surface from the time of peak reflectivity up to time delay t . Assuming the plasma density has a profile of $n_e(x) \sim n_{\text{cr}} e^{-x/L}$, we can estimate the velocity from the relation of normalized reflectivity to the plasma scale length as [5,44]

$$R_{\text{norm}}(t) = \exp\left(-\frac{8v_{ei}L(t)}{3c}\right), \quad (3)$$

with the electron-ion collision frequency

$$v_{ei}(T_e, n_i) \approx 2.9 \times 10^{-6} \ln \Lambda \frac{\bar{Z}^2 n_i [\text{in cm}^{-3}]}{(T_e [\text{in eV}])^{3/2}} [\text{s}^{-1}].$$

where $n_i = n_e/\bar{Z}$ is the average ion density, $\bar{Z}$ is the average ionization state over different species inside the plasma, T_e is the plasma electron temperature, and $\ln \Lambda$ is the Coulomb logarithm ($\Lambda = b_{\text{max}}/b_{\text{min}}$, where b_{max} and b_{min} are on the order of plasma Debye length and closest approach of free electron to ion). By taking $\bar{Z} \approx 5$, $T_e \approx 135$ eV, and $\ln \Lambda \sim 10$ [45], we obtain the average velocity of the critical surface of $\sim 8.55 \times 10^6$ cm s⁻¹. Meanwhile, the average velocity up to 10 ps, calculated from the Doppler spectrometry measurement

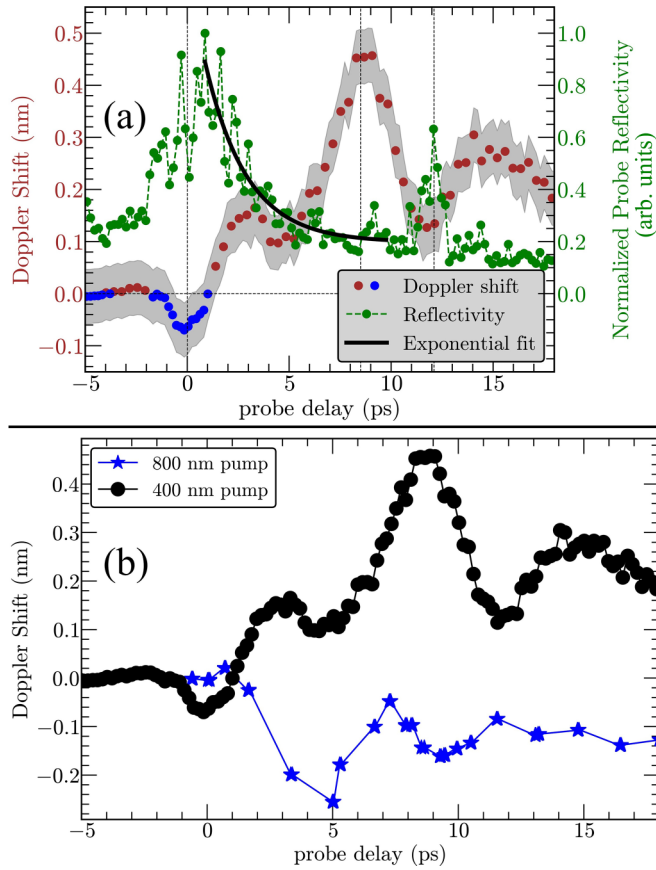


FIG. 3. (a) Simultaneously measured probe reflectivity and Doppler shift for different pump-probe delays. The anticorrelations between reflectivity and Doppler shift are indicated by vertical dotted lines. The black and purple solid lines show the exponential fitting for reflectivity as $R_{\text{normalized}}(t) = e^{-\frac{t-t_{\text{peak}}}{\tau}}$, where τ is the time decay constant for reflectivity. In our case, we get $\tau = 2.04 \pm 0.34$ ps for the first reflectivity peak (black fit). (b) Comparison of Doppler shifts for extreme contrast 400 nm current data vs moderately high contrast 800 nm earlier data (by Jana *et al.* [19]).

for $I_0 = 3 \times 10^{17} \text{ W cm}^{-2}$ [Fig. 2(b)], is $7.43 \times 10^6 \text{ cm s}^{-1}$. The two different approaches agree very well.

We note that the plasma reflectivity and Doppler shift signals are anticorrelated [Fig. 3(a)], somewhat reminiscent of an earlier study [46]. When the redshift increases (or blueshift reduces), the plasma reflectivity reduces, and when blueshift increases (or redshift reduces), the reflectivity increases. This is because during increasing redshift, the critical surface moves inward, effectively increasing the propagation length through plasma, and vice versa. The earlier study, which invoked ultrafast hydrodynamics, pointed out the possibility of density profile modifications to explain the behavior. In our case, the second reflectivity peak around a delay of 12 ps shows a sharp dip in the Doppler shift. However, the full understanding about the secondary peak needs a detailed study and will be pursued in future.

C. Comparison with earlier results

In addition, we also compare the present study performed with the 400 nm SHG pump beam, with the fundamental

800 nm pump beam (contrast $\sim 10^{-9}$ at 100 ps) [19] in Fig. 3(b). The motion of the critical density front is completely opposite for the 800 nm pulses. This implies that the prepulse of the fundamental pump with a lower contrast has caused a significant plasma expansion toward outer direction (showing strong reverse shock), and the inward compression wave could not be detected experimentally. There is, however, an interesting correlation in the oscillatory period in both cases during plasma motion after the pump pulse leaves.

It is important to recognize that the primary objective of the comparison is to show how the contrast level influences shock generation on a timescale of a tenth of a picosecond. Notably, the higher-intensity fundamental laser pulse exhibits lower contrast compared to the second-harmonic pulse, resulting in the generation of a longer preplasma. The effects observed are readily reversed when comparing the second-harmonic laser pulse depicted in Figs. 2(b) and 2(c).

D. Simulations

To understand the observations and get further insights into the dynamics, we performed one-dimensional (1D) hydrodynamic simulations FLASH code version 4.8 [47]. We use the three-temperature treatment for plasma, tabulated equation of state (EoS), flux-limited diffusion model for electron conduction, heat exchange, multigroup opacity, and radiation transport (see the Supplemental Material for simulation details [38]). The plasma pressure is obtained from IONMIX4 tabulated EoS library [48]. Note that the FWHM pulse duration of 186 fs is assumed for $I_0 = 3 \times 10^{17} \text{ W cm}^{-2}$.

The laser energy deposition proceeds via inverse bremsstrahlung, while the resonance absorption ceases to be effective for steep density gradients [49]. In this situation, vacuum heating dominates and the absorption is $\geq 80\%$ for $I_0 \lambda^2 \geq 10^{17} \text{ W cm}^{-2} \mu\text{m}^2$ for incident angle $\theta = 45^\circ - 50^\circ$ [50–52]. However, the condition of this experiment is $I_0 \lambda^2 \leq 10^{17} \text{ W cm}^{-2} \mu\text{m}^2$, and we expect insignificant vacuum heating (resonance absorption is also negligible given the ultrasteep density profile). Therefore, hydrodynamic simulation is sufficient to predict the shock velocities for our intensities. The laser absorption and shock generation can be described in a few stages, depending on the laser intensity profile. Note that the peak of the pulse is located at 360 fs from the origin of the scale.

Due to the extreme contrast of the second-harmonic laser, the laser pedestal at $t \leq -5$ ps does not result in significant preplasma generation. Therefore, we only model the interaction of the main pulse with a steep density gradient target, which corresponds to time delay $t > -2$ ps. The interactions between $-5 \text{ ps} < t < -2 \text{ ps}$ can be described as follows.

Assuming the physical properties of BK7 glass such as density and ionization energy are similar to fused silica, the damage threshold for 400 nm, < 300 fs pulses is around 1 J cm^{-2} [53]. The corresponding intensity for pulse duration of 186 fs is around $5.38 \times 10^{12} \text{ W cm}^{-2}$. The maximum intensity used in our experiment is $1.3 \times 10^{18} \text{ W cm}^{-2}$, several orders of magnitude higher than the damage threshold. The rising edge of the extreme contrast SHG laser pulse has intensities around $10^{12} \text{ W cm}^{-2}$, sufficient to generate plasma on the glass surface. The reflectivity of the plasma is estimated

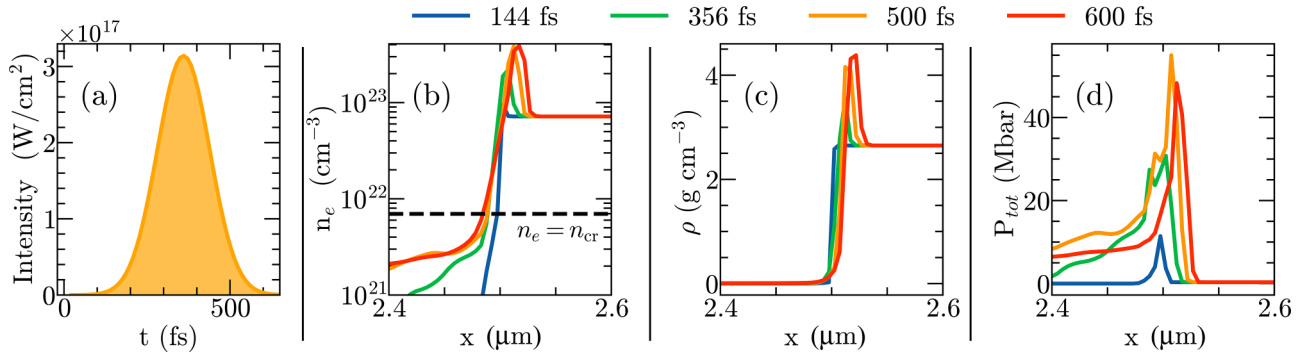


FIG. 4. Simulated temporal evolution of (b) electron number density, (c) mass density, and (d) total pressure for incident intensity of $3 \times 10^{17} \text{ W cm}^{-2}$. The temporal profile is shown in panel (a) with FWHM pulse duration of 186 fs. The simulation is performed using a 1D hydrodynamic code (Flash).

to be $R \approx 0.23$ (see the Supplemental Material for the estimation of reflectivity [38,53]), consistent with the measurement shown in Fig. 3(a) between $-5 \text{ ps} < t < -2 \text{ ps}$. Within this time range, the plasma density scale length is in the limit $L \rightarrow 0$ and the laser absorption on the rising edge of the pulse can be described by normal skin effect. The reflectivity follows from the Fresnel equations [6,54].

At $t > -2 \text{ ps}$, the ablation begins, and the plasma expands outward with density scale length $L = c_s t$, where $c_s = (\gamma Z k_B T_e / m_i)^{1/2}$ is the ion acoustic velocity, where γ is the heat capacity ratio. The subsequent absorption of the laser light is described by inverse bremsstrahlung. Assuming a fully ionized triatomic gas with $\gamma = 1.33$ and $T_e \geq 2.4 \text{ eV}$, the plasma expands outward with velocity $\geq 1.2 \times 10^6 \text{ cm s}^{-1}$. The outward expansion velocity is shown in Figs. 2(b) and 2(c) in blue points.

Figure 4 presents the time evolution of plasma properties during the interactions with the main pulse for an intensity of $3 \times 10^{17} \text{ W cm}^{-2}$. At $t \approx 144 \text{ fs}$, the laser intensity at rising edge is on the order of $10^{16} \text{ W cm}^{-2}$ as shown in Fig. 4(a). However, plasma expansion does not produce an ablation pressure sufficient to generate a shock. This is evident in Figs. 4(b) and 4(c), where we only observe the compression of the electron density at $t \approx 144 \text{ fs}$. The maximum total plasma pressure is only approximately 10 Mbar as depicted in Fig. 4(d). We note that the radiation emission pressure from the plasma is negligible at this time.

At $t \approx 356 \text{ fs}$, the peak of the pulse reached the target surface. The total pressure at this moment reached 30 Mbar. At $t \approx 500 \text{ fs}$ from the origin, the laser pulse is reflected and exits the target surface with a maximum pressure of 50 Mbar. The pressure begins to decrease at the descending edge ($t \approx 600 \text{ fs}$). After this, the laser has completely left the target, while the shock continues to propagate into the target. The electron shock front brings the critical density surface inward. This results in the redshift beyond $t \geq 1 \text{ ps}$ as shown in Figs. 2(b) and 2(c) in red points. The predictions of the velocity of the critical density surface using solely hydrodynamic simulation are consistent with the experimental measurements. This implies that the shock generation is purely hydrodynamic nature.

The shock generation can be attributed to the impulsive excitation by the radiation pressure of an extreme contrast short-pulse laser on the steep density gradient target. The

net radiation pressure on a target can be expressed as $P_{\text{rad}} = (1 + R)I_0/c$, where R is the reflectivity of the target. The first term represents the pressure exerted by the incident wave, while the second term accounts for the pressure exerted by the reflected wave. This also explains why the plasma pressure peaks during reflection at $t \approx 500 \text{ fs}$. The net radiation pressure is $P_{\text{rad}} = 200 \text{ Mbar}$ for $I_0 = 3 \times 10^{17} \text{ W cm}^{-2}$ and $R \sim 1$.

The radiation pressure exerts a force $F = P_{\text{rad}}A$ on the skin depth across the focus spot area A . The acceleration is then $a = F/m = P_{\text{rad}}A/(\rho A \delta) = P_{\text{rad}}/(\rho \delta)$. The maximum velocity by this impulsive force reaches $v = P_{\text{rad}} \Delta \tau / (\rho \delta) \approx 2.2 \times 10^7 \text{ cm s}^{-1}$ with fully ionized plasma skin depth $\delta = c/\omega_{pe} \approx 6.4 \text{ nm}$ and pulse duration $\Delta \tau = 186 \text{ fs}$. For $I_0 = 1.3 \times 10^{18} \text{ W cm}^{-2}$, the radiation pressure is $P_{\text{rad}} = 866 \text{ Mbar}$. The impulsive velocity is $v \approx 5.1 \times 10^7 \text{ cm s}^{-1}$ for $\Delta \tau = 100 \text{ fs}$ and $R \sim 1$.

The shock structure in both the electron and mass density profiles is almost at the same location. This eliminates the possibility of ion motion triggered by the charge separation field in the laser hole-boring (HB) regime [55]. The HB regime is characterized by ion motion driven by the charge separation field, which is absent in our scenario. Additionally, the shock structure in our case differs from the ablation-generated shock, where the shock is generated by the ablation pressure, and the ion motion is driven by the ablation front. To the best of our knowledge, this is the first time that the impulsive excitation of a solid by an extreme contrast, high-intensity, nonrelativistic femtosecond laser pulse has been studied. A key finding was that the shock generation is purely hydrodynamic, caused by the radiation pressure exerted by the laser main pulse. This contrasts with the ablation-generated shock or hole-boring regime, where the latter primarily involves the charge separation field. This represents a mechanism for shock generation in the extreme contrast regime at nonrelativistic intensities.

IV. CONCLUSION

In summary, we have presented the impulsive shock generation on solid density plasma, which is made possible with the extreme contrast of the pulse generated by a high-contrast fundamental pulse. This pressure is analogous to the motion of a solid wall under an “instantly” applied force. The inward motion of the bulk material is present way longer than the

duration of the pulse. The compression happens until the energy transferred by the impulse becomes equal to the compressional energy of the bulk high-density plasma. Additionally, the hydrodynamic nature on long picosecond timescale of the problem distinguishes the current scenario from all earlier measurements on shocks.

ACKNOWLEDGMENTS

G.R.K. acknowledges major support for this research from Department of Atomic Energy through the grant “Physics and Astronomy (Project Identification No. RTI4002), Tata Institute of Fundamental Research” and partially from the Grant No. JBR/2021/00039 of the Anusandhan National Research Foundation (ANRF), both of the Government of India. J.F.O. acknowledges EuroHPC Joint Undertaking for awarding us access to Karolina at IT4Innovations (VŠB-TU), Czechia under Project No. EHPC-REG-2025R01-007 (EU-25-87); supercomputer LUMI, hosted by CSC (Finland)

and the LUMI consortium through a EuroHPC Benchmark Access call (EHPC-BEN-2025B12-021); and Ministry of Education, Youth and Sports of the Czech Republic through the e-INFRA CZ (ID:90140). These works were partly supported by Contract No. PN23210105 funded by the Romanian Ministry of Research, Innovation and Digitalization and of the Extreme Light Infrastructure Nuclear Physics Phase II, a project cofinanced by the Romanian Government and the European Union through the European Regional Development Fund and the Competitiveness Operational Program (Grant No. 1/07.07.2016, COP, ID 1334). Additionally, partial support was given by JSPS Core-to-Core Program, Grant No. JPJSCCA20230003.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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