

Role of femtosecond prestructure of intense harmonic pulses in relativistic laser-solid interactions

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Extreme intensity contrast is essential for ultraintense, femtosecond laser excitation of solids, in particular for studies with structured or ultrathin targets. In order to maximize the contrast in the nanosecond and picosecond range, a very effective approach is the use of second-harmonic generation. In this Letter, we use intense harmonic 400-nm pulses to explore experimentally the fundamental interactions of these pulses with flat and sub- λ grating targets. We show that the presence of a femtosecond-scale structure in the rising edge of the harmonic laser pulse critically modifies the laser-solid interaction. It alters the local field structures and hence induces redirection in the electron trajectories and distributions. We show that it is of particular importance concerning resonant phenomena such as surface plasmon excitation in structured targets. Particle-in-cell simulations fully support and give further insight into our experimental results. Our findings have important implications not only for the use of harmonic pulses on solid targets but also for two-color schemes based on second-harmonic pulses.

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Rapid progress in high-power lasers and high-energy-density science has led to a multitude of applications, for example, the generation and control of high brightness, ultrafast electromagnetic and particle sources, laboratory simulation of astrophysical scenarios, application of extreme shocks to materials, etc. [1,2]. The physics of the interactions driven by such femtosecond terawatt and petawatt laser systems is highly nonlinear and complex. To explore laser-solid interactions with the cleanest relativistic laser pulse, avoiding any preplasma, experiments demand ultrahigh intensity contrast; i.e., the intensity on the picosecond and nanosecond scales preceding the femtosecond peak should fall off below the plasma creation threshold for the target. The intensity contrast crucially determines the preplasma density gradient and, in turn, affects fundamental processes like absorption [3,4], hot electron generation [5,6], high-harmonic generation [7], etc. There have been many recent studies demanding ultrahigh contrast pulses for the generation of homogeneous high-energy-density matter [8], high-harmonic generation [9,10], electron and ion acceleration from ultrathin targets [11–13], and the use of structured targets for creating hot, dense plasma conditions and hence high-flux, electromagnetic and material particle emission [14–21].

Plasma mirrors (PM) are often used for cleaning up the contrast. A two-stage version can improve the contrast by 10^4 with a 50% reflectivity [22,23]. However, the PM is damaged and a fresh surface must be supplied after each shot. Another effective approach to enhance the intensity contrast is the use of a second-harmonic (SH) pulse, the harmonic generation process expected to improve the contrast by many orders of magnitude to an “extreme” level ($\ll 10^{-12}$, the limit of the current measurement capabilities in the field) [8,14,19,24–27]. Moreover, SH pulses have crucial implications in two-color relativistic femtosecond laser schemes for applications in particle acceleration, high-harmonic generation, attosecond control, and generation of positron beams, to list a few [28–32]. Recently, it has been demonstrated [33,34] that efficient generation of SH pulses requires the nonlinear crystals to be driven at high intensities ($> 10^{12}$ W/cm²), which in turn introduces modulations to the femtosecond temporal pulse profiles. Although the exact shape depends on the nonlinear crystal and the input intensity, broadly such intense SH pulses contain femtosecond prestructures in the hundreds of femtosecond scale before the primary laser peak and can have 10–15% of the laser peak intensity. However, these are fundamentally different from the contrast structures at the nanosecond and picosecond levels, as no significant hydrodynamic expansion of the plasma can occur on such femtosecond timescales. Nevertheless, these are intense enough to seriously alter the interactions, especially with ultrathin targets (< 100 nm), structured targets, nanowire surfaces [14,19,35], and grating targets [15–17,20,21].

In parallel studies, laser interactions with grating targets have been explored and key parameters have been identified to efficiently excite surface plasma waves (SPWs) to improve

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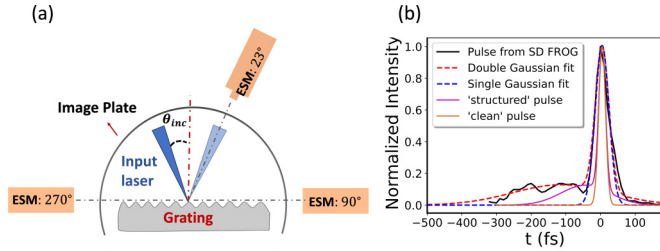


FIG. 1. (a) The experimental schematic for measuring the electron angular distributions and spectra (ESM: electron spectrometer). (b) Temporal profiles of the input laser pulse (with the femtosecond prestructure) measured using the SD-FROG setup [33], the corresponding single and double Gaussian fit temporal profiles, and scaled-down clean pulse and structured pulse with the femtosecond prestructure used in PIC simulations.

laser absorption and to assist in the generation and collimation of relativistic hot electrons [16,17,36–41]. In particular, the generation of SPWs is a resonant phenomenon and acts as a sensitive test to study the role of femtosecond scale prestructures in the fundamental interactions.

In this Letter, we report the role of femtosecond prestructures in the excitation of SPWs in sub- λ grating targets by examining their impact on laser absorption, hot electron angular distributions, and energy spectra. We present experimental data using extreme-contrast ($<10^{-12}$), second-harmonic, femtosecond, 400-nm relativistic laser pulse interactions with sub- λ gratings as well as flat (or plain) targets. Experimental results are well supported with 2D3V particle-in-cell (PIC) simulations performed using the SMILEI code [42]. Here, 2D3V means that the particles are described in a five-dimensional phase space having two coordinate and three velocity components.

The experiment [Fig. 1(a)] was conducted using a chirped-pulse-amplified 150-TW Ti:sapphire laser system delivering 800-nm, 27-fs, 50-nm bandwidth pulses at a 5-Hz repetition rate. The 800-nm pulses were up-converted inside a vacuum chamber (10^{-6} torr) to 400 nm using a 70-mm-diameter, 2-mm-thick type-I lithium triborate (LBO) second-harmonic generation (SHG) crystal (Cristal Laser) [34]. A half-wave plate was placed before the SHG crystal to ensure that the SH pulses are p-polarized. Multiple dichroic mirrors (at 400 nm) were used in series to remove the residual 800-nm components (800/400 energy ratio: $\sim 10^{-5}$). The laser was focused on the target using an f/3 dielectric off-axis parabola, and the focal spot size was $\sim 4\text{--}6\text{ }\mu\text{m}$. The electron angular distributions were measured using image plate (IP) detectors placed in front of the target to cover most of the front angles (a small hole was made in the IPs to let the laser go through). We used Al foils to allow only electrons above 100 keV to reach the IP. Three electron spectrometers characterized the electron energy spectra in three specific directions, i.e., front specular reflection, positive surface ($+90^\circ$), and negative surface ($+270^\circ$) corresponding to direct laser acceleration and electrons accelerated by the surface modes. Each spectrometer had a 0.1-T magnetic field with an IP as the detector. The measurable range of energies in these spectrometers was 0.1–7.0 MeV.

The primary target used in the experiment was a 3600 lines/mm holographic grating target (Edmund Optics) which was sub- λ for our input wavelength of 400 nm. Two important processes arise in laser interactions with a grating target. First, the structures allow a local enhancement of the electric fields in their vicinity, leading to enhanced absorption and hot-electron generation [43]. Second, they allow resonant excitation of SPWs, leading to efficient acceleration and collimation of electrons along the SPW modes. The advantage of a sub- λ grating is to reduce the number of diffraction modes and couple the laser energy to only a single resonant surface mode along the negative surface (the surface opposite to the laser direction; 270° in Fig. 1(b) [44]). The grating used in our experiment had a grating depth of ~ 100 nm and showed a resonance for θ_{inc} [angle of incidence (AOI)] of 23° ; efficient excitation of surface plasma waves occurs at this AOI. A flat BK7 target was used for comparative studies. A first set of experiments was conducted with a fully relativistic pulse at a peak laser intensity of 8×10^{18} W/cm ^{2} , resulting in a “structured” pulse [see Fig. 1(b)]. We then considered a weakly relativistic pulse at a peak intensity of 5×10^{17} W/cm ^{2} . These were generated by driving the LBO crystal at a lower intensity to produce a “clean” pulse (although with lower energy and longer pulsewidth, and hence lower intensity). (See Sec. 2 of the Supplemental Material [45] for exact pulse profiles.)

The angular distribution and the amount of ejected fast electrons differ strikingly for each target as shown in Fig. 2(a) for the fully relativistic pulse. For the case of p-polarized input laser at the resonance angle of $\theta_{\text{inc}} = 23^\circ$, we can see a threefold increase in the electron flux for the grating target as compared to the flat target, estimated by integrating the electron flux along all the directions. Moreover, the electrons are preferentially accelerated along the positive surface direction ($+90^\circ$) for the grating target. For the plain target, we see a more isotropic electron angular distribution. The s-polarized pulse interaction with the gratings shows a tenfold lower electron flux as compared with the p-polarized pulse and it also shows no preferred direction for electron acceleration. The off-resonance case of $\theta_{\text{inc}} = 32^\circ$ for the grating target [green curve in Fig. 2(a)] shows a slightly lower flux along both the surface directions of $+90^\circ$ and $+270^\circ$, indicating a lower surface coupling. The structured pulse vs clean pulse (generated at a lower intensity) angular distribution comparison is shown in Fig. 2(b). We see a clear signature of electrons accelerated along the negative surface for the clean pulse case.

The electron spectra for the grating target for the structured pulse case along three directions, i.e., positive surface ($+90^\circ$), negative surface ($+270^\circ$), and specular ($+23^\circ$) reflection, are shown in Fig. 2(c). Note that the cutoff energies and the temperatures are higher along the positive surface direction, in good agreement with the angular distributions, and indicate preferential electron acceleration along the positive surface direction. This acceleration along the positive surface is, however, not due to the SPW acceleration mechanism, as the SPW mode in a sub- λ grating target is expected along the negative surface direction [44]. The particle acceleration and heating in the upward direction can have two contributions: generation of a current along the surface due to momentum transfer because of partial laser absorption [46], and laser

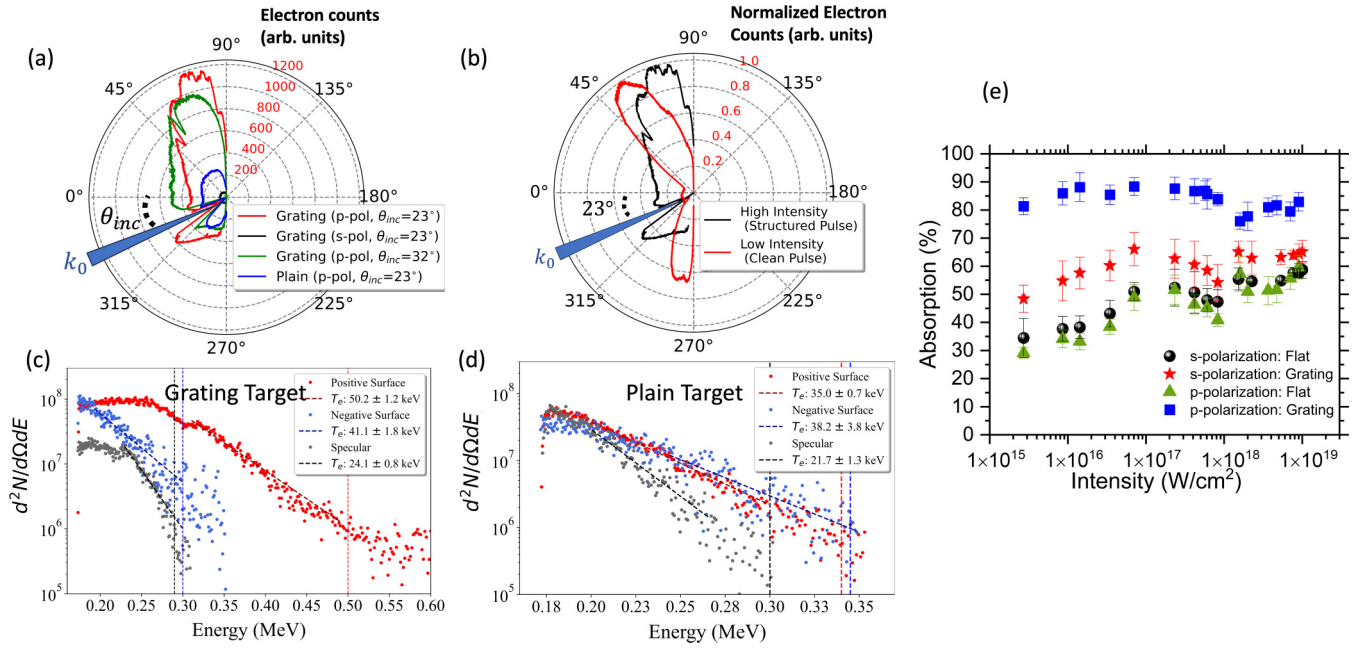


FIG. 2. (a) Electron angular distribution from experiments with a structured pulse for the grating and plain targets. (b) Hot-electron angular distribution comparison between a structured pulse (high intensity) and a clean pulse (low intensity). Experimental hot-electron spectra from the (c) grating target and the (d) plain target in three different directions, i.e., positive surface, negative surface, and specular directions. The dashed vertical lines indicate the respective cutoff energies. (e) Laser absorption data from experiments for the sub- λ grating and plain target for the cases of “p-” and “s-”-polarized input lasers.

vacuum acceleration by the reflected laser [47]. This will result in the presence of a quasistatic magnetic field (also seen in simulations, see Fig. 3) and can have a complex interplay with the particle orbits. Notice that a quasistatic magnetic field in the opposite direction also appears related to the SPW excitation [48]. The corresponding electron spectra for the plain target are shown in Fig. 2(d) where we see similar electron cutoff energies and temperatures along all three directions, again indicating a more isotropic distribution of electrons. The electron energies in our experiment are lower than other results [16] due to the combined effect of lower input intensity,

shorter wavelength, and the role of the prestructure in the suppression of SPWs.

To understand the nature of the interaction clearly, we performed laser absorption measurements in two types of targets, a flat BK-7 glass target and a sub- λ grating target, and the angle of incidence was chosen as 23° to resonantly excite surface plasma waves and enhance absorption in the latter. A detailed description of the absorption measurement is given in Sec. 1 of the Supplemental Material [45]. Figure 2(e) shows that the absorption in a flat target increases from 30 to 60% with increasing input intensity. On the other hand, the grating target shows 80% absorption over the entire intensity range of 10^{16} – 10^{19} W/cm^2 . The increase in absorption is due to a combined effect of surface plasma wave resonance and geometrical effect due to local electric field enhancement. The s-polarization absorption in the grating target is also higher than that in the flat target; this result is attributable to the geometric local electric-field enhancements in the vicinity of the structure [43].

To further understand the experimental results, 2D3V PIC simulations have been performed with the open-source code SMILEI [42]. A detailed description of the simulation geometry and parameters is presented in Sec. 4 of the Supplemental Material [45].

The laser field (B_z) profiles from the grating target are shown in Fig. 3. These are shown at the time instant right after reflection of the input laser from the grating surface, and the two cases correspond to the two temporal shapes considered in the simulations. For the $a_0 = 1.5$ case, we notice a sharp distinction between the two cases: In Fig. 3(a), the clean pulse leads to efficient excitation of the SPW mode along

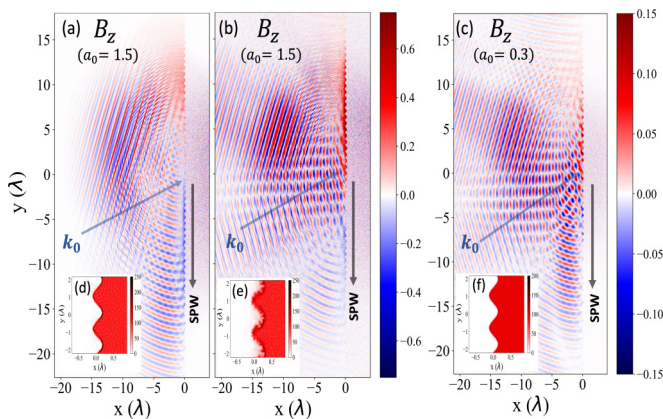


FIG. 3. B_z field profiles after reflection from the grating surface for (a) $a_0 = 1.5$, clean pulse; (b) $a_0 = 1.5$, structured pulse; and (c) $a_0 = 0.3$, structured pulse. Panels (d)–(f) show the corresponding electron densities of the grating when the laser peak hits the target.

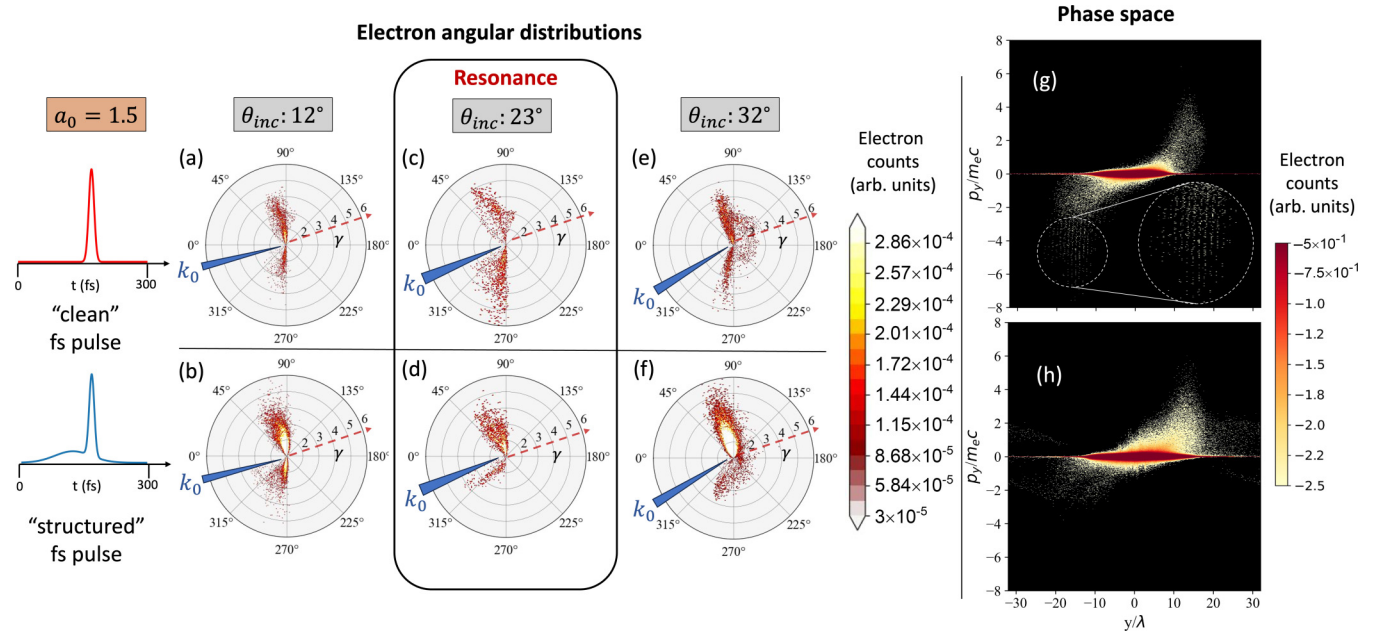


FIG. 4. Electron distributions for the grating target for $a_0 = 1.5$ from the simulations. (a) $\theta_{\text{inc}} = 23^\circ$, clean pulse; (b) $\theta_{\text{inc}} = 23^\circ$, structured pulse; (c) $\theta_{\text{inc}} = 12^\circ$, clean pulse; (d) $\theta_{\text{inc}} = 12^\circ$, structured pulse; (e) $\theta_{\text{inc}} = 32^\circ$, clean pulse; and (f) $\theta_{\text{inc}} = 32^\circ$, structured pulse, respectively. Electron phase-space curves for (g) clean and (h) structured pulses at $\theta_{\text{inc}} = 23^\circ$.

the negative y direction or negative surface (highlighted by the “SPW” arrow in the figure), whereas in Fig. 3(b), for the structured pulse, the SPW mode is strongly suppressed. We also notice a quasistatic B_z field along the positive y direction (or positive surface) in both cases; however, it is stronger for the structured pulse. In Fig. 3(c), we show the simulation results for the $a_0 = 0.3$ case, where we explored both the clean and structured pulses, finding very similar behavior. Here we show simulations for the structured case, keeping in mind that they are representative of both clean and structured pulses at this intensity. As we can see, a strong SPW mode along the negative surface has been excited. This is consistent with the shape of the electron density profiles shown in Figs. 3(d)–3(f), at the time when the laser peak reaches the surface. In Figs. 3(d) and 3(e) we have the electron density profiles for the case $a_0 = 1.5$ without and with the femtosecond prestructures. A significant alteration of the electron densities at the grating surface can be observed when the femtosecond prestructure is present, which in turn affects the field profiles, the laser absorption, and the coupling mechanisms. Note that the femtosecond structure does not generate any significant preplasma scale length, whose presence could have affected the laser energy coupling into the surface waves. However, the strong rippling effect seen in the density profiles can affect the local grating shape and the resonance conditions, and lead to an inefficient surface wave generation. Such a modification of the density profile is also seen in flat targets (see Fig. S7 in the Supplemental Material [45]), where the peak density is reduced and a strong rippling effect is seen for the structured pulse case. This will play a crucial role in basic laser coupling to flat targets and affect all relativistic laser-solid phenomena. Figure 3(f) instead shows the density profile for the $a_0 = 0.3$ case with the structured pulse. The grating structure is preserved without any significant change. We can thus conclude

that, at weakly relativistic peak intensities, the relatively low intensity of the prestructure does not affect the behavior of the interaction of the harmonic pulse with the plasma. In this regime, such pulses are very robust and can be used even for resonant phenomena such as the generation of surface plasma waves.

The angular distribution of electrons from simulations, as shown in Fig. 4, also shows a major effect of the femtosecond prestructure on the directions, flux, and energies of the electrons. These are plotted for three AOIs ($\theta_{\text{inc}} = 12^\circ/23^\circ/32^\circ$), where $\theta_{\text{inc}} = 23^\circ$ is the resonant case for surface plasma wave generation and the other two values are the nonresonant cases. Also, these are plotted considering electrons above 100 keV energy (as in the experiment). For the resonant case (i.e., $\theta_{\text{inc}} = 23^\circ$), the SPW mode along the negative surface (270°) can be seen in Fig. 4(c), which is for the clean pulse case, with γ reaching 6, which corresponds to ~ 2.5 MeV, and the electrons along the positive surface (90°) reaching a maximum of ~ 1.5 MeV. For Fig. 4(d) case, corresponding to the structured pulse, the electrons are preferentially accelerated along the positive surface with a maximum energy of ~ 1.5 MeV, and the SPW mode along the negative surface is suppressed with a maximum energy of ~ 0.5 MeV. Also, we see a significantly higher flux of electrons towards 90° than at 270° . For the nonresonant angles, as shown in Figs. 4(a), 4(b), 4(e), and 4(f), we see that the electron distributions and peak energies remain similar for both pulse shapes, but there is an increase in flux for the structured pulse case. This is because of the higher energy dump in the structured pulse case. Thus, the electron distributions and energy spectra for the grating resonance case are more sensitive to the pulse shape.

The phase space is shown in Fig. 4(g) for the clean pulse and in Fig. 4(h) for the structured pulse. For the clean pulse,

we can see the formation of electron bunches separated by λ (highlighted by the dashed circle, and a zoomed inset [better visible in the digital version]) along the negative surface direction, consistent with the SPW acceleration mechanism. Such a bunching effect is significantly suppressed in the structured pulse case. Also, for the structured pulse, the acceleration is stronger along the positive surface, but due to the lack of electron bunches separated by λ , we cannot associate it with the SPW acceleration mechanism.

To conclude, we have performed experiments and simulations to study the fundamental interactions of extreme-contrast 400-nm pulses with flat and sub- λ grating targets. We show that femtosecond prestructures, which are inherently produced in efficient SHG of femtosecond, broadband terawatt/petawatt class lasers, modify the local electron densities, field profiles, and electron distributions with regard to resonant SPW excitation in the grating target. We also present that, at weakly relativistic intensities, the relatively low intensity of the prestructure does not affect the laser-solid

interactions. We emphasize that the femtosecond prestructures play a crucial role in such extreme contrast interactions with solids. The modifications of the SPW response can serve as a sensitive indicator of the prestructures at the femtosecond levels.

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