

Study of 2ω and $3/2\omega$ harmonics in ultrashort high-intensity laser plasma interaction

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Abstract. An experimental study is presented on measurements of optical spectrum of the laser light scattered from solid surface irradiated by Ti:sapphire laser pulses up to an intensity of 1.2×10^{18} W cm⁻². The spectrum has well-defined peaks at wavelengths corresponding to 2ω and $3/2\omega$ radiations. The spectral features vary with the laser intensity and show blue-shift with increasing laser intensity. At a constant laser fluence, the spectrum is red-shifted with increasing laser pulse duration. The observed results are explained in terms of the density scale length variation of the plasma and laser chirp.

Keywords. Parametric instability; three-halved harmonics; ultrashort laser plasma interaction; pre-pulse parameters.

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1. Introduction

Existence of a pre-pulse ahead of any fs laser pulse has been a matter of concern for the study of ultrashort high-intensity laser plasma interaction, specially in developing laser plasma as a source of high-energy X-rays [1] and particles [2,3] (electrons, protons, ions) for potential applications in radiography [4], nuclear physics [5] etc. The driving laser used for such applications is invariably a chirp pulse amplification (CPA) based system. Intense laser pulses from such laser systems may have many pre-pulses like picosecond pedestal arising from imperfect compression, a nanosecond pedestal due to amplified spontaneous emission from the high-energy amplifiers, pre-pulse leakage from pulse selector etc. These pre-pulses can lead to uncontrolled formation of plasmas of relatively much longer scale length before the actual high-intensity main pulse arrives. A knowledge of pre-pulse parameters, namely intensity, pulse duration and fluence, are essential for understanding the ultrashort laser-matter interaction, as well as to control the source parameters. *In situ* monitoring of ultrashort high-intensity laser-plasma interaction conditions can be done by the spectral measurements of scattered laser radiation. For instance, the emission of 2ω and $3/2\omega$ radiations arises due to different processes which occur at plasma regions of quarter critical density and near the critical density respectively.

The $3/2\omega$ radiation is generated by the onset of two-plasmon decay (TPD) and stimulated Raman scattering (SRS) [6] by one of these plasmons. The TPD and SRS instabilities take place in the plasma region where the electron density is less than or equal to the quarter critical density. In the TPD processes, a laser photon decays and produces two plasmons. The $3/2\omega$ radiation is generated as a result of stimulated Raman scattering by one of these plasmons. The instability threshold is determined by the density scale length [7]. On the other hand, the 2ω radiation is produced by electron plasma wave coupling at the critical density surface of the plasma. The other mechanism of generation of 2ω is when the intense laser field drives a relativistic oscillation of plasma surface, which causes a periodic phase modulation of the reflected light and, hence, the emission of harmonics of the laser frequency. The integral harmonic emission growth rate depends on the density scale length which is determined by the pre-pulse and the laser parameters, namely intensity and pulse duration. In this paper, we report an experimental study on measurement of the spectrum of the light scattered from a solid surface irradiated by Ti:sapphire laser pulses of intensity up to $1.2 \times 10^{18} \text{ W cm}^{-2}$. The spectrum has well-defined peaks at wavelengths corresponding to 2ω and $3/2\omega$ radiations. The optical spectrum is measured for laser intensities in the range of 8.2×10^{16} – $1.2 \times 10^{18} \text{ W cm}^{-2}$ for a constant pulse duration of 45 fs and for pulse durations varying in the range of 45–800 fs, for a fixed fluence of $1.6 \times 10^4 \text{ J cm}^{-2}$. The spectral features vary with the laser intensity and show blue-shift with increasing laser intensity. On the other hand, at a constant laser fluence, the spectrum is red-shifted with increase in the laser pulse duration. The observed results are explained from the variation of the density scale length plasma.

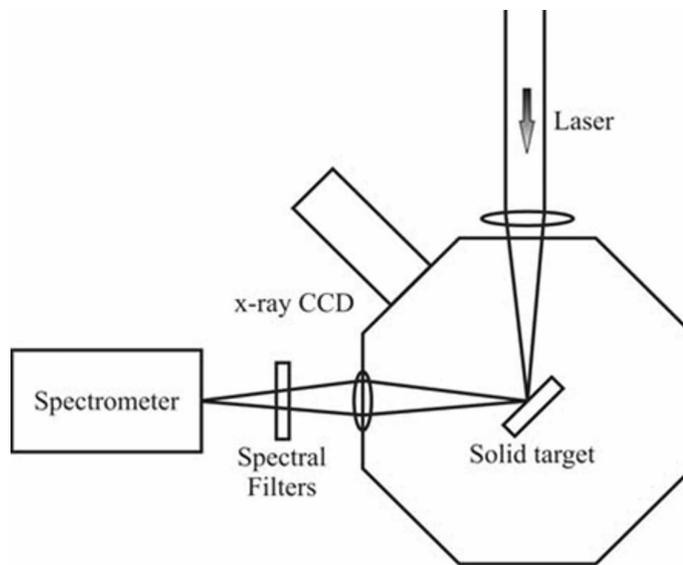


Figure 1. The experimental set-up.

2. Description of the experiment and results

A schematic of the experimental set-up is shown in figure 1. Experiments were carried out with femtosecond pulses of 45 fs (FWHM) duration, obtained from a Ti:sapphire laser system operating at a 10 Hz pulse repetition rate. The laser has a central wavelength of 790 nm with a bandwidth of 16 ± 2 nm after the compressor. The picosecond intensity contrast ratio measured with third-order autocorrelator was better than 10^5 , and the nanosecond intensity contrast ratio measured with fast photodiode was better than 10^6 . Plasma was produced by focussing the laser pulses onto a thick copper target placed at 45° using an $f/8$ lens. The target chamber was evacuated to a pressure of 10^{-5} Torr. Maximum pulse energy used in the current experiments was 150 mJ corresponding to a focussed intensity of $\sim 1.2 \times 10^{18}$ W cm $^{-2}$, for a measured focal spot diameter of 18 μ m (FWHM). The target was raster scanned so that laser pulse interacted with the fresh surface of the target, every shot. The light scattered in the specular reflection direction was focussed with an $f/4$, 200 mm focal length lens onto a fibre-coupled spectrograph (USB 2000, Ocean Optics). The spectrograph operates in the spectral range of 200–900 nm with a resolution of 0.3 nm. Coloured glass filters (Schott BG 39) were used to attenuate the strongly scattered laser light at the fundamental wavelength. The image processing and capturing were done with the indigenously developed software ‘Tarang’.

Figure 2 shows the optical spectrum for three different laser intensities. Here, the laser intensity was varied by changing the laser energy, while keeping the pulse duration and the focal spot fixed. At a low intensity of 8.2×10^{16} W cm $^{-2}$, the spectrum shows 2ω emission at a wavelength of 398 nm. As the intensity is increased to 3.6×10^{17} W cm $^{-2}$, $3/2\omega$ emission appears at a wavelength of 521 nm, along with the 2ω emission at 392 nm. On further increasing the intensity to 1.2×10^{18} W cm $^{-2}$, only a more blue-shifted $3/2\omega$ emission (centred at 515 nm) is seen. The saturation of the signal was avoided using neutral density filters in front of the spectrograph.

The $3/2\omega$ emission depends on the plasma density scale length. The growth rate of the two-plasmon decay instability in an inhomogeneous plasma increases with plasma scale length at the quarter critical density. It appears that the foot of the laser pulse, tens of ps before the peak of the pulse, has enough intensity to create plasma (pre-plasma), with which the main pulse interacts, instead of interaction with a solid target. As the laser intensity is increased, the plasma formation takes place at an earlier time, leading to a longer scale length pre-plasma. As the scale length of the pre-plasma increases with increasing laser intensity, there is an increase in the $3/2\omega$ emission intensity. On the other hand, increase in the scale length means the laser beam gets reflected at a larger distance from the critical density, leading to lesser excitation of the plasma wave at the critical density (by the evanescent wave), resulting in lower second-harmonic generation at the critical density. Hence one gets reduction in second harmonic generation with increase in the laser intensity.

It may be noted that the 2ω and $3/2\omega$ emissions are observed approximately at 395 ± 3 nm and 518 ± 3 nm respectively. This means that both are blue-shifted from the expected wavelengths of 398 ± 2 nm and 531 ± 2 nm respectively. The observed blue-shift may be due to the ionization blue-shift of the fundamental

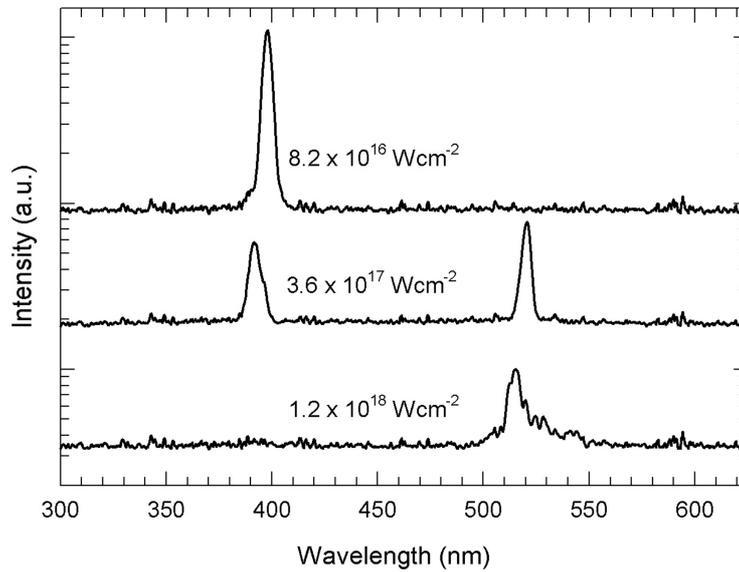


Figure 2. The scattered light spectrum at three different laser intensities, showing second and 3/2 harmonic emissions.

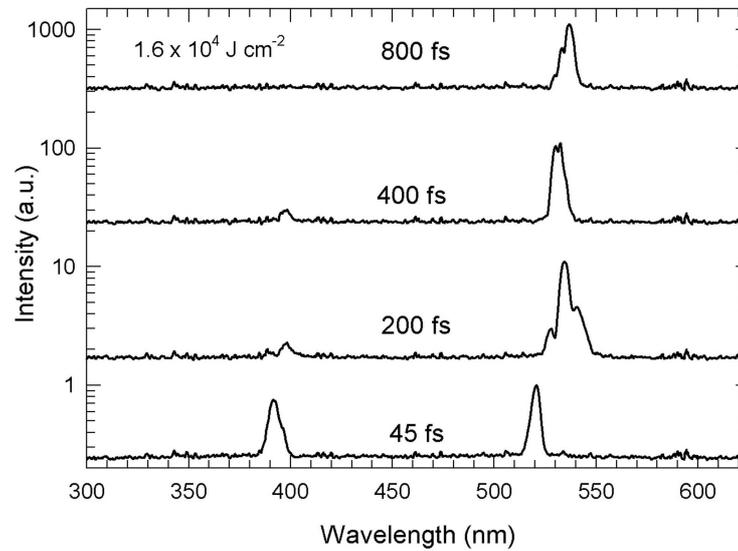


Figure 3. Optical spectrum as a function of laser pulse duration for a fixed fluence of $1.6 \times 10^4 \text{ J cm}^{-2}$.

wavelength of laser pulse [8] or due to Doppler shift due to the forward motion of the preformed plasma.

Figure 3 shows the variation of scattered light spectrum with the laser pulse durations in the range of 45–800 fs, for a fixed fluence of $1.6 \times 10^4 \text{ J cm}^{-2}$

(corresponding to 3.6×10^{17} W cm⁻² for the 45 fs pulse). The laser pulse duration is varied by changing the separation between the two gratings in the pulse compressor, with no change in the laser pulse energy. It can be observed that the 2ω signal increases on decreasing laser pulse duration. On the other hand, $3/2\omega$ emission does not show much difference on increasing the pulse duration though the laser intensity is ~ 18 times higher at 45 fs as compared to intensity at 800 fs.

The change in pulse duration of the ultrashort pulse is accomplished by adjusting the separation between compressor gratings. Beyond 100–150 fs pulse duration, the foot of the ultrashort pulse vanishes, and one gets interaction of the laser pulse (with the pulse duration varying), with no pre-plasma. In this regime, increasing the pulse duration means increasing the density scale length ($c_s\tau$). Hence, by increasing the pulse duration, the second harmonic intensity reduces because of the increasing distance between the reflecting surface and the critical density surface. On the other hand, for the $3/2\omega$ emission, the increase in scale length means a longer region for the two-plasmon decay instability to grow. This compensates for the lower intensity, thereby keeping the intensity of the $3/2\omega$ emission unchanged, as observed in figure 3.

The other interesting observation is the red-shift in both 2ω and $3/2\omega$ signals. This can be understood from the fact the pulses are negatively chirped as pulse duration is increased. As the pulse is stretched, the two-plasmon decay instability threshold is reached more and more later in time (towards the peak of the laser pulse) leading to a red-shift in the $3/2\omega$ emission.

A study on the dependence of 2ω and $3/2\omega$ spectra on laser parameters is carried out. The study showed drastically different behaviour of the second-harmonic intensity and the $3/2\omega$ emission intensity when the laser pulse intensity is decreased by lowering the pulse energy or by increasing the pulse duration. In either case, the observed behaviour can be explained in terms of the change of density scale length.

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