Evolution of plasma produced by intense femtosecond lasers and observation of backscattered three-halved harmonic generation

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1. Introduction

Two-plasmon decay (TPD) leads to the generation of halved harmonics that were extensively investigated at initial stages of laser plasma investigations using pico- and nano-second pulses [1,2]. Laser plasma studies using femtosecond pulses have shown a specific peculiarities of 3ω/2 harmonic generation [3,4] that distinguish them from previously observed properties of halved harmonics [5,6].

Recently we investigated the backscattered 3ω/2 harmonic generation from the surfaces of low-Z and high-Z targets (Z means the atomic number) [7]. We have shown that in the case of multi-shot regime, when each next shot interacts with the same spot of the surface as previous one, the specific conditions of the plasma produced at the target surface lead to the generation of backscattered 3ω/2 harmonic that did not appeared in a single-shot regime. We proposed the mechanism of phase-matching conditions that allowed combining the backscattered photon with the longitudinal electron waves caused by TPD. However, the dynamics of plasma created at multi-shot conditions remained unstudied.

In this paper we analyse the evolution of low-Z (boron) plasma in single-shot and multi-shot regimes using time-resolved shadowgram technique and compare the plasma dynamics with the characteristics of backscattered 3ω/2 harmonic radiation.

2. Experimental arrangements

The output characteristics of CPA Ti:sapphire laser (TSA-10F, Spectra Physics) operated at 10 Hz pulse repetition rate were as follows: wavelength: λ=795 nm, pulse energy: E=10 mJ, pulse duration: t=150 fs. We also were able to vary the output pulse duration from 130 fs to 1.6 ns by changing the distance between the gratings of the compressor of Ti:sapphire laser.

The radiation of this laser was divided by a beam splitter at the ratio of 9:1. The main (pumping) beam was propagated through the delay line and focused by a 10-cm focal length lens at the angle of interaction of 20° on the target placed in a vacuum chamber. The beam waist radius (HW1/e²M) was measured to be 10 µm. The maximum intensity on the target surface was as high as 10¹⁶ W cm⁻¹. The boron slab was used as a target in these experiments.

The radiation reflected from the beam splitter (probe beam) was frequency doubled (λ=397.5 nm) in a nonlinear crystal and irradiated the laser plasma and target at the orthogonal direction. This radiation was used for the analysis of plasma absorption characteristics at different delays between the pumping and probe pulses. We used the shutter at different positions allowing the single- and/or the multi-shot regimes to be provided for both beams. The radiation of probe beam propagated through the laser plasma.
was registered by CCD camera. The spectral characteristics of generated plasma and backscattered harmonic radiation were registered by a fiber optics spectrometer (USB2000, Ocean Optics).

3. Results and discussion

The 10 Hz pulse repetition rate radiation was interacted with a boron slab without the change of target position. We were able observing the backscattered $2\omega$ radiation generated from the initial pulses interacted with the target. After 1 s to 5 s from the beginning of interaction the backscattered $3\omega/2$ harmonic radiation was appeared. This process lasted during 10 s to 20 s depending on experimental conditions (target type, pulse duration, focusing lens position, etc) and then vanished due to the distortion of the optimal conditions of backscattered $3\omega/2$ harmonic generation. After that the shutter was closed and the target was shifted so that the next set of interaction with the target was performed at a fresh surface. No $3\omega/2$ harmonic generation was observed in the case of single-shot interaction as well as in the case of the variations of target position from shot to shot at 10-Hz pulse repetition rate.

We measured the efficiencies of $2\omega$ and $3\omega/2$ harmonic generation to be $10^{-6}$ and $2 \times 10^{-7}$, respectively. The appearance of $3\omega/2$ emission was unstable as well as its amplitude. The detailed analysis of spectroscopic measurements of backscattered $3\omega/2$ and $2\omega$ radiation from the low-Z and high-Z targets is presented in [7].

The motivation of our present studies was an analysis of dynamics of plasma generated in single-shot and multi-shot regimes in the cases of various pulse durations in the conditions of backscattered $3\omega/2$ harmonic generation. The multi-shot regime, when the laser pulses interacted with the same spot of boron surface at 10-Hz pulse repetition rate, confirmed our assumptions of variations of plasma properties. Our analysis of the plasma shadowgrams at such conditions have shown that the process of plasma formation is continued after initial shots inside the thin hole drilled by a femtosecond radiation (Fig. 1).

We analyzed the shadowgrams of laser plasma generated at the boron surface in a single-shot regime of interaction when the fresh surface was provided for each next shot at the delays between the main and probe pulses varying from $-6$ ps to 6 ns. The plasma generation was accompanied by a growth of probe beam absorption. The growth of the sizes of dense plasma was observed at the initial stages of plasma formation (up to delays of 126 ps). In some cases the “plasma bullets” were seen in front of the whole plasma volume (see the shadowgram registered at 186 ps delay).
that was previously observed by Vogel et al [8]. At the next stages of plasma formation its density decreased that led to the increase of propagation of a probe beam through the plasma area. This process was observed at the delay range of 180 ps to 1 ns. After that the further growth of plasma area and inhomogeneous propagation of 397.5-nm radiation through the absorbed area were observed. Finally, after 6 ns from the beginning of laser-surface interaction, the absorption area of plasma was disappeared indicating the decrease of electron density well below the level corresponding to the critical plasma density for 397.5-nm radiation.

The importance of time characteristics of laser pulses was analyzed in the case of single-shot interaction. We observed a strong backscattered $2\omega$ emission in the case of 400-fs pulses, whereas only a broadband plasma emission together with suppressed second harmonic radiation were registered in the case of short pulses. A difference between the short-pulse- and long-pulse-induced backscattered emission was clearly seen in the multi-shot regime, when the $3\omega'2$ harmonic radiation was dominated in the spectra induced by 600-fs pulses, whereas only the second harmonic emission was seen in the spectrum of backscattered radiation dominated by a plasma emission in the case of 130-fs pulses.

Most of our studies were carried out using 200-fs laser pulses. We also analysed the plasma images at different pulse durations. The generation of “plasma bullets” was observed in the case of short pulses. These shadowgrams were registered at the single-shot regime. The generation of plasma inhomogeneities in the case of short laser pulses can lead to the infringement of the optimal conditions of phase matching for three-halved harmonic generation. Indeed, we observed a strong difference in conversion efficiency of $3\omega'2$ harmonic generation, when such radiation completely disappeared from the spectral distribution of backscattered radiation in the case of shortest pulses.

The bullet-like and jet-like structures in laser-produced plasma has been observed previously using various techniques, such as optical probing, shadowgraphs, measurements of angular and energy distribution of suprathermal electrons, interferometry, and optical emission. The pump lasers used in these investigations also varied over a wide intensity range, from $10^8$ W cm$^{-2}$ to $10^{19}$ W cm$^{-2}$. The extensive numerical investigations of such fast moving plasma blocks in a plasma layer have been performed in [9]. The mechanism for this phenomenon is a ponderomotive force resulting from the spatial gradient of the electromagnetic momentum flux density produced within the plasma. Here we should underline that we were able observing the plasma bullets both in the cases of single-shot (that was commonly used in previous observations of this phenomenon) and multi-shot regimes (that wasn’t reported previously, to our best knowledge).

$3\omega'2$ harmonic radiation was generated due to the excitation of parametric instability in the plasma that led to the decay of light wave of frequency $\omega$ into two plasmons of frequency $\omega'2$ in the vicinity of plasma region of 1/4 of critical density. This process in turn leads to the generation of frequency $3\omega'2$ due to the different mechanisms. The first process is the coalescence of parametrically excited plasma oscillation ($\omega'2$) with incident light wave ($\omega$), and the second one is a very unlikely process of coalescence of three plasma oscillations ($3 \times \omega'2$). There is also the third mechanism for the generation of $3\omega'2$ emission, a combination of the plasmon wave with the pump photon reflected from critical density.

Our studies have shown that three-halved harmonic radiation is produced in a drilled hole. Unfortunately, the plasma dynamics during the interaction process cannot be thoroughly studied, as measurements inside the hole would be necessary. There are some questions yet unresolved concerning the phase-matching conditions, the initial density gradient of the
plasma inside the crater, and scale length at multi-shot regime. More detailed analysis of plasma conditions inside the hole would be available in the case of transparent targets (i.e., glass plates). The plasma dynamics is mainly governed by the ponderomotive force at intensities used and not by a simple hydrodynamic expansion. However, in the case of multi-shot regime the latter process can influence the conditions of backscattered $3\omega/2$ harmonic generation.

### 4. Conclusions

In conclusion, we presented our studies of boron plasma evolution using time-resolved shadowgram technique. These investigations were motivated by our observations of backscattered $3\omega/2$ harmonic generation in pulse repetition rate regime, when the multi-shot interaction with the same spot of the target led to the fulfillment of phase-matching conditions for three-halved harmonic generation in the case of low-Z targets. The shadowgram and spectroscopic studies have shown that this process occurs after tens shots inside the drilled hole. Two different plasma distributions were seen in the cases of single- and multi-shot regimes. This leads to the difference in plasma conditions inside the drilled hole leading to the phase matched $3\omega/2$ harmonic generation. A comparison of low- and high-Z targets has shown a difference in the dynamics of plasma formation. Our previously observed difference in the backscattered $3\omega/2$ harmonic generation in the cases of low- and high-Z targets can be explained by above-mentioned peculiarities of plasma formation in the drilled hole at multi-shot regime of laser-surface interaction.

### REFERENCES