RESONANT HIGH-ORDER HARMONIC GENERATION OF PICOSECOND ND:YAG LASER RADIATION IN LEAD PLASMA

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1. Introduction

The advantages of high-order harmonic generation (HHG) of laser radiation in a plasma plume could largely be realized with the use of low-excited and weakly ionized plasma, because the limiting processes governing the dynamics of the laser frequency conversion would play a minor role in this case. This has been confirmed by several studies concerned with high-order harmonic generation in the plasma plumes. A substantial increase in the cutoff of the generated harmonics, the emergence of a second plateau in the intensity distribution of highest order harmonics, the high efficiencies obtained with several plasma plumes, the resonance enhancement of individual harmonics, the efficient harmonic enhancement in the plasma plumes containing clusters of different materials and other properties have demonstrated the advantages of the application of specially prepared plasmas for the HHG. In this connection, a search for new plasma media and definition of the best experimental conditions, such as pulse duration of driving laser field, for efficient HHG in different spectral ranges is a way for further enhancement of harmonic yield. Some metal plasmas, such as indium and tin [1,2] have proven that, at appropriate experimental conditions, they can be considered as excellent plasma media for the enhanced harmonic generation in the 80 – 40 nm spectral range using ultrashort laser pulses. Moreover, some harmonic orders (13^{th} harmonic from In plasma and 17^{th} harmonic from Sn plasma in the case of 800 nm, femtosecond radiation) demonstrated the enhancement attributed to the influence of the ionic transitions.

In this work we present the results of studies of the HHG in the lead plasma. We compare those enhanced harmonics in the cases of laser ablation of pure Pb and its alloy. We also analyze the resonance-induced enhancement of the 11^{th} harmonic of Nd: YAG laser from this plasma in the presence of different gases.

2. Experimental

The passive mode locking Nd:YAG laser (λ = 1064 nm) has generated a train of pulses of 38 ps duration and 1.5 Hz pulse repetition rate. Two-stage amplification of single pulse was followed by splitting of this pulse on two parts, one (heating pulse) with the energy of 5 mJ, which was used for plasma formation on the target surface, and another (probe pulse) with the energy of up to 28 mJ, which was used, after some delay, for frequency conversion in the prepared plasma. The heating pulse was focused inside the vacuum chamber containing ablating target to create the plasma plume. The intensities of heat and probe pulse was up to 7×10^{10} W cm^{-2} and 4×10^{I3} W cm^{-2}, respectively The delay between these two pulses during most of experiments was maintained at 50-70 ns, which was optimal for efficient harmonic generation in laser-produced plasmas. The probe and converted radiation were analyzed using the vacuum monochromator (VMR2, LOMO), and the harmonic radiation in the range of 50 – 250 nm was detected using the sodium salicylate luminophore and photomultiplier tube. Various gases were inserted inside the vacuum chamber containing plasma plumes to analyze the influence of absorption and dispersion properties of these gases on the variations of the harmonic spectra.

3. Results and Discussion

We present some specific features of lead and alloy plasmas as the sources of HHG using picosecond laser pulses. The goal of these studies was to define whether the components of Pb-containing plasma influence the resonance-enhanced harmonic generation efficiency. Changing a composition of plasma from pure lead was accomplished using an alloy of this material, which consisted of tin and lead (at a volume ratio of ~5:3). We also compared plasma harmonic spectra from pure lead and alloy with those from pure tin. Another alloy target was a brass, which consisted of copper and zinc (65/35%). Additionally, we studied the variations of harmonic spectra from lead plasma while adding different gases in the vacuum chamber thus changing the components inside the plasma plume.

In the meantime, a lead plasma demonstrated the enhanced 11th harmonic (Fig. 1,a), which was stronger than lower orders. This peculiarity of lead harmonics was maintained at variations of different conditions of experiments (confocal parameter, plasma length, etc). The harmonics from mixture of lead and tin plasmas comprised both harmonic spectra, while maintaining the enhanced 11th harmonic (Fig. 1, b). We analyzed the harmonics from lead plasma at variable experimental conditions. The delay between heating and driving pulses is crucial for optimization of the HHG. The increase of delay allows the appearance of plasma particles along the path of converting pulse, which leads to the growth of HHG efficiency. Further increase of delay leads to saturation of the HHG at ~70 ns and gradual decrease of conversion efficiency at longer delays (>110 ns). We also investigated the influence of the distance between the target and the optical axis of propagation of the fundamental radiation. This distance was varied by a manipulator, which controlled the position of the target relative to the waist of the converting picosecond radiation. The common feature of these studies was an abrupt decrease of harmonics at irradiation of targets above the appropriate level. The reason of these observations is obvious and related with the over-excitation of plasma, which leads to the appearance of abundance of free electrons. The latter cause a phase mismatch between the waves of driving field and harmonics. This effect is especially important for lower order harmonics.

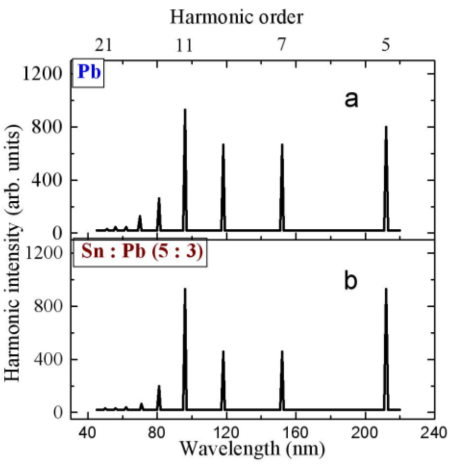

Fig.1. Harmonic spectra from (a) Pb and (b) Sn:Pb (5:3) alloy plasmas

The analysis of influence of fundamental (probe) intensity on efficiency of 11th harmonic generation in lead plasma was carried out. The dependence of harmonic signal on fundamental intensity was close to linear in a rather wide range (Fig. 2). With growth of laser intensity the decrease of power dependence occurred practically monotonously without presence of obvious oscillations. As frequency shift from 11th photon resonance between ground and exited states $6s^2 6p\ ^2P_{1/2}$ - $6s^2 8d\ ^2D_{3/2}$ for PbII was ~4.5 cm^{-1} only, the analysis of given behaviour was carried out in resonant approach. In Fig.2 the calculated dependence obtained at fitting of ac Stark shift, relaxation time, and generalized matrix elements is presented by solid line.

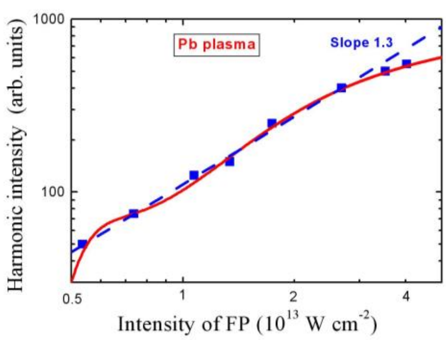

Fig.2. Comparison of experimental (dots) and calculated (solid lines) dependences of 11th harmonic signal on fundamental pulse intensity

The insertion of gases in the vacuum chamber containing targets led to variations of harmonic spectra compared with the case of pure lead plasma. We used four noble gases (He, Ar, Kr, Xe) possessing different dispersion, as well as different absorption, in the region of interest (50 – 250 nm). We used the He pressures up to 100 torr above which the optical breakdown of the gas (without ignition of the metal plasma) was observed. The abrupt decrease of 11th harmonic with the increase of He pressure. Meanwhile, some harmonics, in particular the 9th one, showed less decrease, and the 5th harmonic became even stronger compared with the plasma formation at vacuum conditions. Approximately same features were observed in the case of insertion of xenon, while a decrease of 11th harmonic was not so pronounced.

4. Conclusions

Our studies have demonstrated the variations of high-order harmonics from the Pb plasma using the mode-locking picosecond laser. The important peculiarity of these studies was the observation of the variations of enhanced harmonic in the vacuum ultraviolet range (50-200 nm), which thought to be originated from the resonance-induced growth of the nonlinear optical response of lead plasma at the wavelength of 11th harmonic lying close to the strong ionic transition (Pb II, λ = 96.72 nm). Our experiments with different gases added to the plasma volume have shown the role of propagation effect on the abrupt variation of resonant harmonic intensity.

[1] R. A. Ganeev, M. Suzuki, T. Ozaki, M. Baba, and H. Kuroda,” Strong resonance enhancement of a single harmonic generated in extreme ultraviolet range,” Opt. Lett. **31**, 1699-1701 (2006).

[2] M. Suzuki, M. Baba, R A Ganeev, H. Kuroda, T. Ozaki, “Anomalous enhancement of single high-order harmonic using laser ablation tin plume at 47 nm,” Opt. Lett. **31**, 3306-3308 (2006).