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Anomalous Single Harmonic Generation due to Strong Resonance in Extreme Ultraviolet Region

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Abstract

We demonstrate the generation of high harmonics (up to the 65th order, wavelength: 12.24 nm) of a Ti:sapphire laser radiation after the propagation of femtosecond laser pulses through the low-excited boron plasma produced by a prepulse radiation on the surface of different targets. High-order harmonics generated from the surface plasma of most targets showed a plateau pattern. The harmonic generation in these conditions assumed to occur due to the interaction of femtosecond pulses with ions. The conversion efficiencies in the plateau region were varied between 10^{-7} to 8×10^{-5} depending on the target. A 13th harmonic (61.2 nm) with conversion efficiency of 8×10^{-5} and output intensity almost two orders of magnitude higher than neighboring harmonics is obtained.

1. Introduction

Since the time when first ruby laser was demonstrated, future development of x-ray laser remains as one of a dream of laser physicists and almost all researchers engaged in laser applications involving biomedical applications and basic medicine. High-order harmonic generation are very attractive coherent source in the XUV and soft x-ray region. These unique features will have a large impact on x-ray laser application. An example is the precise measurement of time-resolved emission spectroscopy in live tissue, which is tremendously difficult even with advanced synchrotron radiation sources. Nonlinear frequency conversion sill allows tenability of the x-ray laser wavelength, allowing us the measure real atomic structure via photoelection holography. Such methods will allow the precise measurement and observation of various nano-structures, with which we hope to renew our understandings of new fields such as nanotechnology, nanobiology, and laser medicine. To apply for such applications, it is necessary to generate the strong high brightness of high-order harmonics.

The laser plasma has long been used as a medium for harmonic generation. The high-order harmonic in XUV region has been investigated in laser plasma formed by optical breakdown in gases and at the surface of solids target [1-3]. However the maximum observed order of harmonics reported in those studies was limited to the 27th one due to some concurred effects in high-excited plasma. No plateau pattern for high harmonics was reported in those studies. In this paper, we demonstrate the efficient generation of coherent XUV radiation after the propagation of femtosecond pulse through the low-excited plasma produced on the surface of different solid targets [4-6]. The harmonics up to the 65th (12.24 nm) order and the conversion efficiencies up to 8×10^{-5} were achieved in these studies.

2. Experiments

The pump laser used in this research was consisted of chirped-pulse amplification Ti:sapphire laser (Spectra-Physics, Tsunami + TSA10F) operated at a 10-Hz pulse repetition rate, whose output was further amplified using a three-pass amplifier. A portion of uncompressed radiation (pulse energy: 15 mJ, pulse duration: 210 ps, central wavelength: 796 nm) was split from the main beam by a beam splitter and used as a prepulse. This prepulse was focused by a spherical lens on a solid target located in the vacuum chamber and produced a plume predominantly consisting on neutrals and singly charged ions. The composition of plasma was analyzed using the time-integrated spectral measurements of plume in XUV, UV, and visible ranges. The focal spot diameter of prepulse beam on the target surface was adjusted to be approximately 600μm. The intensity of picosecond prepulse, I_{pp}, on the target surface was varied from 7×10^9 W cm^{-2} to 8×10^{10} W cm^{-2}. After some delay, the femtosecond main pulse (pulse energy: 8 mJ, pulse width150 fs, central wavelength: 796 nm) was focused on the target plasma from the orthogonal direction using 200-mm or 100-mm focal length lenses. Our experiments were carried out up the maximum intensity of the femtosecond main pulse of $I_{fp}=1\times10^{15}$ W cm^{-2}. The high-order harmonics were analyzed by a flat-field grazing-incidence XUV spectrometer with a Hitachi 1200-groove/mm grating. An additional gold-coated grazing-incidence cylindrical mirror was used for the image translation from the plasma area to the detector. The XUV spectrum was detected by a microchannel plate with phosphor screen and recorded by CCD camera. The details of calibration were described in Ref.7.

3. Results an discussions

We used different solid targets (Ag, B, In, C, W, Se, Mo, Nb, Si, Zr, Mg, Cd, Pd, Al, Cr, Ta) for the preparation of suitable laser plumes for our experiments. The choice of targets was stipulated by our aim to analyze the influence of atomic number, ionization potentials, and other parameters of samples on the HHG from different plumes. Our efforts were concentrated on determination of the optimal conditions for achieving the maximum harmonic order (i.e., cutoff energy) as well as maximum conversion efficiency for high harmonics using low-excited laser plasma. The plateau pattern of high-order harmonic distribution was appeared in these experiments. In particular, high harmonics up to the 65th order at a wavelength of 12.24 nm were observed in experiments with boron plasma at loosely focused conditions (b=6 mm, L_p=0.6 mm, b is confocal parameter, L_p is the plasma sizes). Figure 1 shows the typical spectra of the high-order harmonic generation by boron and silver plasma. The HHG from the B plume appeared to be similar to those observed in gas-jet

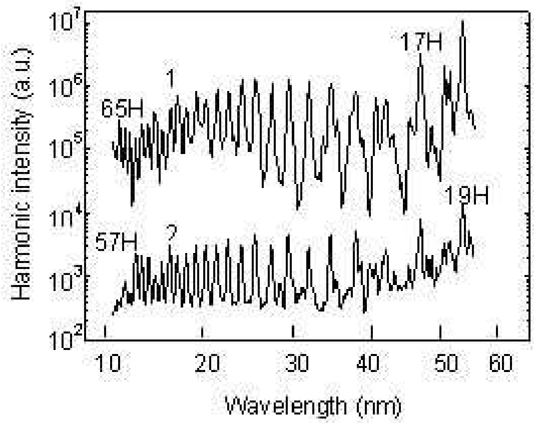

Figure 1. Harmonic output as a function of the wavelength of generated XUV radiation in the cases of (1) boron and (2) silver.

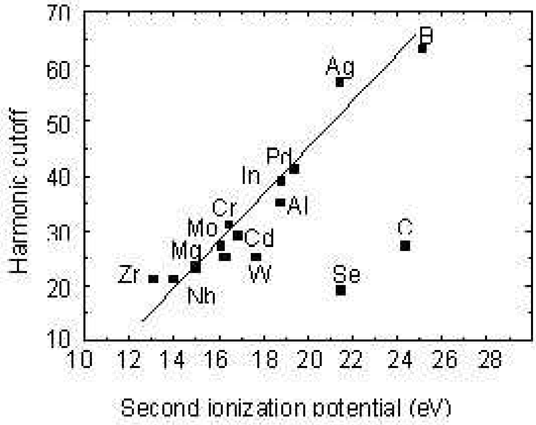

Figure 2. Dependence between the cutoff harmonic and ionization potential of ions (second ionization potential) for different targets.

experiments, with characteristic shape of plateau for the harmonics exceeding 17th order (Fig. 1, curve 1). The conversion efficiencies were in the range of10^{-4} (for the 3rd harmonic) to 10^{-7}. Figure 2 shows the dependence between the cutoff harmonic and ionization potential of ions (second ionization potential) for different targets. These observations underline a decisive role of free electrons appearing during further ionization of singly charged particles that led to the restriction of cutoff energy. The conversion efficiencies in the plateau region were varied between 10^{-7} to 8×10^{-6} depending on the target. Most intriguing pattern was appeared in the case of indium plasma in Fig. 3. We observed anomalously strong 13th harmonic radiation (wavelength: 61.2 nm), which almost two orders of magnitude exceeded the neighbor ones. Figure 4 shows the line trace of indium. The conversion efficiency of 13th harmonic was measured to be 8×10^{-5}. For the confirmation that this strong line in the range of 61.2 nm was originated from the nonlinear process rather than from a simple re-excitation of plasma line we investigated the influence of 13th harmonic output on polarization characteristics of the main pulse. The 25 degree rotation of quarter-wave plate led to the complete disappearance of 61.2 nm emission, as it should be assuming the origin of HHG, while the excited lines of plasma spectrum observed at different polarizations of main beam remained unchanged. Another test of this process was carried out by the variation of the wavelength of main beam

4. Conclusion

We demonstrated the generation of high harmonics (up to 65th order, wavelength: 12.24 nm) after the propagation of femtosecond laser pulses through the low-excited surface plasma created by a prepulse radiation. High-order harmonics generated from most plumes showed a plateau pattern. The harmonic generation in these conditions assumed to occur due to the interaction of femtosecond pulses with ions.The conversion efficiency for the harmonics generated in the plateau region was varied in the range of 10^{-7} to 8×10^{-6} depending on target. The main contribution to the limitation of harmonic generation efficiency was caused by free-electrons-induced self-defocusing of main beam.

Our observation of the considerable resonance-induced enhancement of a single harmonic wavelength: 61.2 nm) in the plateau region with the efficiency of 8×10^{-5} in the case of In plume can offer some expectation that analogous processes can be realized in other plasma samples even in the shorter wavelength range where the highest harmonics were achieved. It is difficult to realize this approach in gas puffs due to the necessity of the preparation of specific conditions for the excitation of appropriate levels of nonlinear medium prior to laser-matter interaction. An enhancement of HHG efficiency should include the optimization of both macroscopic and microscopic responses. From the macroscopic point of view, such an optimization dominantly means an achievement of ideal phase matching conditions. Microscopic optimization includes the methods of the growth of polarization of harmonics that depends on nonlinear susceptibility of a separate harmonic. The last parameter considerably depends on the relation between the excited states of nonlinear medium in the vicinity of harmonic wavelength. To our opinion, the strong 13th harmonic was a result of resonance-induced enhancement of nonlinear susceptibility in the vicinity of 61 nm.

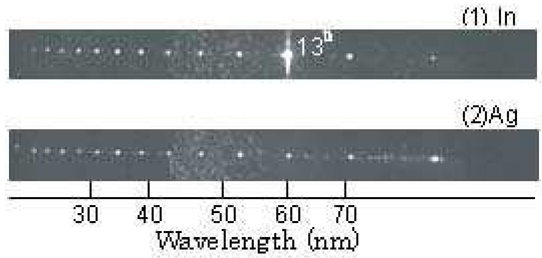

Figure 3. High-order harmonic spectra from (1) indium and (2) silver plumes

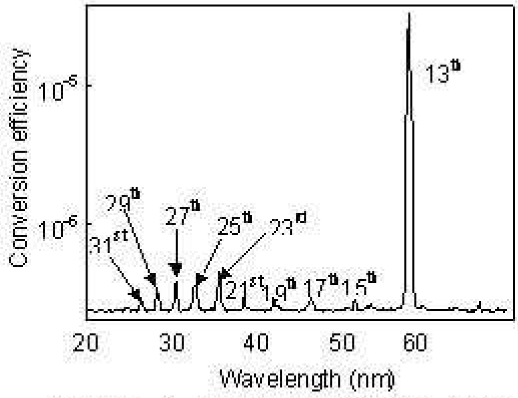

Figure 4. Spectrum of the high order harmonic generation was obtained using the indium plasma. The conversion efficiency of 13th harmonic is 8×10^{-5}.

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