High-order harmonic generation from laser plasma produced by pulses of different duration

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The high-order harmonic generation was analyzed by interaction of the femtosecond pulses with the laser plasma produced on the surfaces of various targets. The plasma formation was accomplished by the interaction of the prepulse radiation of different pulse duration (160 fs, 1.5 ps, 210 ps, and 20 ns) with the low-*Z* (lithium, boron, carbon), medium-*Z* (manganese, zinc, nickel), and high-*Z* (silver, barium) targets. We showed that plasma formation conditions play a crucial role in harmonic generation and the optimization of this process mostly depends on the energy of prepulse rather than its intensity at the target surface. These studies also demonstrated that the delay between the prepulse and femtosecond pulse is another important parameter, which distinguishes harmonic generation in the cases of the low- and high-*Z* targets.

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I. INTRODUCTION

Plasma formation during high-order harmonic generation (HHG) in laser-gas jet experiments has long been considered as a restricting factor, which diminishes the optimal conditions for the HHG in the extreme ultraviolet (xuv) range. Various methods were used to decrease the negative influence of free electrons on the phase matching conditions and laser radiation divergence. The prolonged gas media, waveguides with variable diameter, optimization of coherence length, adaptive control of the phase relations between the driving and harmonic waves, etc., were applied to enhance the harmonic conversion efficiency in the xuv region [1–3]. Since in most of the reported HHG experiments the driving radiation intensity exceeded the barrier suppression intensity of used gases, the presence of ions and free electrons were considered as an inevitable factor diminishing the HHG efficiency [4–6].

In the meantime, in the case of harmonic generation from the laser ablation of solid targets, the presence of ions was considered as the origin of harmonic generation [7,8]. The initial studies of the HHG from laser plumes [7–11], which were carried out at the conditions of high-excited, high-charged plasma, have shown the low-order harmonic cutoffs and the absence of the plateaulike distribution of harmonics in the short-wavelength range. The application of low-excited, singly charged laser ablation allowed for considerably improving the conditions of harmonic generation from the laser plasma [12]. The high harmonics in the range or orders from 40 to 60 have been obtained at such plasma conditions from almost all solid targets of the periodic table [13,14]. Some plasma samples have shown high conversion efficiencies at the plateau region (close to 10^{-5} [15]). The plasma harmonics have demonstrated relatively high cutoffs (65th, 71st, and 101st harmonics in the cases of B [16], V [17], and Mn [18] targets, respectively). The harmonic generation from some of these plasmas originated from the interaction of the femtosecond pulses with the doubly charged ions. Another interesting peculiarity of this process was the observation of the resonance-induced enhancement of single harmonics. This mechanism considerably increased the high harmonic yield at the specific spectral ranges close to the resonance transitions of ions. The conversion efficiencies up to 10^{-4} have been demonstrated at these resonance conditions [19,20].

These studies have underlined a role of plasma conditions for achieving the maximum conversion efficiencies and harmonic cutoffs. Various techniques were applied for the analysis of the ionization level, electron and ion concentration, spectral characteristics, and nonlinear optical parameters of laser plasma. Among them are the calculations using HYADES code [21], time-integrated [14] and time-resolved [22] spectral analysis of the plasma in the visible and uv ranges, analysis of the divergence of the femtosecond beam propagated through the laser plasma [23], *z*-scan studies of laser ablation [23], etc. This allowed for defining the conditions of the efficient generation of coherent xuv radiation through the harmonic generation in laser-ablated plumes.

Meanwhile, the important parameter that has never been explored during plasma HHG studies is a prepulse duration. Since almost all of the HHG studies from plasma plumes were carried out by using the prepulses of a few hundreds picosecond pulse duration, the conclusion was drawn about the optimal intensity of the prepulse radiation at the target surface [$\sim(1-4)\times10^{10}$ W cm^{-2}, depending on the absorptive properties of targets] [13,20,21]. It would be interesting to compare the HHG from the plasmas produced by the prepulses of different duration, since it can clarify which parameter (energy or intensity) of the prepulse plays a crucial role in the formation of optimal plasma.

Another interesting issue is the influence of the atomic number (*Z*) of target materials on the HHG at different delays between the prepulse and the main pulse. It would be worth analyzing whether the optimal delay between the two pulses is sensitive to the target element. One can expect a difference in the HHG from the low- and high-*Z* targets, since the heating as well as the expansion and the subsequent cooling and recombination of the plasma depend on the

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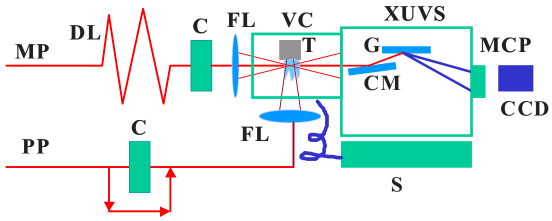

FIG. 1. (Color online) Experimental setup. MP, main pulse; PP, prepulse; DL, delay line; C, grating compressors; FL, focusing lenses; VC, vacuum chamber; T, target; S, USB2000 spectrometer; XUVS, xuv spectrometer; CM, gold-coated cylindrical mirror; G, gold-coated grating; MCP, microchannel plate; CCD, charge-coupled device.

atomic weight and density of the target material. Different velocities and recombination properties of the low- and high-Z atoms and ions can considerably change the concentration characteristics of plasma at different delays. The optimization of the delay between the pulses for specific plasma samples allows for achieving the maximum harmonic intensities and highest harmonic cutoffs.

In this paper, we present our studies of the HHG from the laser plumes produced by the interaction of the prepulse radiation of different pulse duration (160 fs, 1.5 ps, 210 ps, and 20 ns) with the silver, manganese, barium, lithium, zinc, nickel, boron, and carbon targets. We show that the plasma formation plays a crucial role for achieving the efficient harmonic generation and the optimization of this process mostly depends on the prepulse energy rather than its intensity at the target surface. We also show the difference in harmonic generation in the cases of the low- and high-Z targets, when the delay between the prepulse and femtosecond pulse becomes a crucial factor for achieving the efficient HHG from laser ablation.

II. EXPERIMENTAL ARRANGEMENTS

The experimental setup of the HHG from laser plasma was described in our previous studies [24]. Here we present some details of these experiments. The pump laser was a commercial, chirped pulse amplification Ti:sapphire laser system (Spectra Physics, TAS-10F), whose output was further amplified using a homemade three-pass amplifier operating at a 10 Hz pulse repetition rate. A prepulse was split from the amplified laser beam by a beam splitter before a main pulse compressor. The prepulse duration was 210 ps. We also varied the pulse duration of this radiation by compressing it in an additional compressor stage (Fig. 1). In particular, we used the 160 fs and 1.5 ps prepulses by adjusting the distance between the gratings of this compressor. The pulse energy of these three prepulses was adjusted to be approximately 10 mJ.

A spherical or cylindrical lens focused the prepulse on a solid target placed within a vacuum chamber to generate a laser ablation plume. High-Z (barium and silver), medium-Z (zinc, nickel, and manganese), and low-Z (carbon, boron, and lithium) materials were used as the targets in these studies. The width of the line focus at the target surface in the case of the focusing by a cylindrical lens was adjusted between 50 to 200 μm at 2 mm length of the plasma, and the intensity of 210 fs prepulse at the target surface was varied in the range of $I_{pp}=(0.7-7.0)\times 10^{10}$ W cm^{-2}. In the case of spherical focusing, the area of ablation was adjusted in the range of 0.6 mm and the 210 ps prepulse intensity at the target surface was varied in the range of $I_{pp}=(1-5)\times 10^{10}$ W cm^{-2}. In the case of 160 fs and 1.5 ps prepulses, the intensities at the target surface were considerably higher, since the geometrical characteristics of these studies were similar for three prepulse durations, while keeping the same pulse energy.

After the proper delay with regard to the prepulse irradiation, a main pulse ($\lambda=795$ nm, $E=12$ mJ, $t=115$ fs) was focused by a spherical lens ($f/10$) on the ablation plume from the orthogonal direction. The maximum intensity of the femtosecond laser beam at the focal spot was 6×10^{16} W cm^{-2}. Since this intensity considerably exceeded the barrier suppression intensity of singly charged ions, the position of laser focus was adjusted to be either before or after the laser plume to maximize the high harmonic yield. The intensity of the main pulse at the plasma area was varied between 7×10^{14} to 3×10^{15} W cm^{-2}. A magnesium fluoride window possessing small nonlinear refractive index was used in a vacuum chamber containing targets to exclude the self-phase modulation and white-light generation during the propagation of femtosecond radiation. The grating compressor was adjusted to achieve the shortest duration of the main pulse at the plasma area after the propagation through the focusing lens and chamber window. Our experiments were carried out at loose focusing conditions [$b>L_p$, b is the confocal parameter of the focused radiation, and L_p is the plasma length; $b=3$ mm, $L_p=0.6$ mm (in the case of the spherical focusing of prepulse radiation), and $L_p=2$ mm (in the case of the cylindrical focusing of prepulse radiation)].

The spectrum of generated high harmonics was analyzed by a grazing incidence xuv spectrometer with a gold-coated flat-field grating (Hitachi, 1200 grooves/mm). A gold-coated grazing incidence cylindrical mirror was used for the image translation of the harmonics from the ablation plume onto the detector. The xuv spectrum was detected using a microchannel plate with a phosphor screen (Hamamatsu, F2813-22P), and the optical output from the phosphor screen was recorded using a CCD camera (Hamamatsu, C4880). The details of the absolute calibration of the harmonics measured by this xuv spectrometer were described elsewhere [15]. The fiber spectrometer (Ocean Optics, USB2000) coupled with image translation lens pair was used to measure the spectra of laser ablation plume in the visible and uv ranges.

III. RESULTS AND DISCUSSION

The main goal of these studies was to define whether the prepulse duration influences the harmonic generation efficiency from the laser plasma. Another goal was to analyze the influence of the atomic weight of the targets on the HHG at different delays between the prepulse and main pulse. Below we present our studies of these two problems by using (a) the prepulses of different pulse duration, (b) different delays between the prepulse and main pulse, and (c) the targets of different atomic numbers.

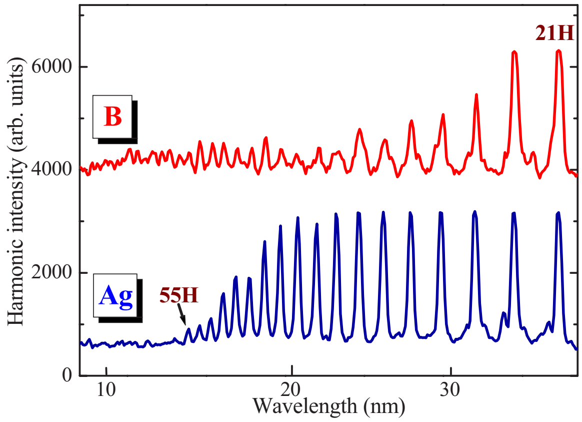

FIG. 2. (Color online) Harmonic spectra obtained from the silver and boron plasmas at the 17 ns delay between the 210 ps prepulse and 115 fs main pulse.

The properties of harmonic generation from some of the targets under investigation were analogous to those reported previously [14]. The plateaulike harmonic distribution was observed for most plasma samples. Figure 2 presents the harmonic spectra from some of the targets used in these experiments. The best harmonic generation efficiency was obtained in the case of silver plasma. The estimated conversion efficiency at the plateau region in that case was 8×10^{-6}, since our experimental conditions were close to those reported in Ref. [15] where the calibration of the absolute value of conversion efficiency was established.

Manganese plasma showed the highest harmonic cutoff among the samples under investigation. The harmonics up to the 93rd order were obtained (see inset in Fig. 3). The enhancement of a group of harmonics (from the 33rd to 43rd order) was observed during these studies of manganese plasma (Fig. 3). The 33rd harmonic (λ=24.1 nm) showed highest enhancement among other enhanced harmonics. The enhancement of 24.1 nm radiation (E_{ph}=51.7 eV) could be associated with the resonance-induced growth of conversion

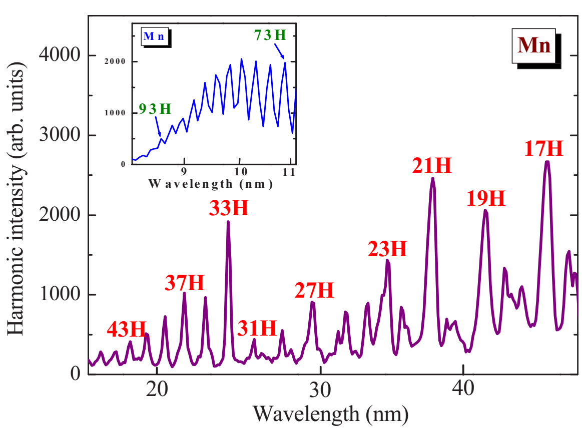

FIG. 3. (Color online) Part of the harmonic spectra obtained from the manganese plasma at the 88 ns delay between the 210 ps prepulse and 115 fs main pulse. Inset, harmonic distribution at the cutoff region.

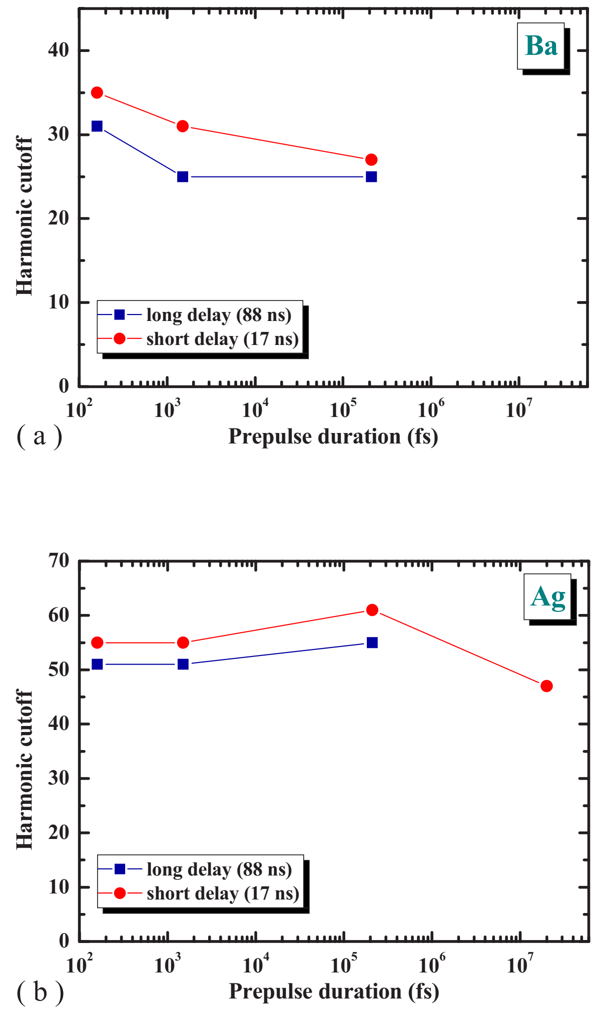

FIG. 4. (Color online) Harmonic cutoffs obtained from the high-Z plasmas prepared by using the prepulses of different duration in the cases of short (17 ns) and long (88 ns) delays. (a) Ba, (b) Ag.

efficiency in the vicinity of the Mn ionic transitions with strong oscillator strengths. The Mn II and Mn III transitions in the range of 51 to 52 eV were studied in past works and proved to possess strong oscillator strengths. The detailed analysis of the HHG from Mn plasma using shorter pulses (t=35 fs) will be reported soon [18]. Here we must underline the role of doubly charged Mn ions in the generation of highest harmonics, since the three-step model [25] cannot explain the origin of the harmonics exceeding the 31st order by assuming the HHG as a result of the interaction of the main pulse with the singly charged Mn ions.

Figures 4–6 show the influence of the prepulse duration on the harmonic cutoff (H_c) in the cases of relatively heavy [Ba (Z=56) and Ag (Z=47)], mid-weight [Zn (Z=30), Ni (Z=28), and Mn (Z=25)], and light [C (Z=6), B (Z=5), and Li (Z=3)] targets. One can see the different behavior of the H_c (t_{pp}) dependences for these three groups of targets. In the case of the high-Z targets (and in most cases of the medium-Z targets), no considerable change in the harmonic cutoffs was observed at the 160 fs, 1.5 ps, and 210 ps pulse durations of prepulse radiation. In some cases, we used the sec-

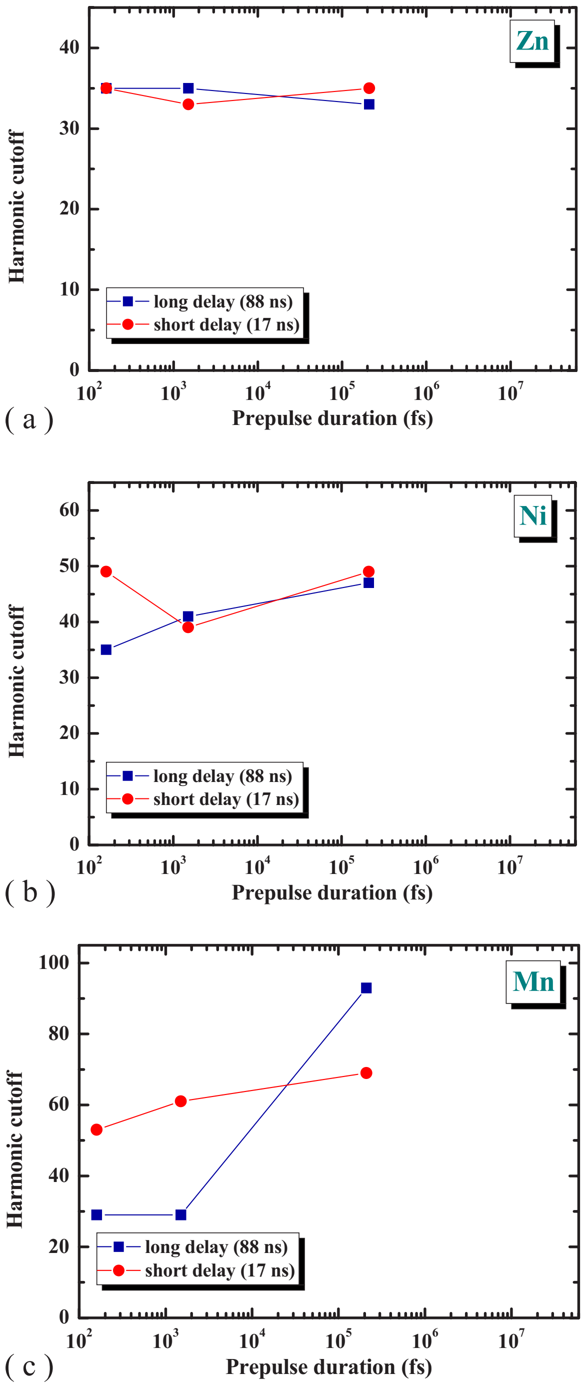

FIG. 5. (Color online) Harmonic cutoffs obtained from the medium-*Z* plasmas prepared by using the prepulses of different duration in the cases of short (17 ns) and long (88 ns) delays. (a) Zn, (b) Ni, (c) Mn.

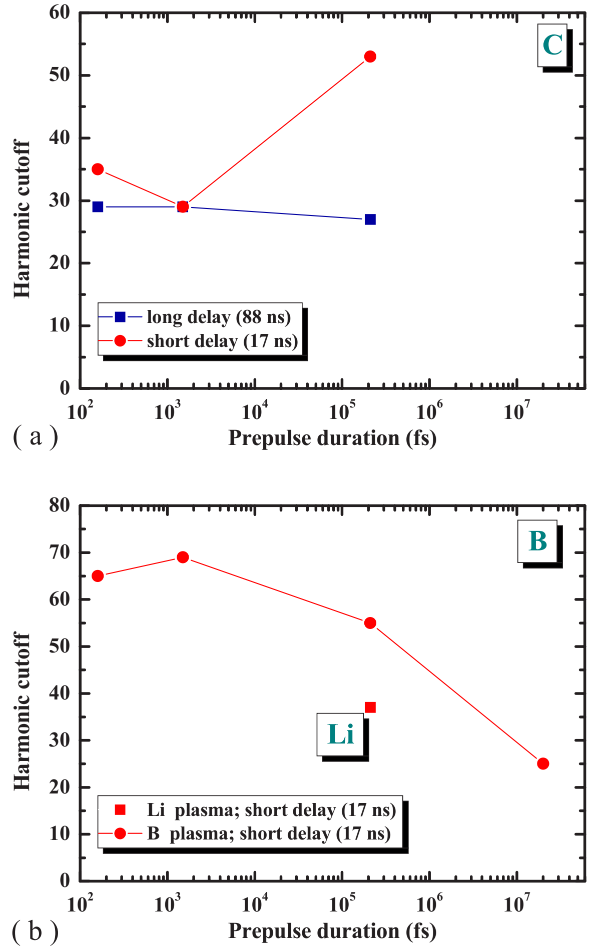

FIG. 6. (Color online) Harmonic cutoffs obtained from the low-*Z* plasmas prepared by using the prepulses of different duration in the cases of short (17 ns) and long (88 ns) delays. (a) C, (b) B and Li.

ond harmonic of Nd:YAG laser as a prepulse radiation of longer pulse duration (t_{pp}=20 ns) and also did not observe a considerable difference (for heavy targets) in harmonic cutoffs with regard to shorter prepulses.

In the case of the low-*Z* targets, a more pronounced H_c (t_{pp}) dependence was found compared to the high-*Z* samples. However, in that case we also did not observe the restricting factors, which stopped the HHG at significantly different intensities of heating radiation at the target surface. The only exception was the case of lithium plasma, where the high harmonics were generated only by using the relatively long (t_{pp}=210 ps) prepulses and short delays. The main difference in the HHG properties in that case was related with the delay between the prepulse and main pulse, rather than the prepulse intensities. Let us remind again that we maintained the same pulse energy of prepulse radiation at the variable pulse durations.

These observations showed the main parameter of laser prepulse, which is responsible for the creation of the optimal plasma conditions for the HHG. This is a pulse energy, which is responsible for the ablation of targets and preparation of the plasma where the efficient HHG can be achieved. Since the most important parameter of plasma (from the point of view of harmonic generation ability) is a concentration of singly charged (or in some cases doubly charged) ions, the conditions of plasma formation should be maintained with the appropriate manner to achieve the best characteristics of harmonic generation.

We analyzed the time-integrated spectra from the laser plasmas in the visible and uv ranges in the cases of different

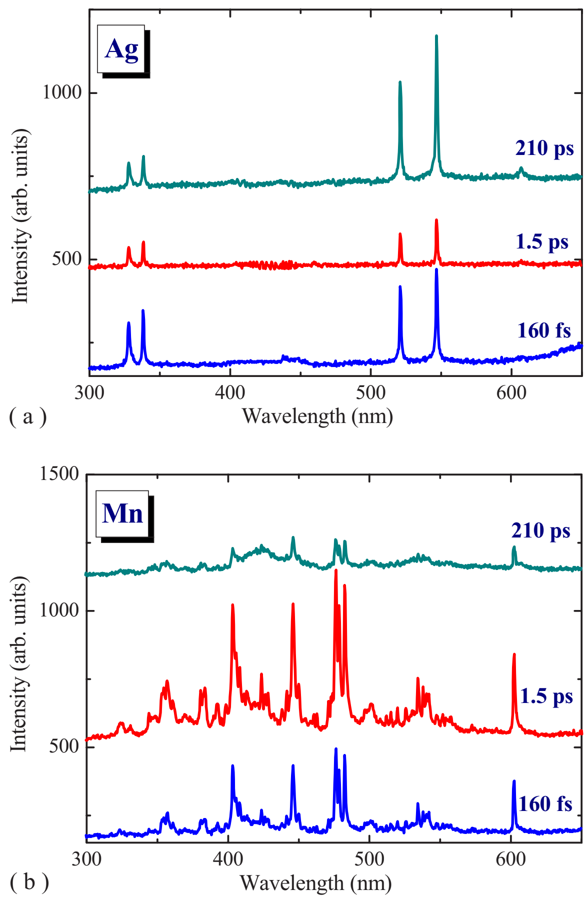

FIG. 7. (Color online) Plasma spectra emitted from the (a) Ag and (b) Mn plumes in the cases of the excitation by the 160 fs, 1.5 ps, and 210 ps prepulses.

pulse durations of prepulse radiation. Figure 7 shows some of these spectra obtained from the Ag and Mn plasmas prepared for the efficient harmonic generation. Though some of these spectra varied from each other, the overall pattern remained the same at different prepulse durations. These studies showed that the pulse duration of heating radiation does not considerably change the emission spectra of targets. The same can be said about the emission in the xuv region that confirmed the approximately equal plasma conditions at considerably different intensities of target excitation.

The harmonic generation strongly depended on the delay between the prepulse and main pulse. At very small delays (less than 5 ns), no harmonics were generated from all plasma samples due to insufficient concentration of the particles at the axis of the main beam. However, the harmonic properties of different plasma plumes considerably distinguished from each other at longer delays depending on the atomic number of the targets. In the case of 17 ns delay, the harmonic generation was observed from all of the samples, while in the case of 88 ns delay, only the high- and medium-Z plumes produced harmonics (Figs. 4 and 5). No harmonics were obtained at long delays in the case of the low-Z plumes (excluding carbon, Fig. 6). At the same time, the harmonic cutoffs from the high- and medium-Z plumes remained approximately unchanged for both the short and long delays.

Such a behavior can be attributed to the dynamic characteristics of the plasmas in the cases of the low- and high-Z target materials. Light particles possess higher velocity and this can lead to the depletion of the particles concentration in the laser plasma after a few tens of nanoseconds. At the same time, the high- and medium-Z particles for a longer time remain close to the target surface where the HHG occurs. For heavy targets, the dependence of the high harmonic yield on the delay between the pulses is not so pronounced compared to the low-Z materials. The difference between these dependences was clearly seen in the case of longer delays due to different concentration of the particles in the interaction area of a laser plume for the low- and high-Z targets. Based on this, the search of the optimal plasma conditions should take into account the role of the atomic characteristics of the targets.

Previously, second ionization potential of target materials was assumed as playing an important role in the definition of the harmonic cutoffs from different laser plumes [13,14,18]. It was found that the harmonic cutoffs observed from different plumes directly depended on the second ionization potentials of materials. This finding underlined the role of singly charged ions in harmonic generation. The studies reported in the present paper revealed the additional parameters of target material and prepulse, which should be taken into account during further search of higher harmonic cutoffs and harmonic yields from laser ablation.

From the empirical rule H_c (harmonic cutoff) $\approx 4I_i(\text{eV}) - 32.1$ [21], one can assume that the highest cutoff can be achieved in the case of highest ionization potentials (I_i) of the particles participating in the HHG. Indeed, in the case of singly charged ions (which are proven to be the main source of harmonics in most plasma HHG experiments), highest second ionization potentials of used targets corresponded to the highest cutoffs ever observed [16,21]. At the same time, as it was mentioned in the introduction to this paper, the plasma HHG studies have also revealed in some cases the role of doubly charged ions in achieving the highest harmonic cutoff energies (V [17], Mn [18]). The ionization potentials of V III and Mn III (i.e., the third ionization potentials of vanadium and manganese) and the observed harmonic cutoffs (71st and 101st orders, respectively) obeyed the above-mentioned empirical relation. One could expect to achieve higher harmonics by applying the materials with higher third ionization potentials.

In the present studies, the targets under investigation were chosen from the point of view of their highest third ionization potentials among other solid materials. However, we failed to achieve the higher harmonics with regard to previously reported data. The reason for this fault could be related with the specific phase-matching conditions in the case of a few plasma samples, which led to the efficient harmonic generation through the interaction of the radiation with the doubly charged ions, while in the case of other plumes, the influence of the enhanced free electrons concentration led to the growth of phase mismatch and self-defocusing.

Laser-induced plasma itself is a complex phenomenon, especially from the point of view of its nonlinear optical properties. The behavior of laser ablation changes considerably, depending on the equation of state, ionization potential, and cohesive energy of the material. The processes that determine harmonic generation from the plasma plume are complex and may include various factors that are not considered for gas harmonics. For example, the nonlinear medium is already weakly ionized for plasma harmonics, whose level depends on the prepulse characteristics. If the free electrons density is too high (which is the case when doubly charged particles exist in the plasma), it can induce the phase mismatch between the pump and harmonic waves, or defocus the pump beam. Both of these processes can reduce or even stop the harmonic generation.

IV. CONCLUSIONS

In conclusion, we have reported our studies of the two parameters (the temporal characteristics of heating prepulse radiation and the delay between the prepulse and main pulse), which play an important role during harmonic generation from the laser plasmas produced on the surfaces of different targets. In the case of the low-Z targets, we observed a strong dependence of harmonic yield and harmonic cutoff on the delay between the prepulse and the main pulse. At the same time, this dependence was less pronounced in the case of the high- and medium-Z materials.

Our studies reveal the insignificant role of the temporal characteristics of prepulse radiation in creation of the optimal plasma conditions for the efficient harmonic generation. We varied the prepulse duration in the range of a few orders of magnitude (from 160 fs to 20 ns) and in most cases observed approximately the same harmonic cutoffs at short delays for both the light and heavy targets.

From these studies we concluded that it is rather the prepulse energy than its intensity that influences the harmonic cutoff from different plasma plumes, i.e., the cutoff stays unchanged at different prepulse durations. The harmonic cutoff in the case of the low-Z targets was found to be highly dependent on the delay between the prepulse and main pulse. These findings allow further optimizing the harmonic generation from the plasma plumes by choosing the appropriate target materials and the delays between the pulses.

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