Influence of the main pulse and prepulse intensity on high-order harmonic generation in silver plasma ablation

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We present experimental studies of high-order harmonic generation in silver plasma ablation performed with the Ti:sapphire laser beams of the Advanced Laser Light Source (800 nm wavelength, 360 mJ total energy). We have independently varied the intensity of the prepulse (which creates the plasma ablation) and the intensity of the main pulse (which generates the harmonics), and studied their influence on the harmonic spectrum. We show here that the presence of doubly ionized atoms in the ablation, created either by a strong prepulse intensity or with the irradiation of the main pulse, is ineffective for the generation of harmonics.

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I. INTRODUCTION

After nearly two decades of research, high-order harmonic generation (HHG) has become one of the most promising techniques for the development of ultrashort radiation sources in the soft x-ray spectral region. HHG is produced by focusing short laser pulses in a nonlinear medium, thus generating the harmonics via the nonlinear interaction between the high electric field of the laser pulse and the medium. Extensive investigations of this phenomenon have been carried out using gas jets, which have resulted in novel coherent soft x-ray sources, with wavelength in the range of few nanometers and pulse duration as short as hundreds of attoseconds. The harmonic spectrum has extended to wavelengths as short as 0.95 nm [1], and pulses as short as 170 as have been measured [2]. However, typical gas harmonics present some disadvantage due to low conversion efficiency. In spite of the high potential for applications of this novel radiation source, very few have been able to exploit its ability to generate nonlinear effects, and results are limited to a few observations of multiphoton absorption processes [3].

Recent investigations have shown that this weakness of gas harmonics can be overcome using ablated plasma as the nonlinear medium [4,5]. An especially interesting observation unique to ablation harmonics is the intensity enhancement of a single harmonic, attributed to resonance with a strong radiative transition [6]. The ablation harmonic technique uses two laser beams; the first subnanosecond long pulse creates the ablation, and the second femtosecond short pulse, which is delayed compared to the first pulse, is focused inside the ablated plasma and creates the harmonics. In this way, conversion efficiencies as high as 10^{-4} from the pump laser to the harmonics have been achieved [6]. However, these experiments used beam splitters to split the prepulse from the main pulse, and thus there was a limit in the range of conditions that we could explore. Further improvements in conversion efficiency will require systematic study of the influence of the various laser parameters on ablation harmonics.

In this paper, we present our experimental studies on the influence of the prepulse and main pulse intensities on the HHG conversion efficiency of a femtosecond laser in silver plasma. We analyze the problem of how to increase the number of harmonic photons, by increasing either the energy of the prepulse or that of the main pulse.

II. EXPERIMENTAL CONFIGURATION

A key point of these studies is the use of two temporally synchronized laser beams from a high-power Ti:sapphire laser system. This allowed us to independently control the characteristics of two pump lasers (prepulse and main pulse) and investigate the conditions for efficient harmonic generation in plasma. For this purpose, we used the 10 Hz, multiterawatt, 35 fs beamline of the Advanced Laser Light Source (ALLS) facility [7]. The output of this beamline was configured into two beams before compression, with each beam having a maximum energy of 200 mJ and pulse duration of 210 ps. Each beam is equipped with a variable energy controller, which can continuously vary the energy of each beam independently using a computer. One of the two beams is sent to a vacuum grating compressor, which compresses the 210 ps pulse to 35 fs with maximum energy of 150 mJ. We name the two beams used in this work line 1 (210 ps duration, 200 mJ maximum energy) and line 2 (35 fs duration, 150 mJ maximum energy).

The generation of harmonics from ablation requires two laser pulses: first, a long pulse to create the ablation, and second, a short pulse to generate the harmonics. In the experiment, laser pulses from line 1 are used as the prepulse, which are first focused on a solid target to create low-ionized ablation. A temporal delay line is introduced into line 2 before compression, and the output pulse is focused onto the ablation as the main pulse, to generate the harmonics. The ablation contains mostly neutral and singly ionized atoms, which interact nonlinearly with the high-intensity electric

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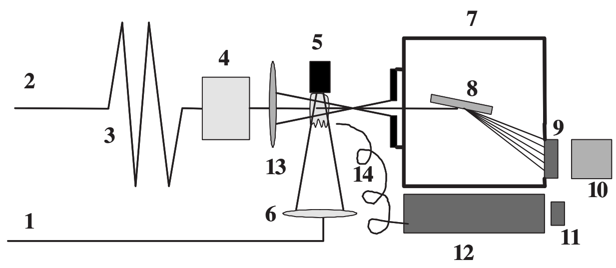

FIG. 1. Schematic of the HHG from plasma. 1, line 1, 2, line 2, 3, delay line; 4, compressor; 5, target, 6,13, focusing lenses; 7, xuv spectrometer; 8, grating; 9, MCP; 10,11, CCDs; 12, uv spectrometer; 14, fiber waveguide.

field of the main pulse. The result of the interaction of the femtosecond pulse with such an ablation is the HHG of the main pump laser, which can be explained by a three-step model [8]. The initial Coulomb potential of a bound electron is lowered by the high-intensity laser field, thus allowing the electron to ionize by the tunnel effect. Then the electron oscillates in the laser field, and eventually the electron recollides with its parent ion. If the collision results in radiative recombination, then the excessive energy is emitted as harmonic radiation.

A schematic diagram of the experimental setup is shown in Fig. 1. To create the ablation, we focused the prepulse from line 1, using a planoconvex lens (focal length $f=150$ mm) on a silver slab target. Then the main pulse from line 2 was focused onto the ablation using a MgF_2 planoconvex lens ($f=680$ mm). The harmonic spectrum is spectrally dispersed by a spectrometer with flat-field Hitachi grating (1200 lines/mm). The spectrum is then detected by a microchannel plate (MCP) with a phosphorus screen readout, and is finally recorded using a charge-coupled device (CCD).

III. RESULTS

Figure 2 shows a typical harmonic spectrum generated in the silver ablation obtained from a single shot. The ablation was produced by focusing a prepulse (2.5 mJ, 210 ps, 800 nm wavelength) onto a silver slab target. The focal spot size was adjusted to 600 μm, corresponding to a prepulse intensity $I_{pp}=0.32\times10^{10}$ W cm^{-2}. After a temporal delay of 100 ns, the high-intensity main pulse (15 mJ, 35 fs, 800 nm wavelength) is focused onto the ablation to generate the harmonics. The spot size of the main pulse at the ablation is adjusted to maximize the harmonic intensity. The optimal position of the focal spot was found to be at the distance of 6 mm before the laser-produced plume. The typical beam waist of the focused main pulse is 200 μm, resulting in a main pulse intensity at the plasma position of $I_{mp}=1.1\times10^{15}$ W cm^{-2}. As demonstrated in Ref. [9], the harmonics from silver ablation have strong similarities with gas harmonics, with a long plateau of harmonic distribution. We found that the ablation harmonics have a perturbative region for relatively low orders, followed by a plateau region, and finally a cutoff at the harmonic wavelength $\lambda=13.5$ nm.

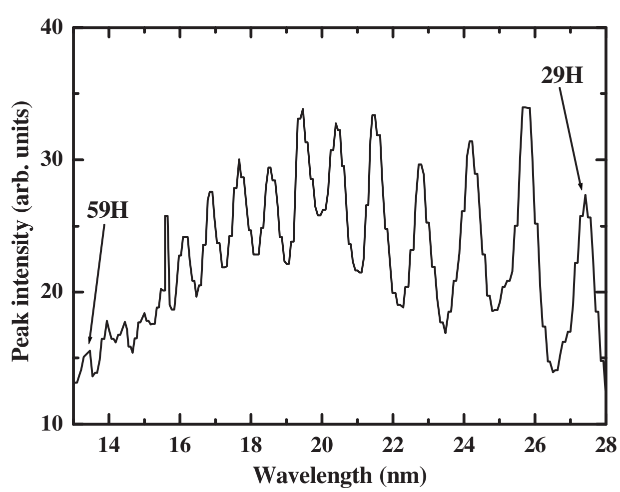

FIG. 2. Example of harmonic spectrum obtained using the silver target.

High-order harmonics (up to the 59th order) and prolonged plateau pattern were observed during these studies. We performed a systematic study of the HHG from the Ag plume to maximize the harmonic efficiency, and cutoff energy. The optimal conditions for this process were created by the weak focusing of the prepulse. In such cases the optimal plasma spectrum in the visible range consisted of the excited Ag I and Ag II lines. At tight focusing conditions and higher intensities of prepulse, the Ag III and Ag IV lines also appeared in the plasma spectrum. Saturation in the conversion efficiency was observed with an increase of the prepulse intensity. This is attributed to the observed generation of multiply charged ions at higher prepulse intensities, and the ionization-induced defocusing of the driving laser radiation due to the generation of a large amount of free electrons in the Ag plume. The important parameter here is the time delay between the prepulse and driving pulse.

Apart from silver, the high-order harmonic spectrum was recorded for other targets, such as In and Sn. Among these targets, the highest harmonic conversion efficiency in the plateau region was obtained in the case of the Ag plasma. We observed a direct relation between the cutoff energy of the harmonics generated from different targets and the second ionization potential of the target atoms, which has underlined the role of the free electrons appearing during further ionization of singly charged particles and leading to the restriction of cutoff energy. This observation also points to the decisive importance of the singly charged ions in the HHG process, which was also confirmed in previous studies of harmonic generation from laser plumes [5,6,9].

A. Effect of prepulse intensity on ablation harmonic spectrum

In this section, we present experimental results investigating the influence of the prepulse intensity on the harmonic intensity. For this experiment, we fixed the intensity of the main pulse and varied that of the prepulse from 2.2×10^{9} to 4×10^{10} W cm^{-2}. For the experimental conditions investigated, we did not observe any significant extension in the cutoff energy with an increase of the prepulse intensity.

There are two ways of representing the harmonic output: in the first method, one takes the maximum value of the peak

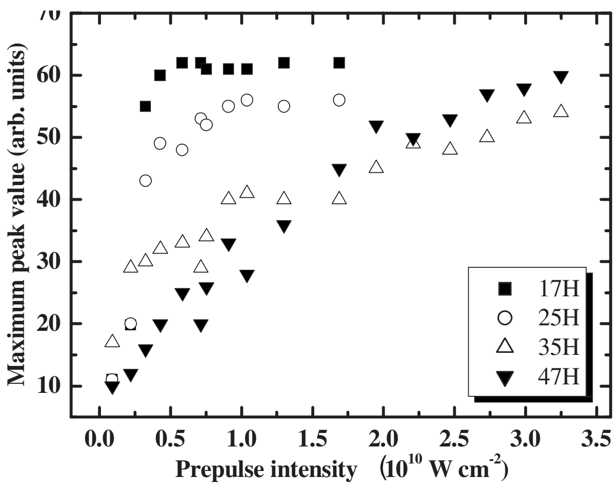

FIG. 3. Peak spectral intensities of the 17th, 25th, 35th, and 47th harmonics as functions of the prepulse intensity at 100 ns delay between the main pulse and prepulse. The main pulse intensity kept constant and was equal to 1.1×10^{15} W cm^{-2}.

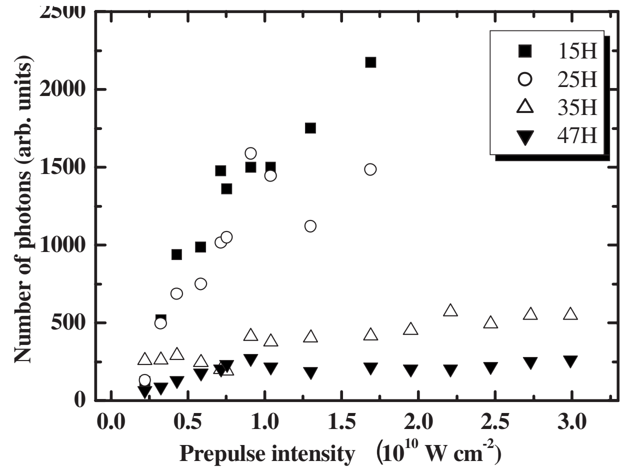

FIG. 4. Integrated harmonic intensities of the 17th, 23rd, 25th, 35th, and 47th harmonics as functions of the prepulse intensity at 100 ns delay between the main and prepulse. The main pulse intensity kept constant and was equal to 1.1×10^{15} W cm^{-2}.

(peak spectral intensity), and in the second method, one integrates the line over the bandwidth, which gives the total number of photons representing the line. In this paper we use both methods, because we observed a difference in the bandwidth of the harmonics under different conditions.

Figure 3 shows the peak spectral intensities of the 17th, 25th, 35th, and 47th harmonics, as a functions of the prepulse intensity, with the main pulse intensity fixed at 1.1×10^{15} W cm^{-2}. The curves in this figure show two tendencies: at low prepulse intensities (less than 0.8×10^{10} W cm^{-2}), the harmonic spectral intensity increases quickly with the prepulse intensity, both for low-order and high-order harmonics. However, whereas the harmonic spectral intensity saturates quickly for the low-order harmonics (in particular, for 17th and 25th orders), that of the higher-order harmonics (35th and 47th) continues to increase, though more gradually. The harmonic intensity of the 35th and 47th harmonics does not grow indefinitely, and they too start to saturate at intensity levels comparable to those of the 17th and 25th harmonics.

However, when we integrate within the bandwidth of each harmonic, the total number of harmonic photons increases with the prepulse intensity in a more gradual way for all orders, but saturates for the higher orders, and do not show a saturation for the lower orders (Fig. 4), contrary to the previous case. We could not determine the saturation limit for lower-order harmonics because of the strong plasma emission at higher prepulse intensity, which makes it difficult to identify the harmonic spectrum.

In fact, when the prepulse intensity is increased, initially the peak spectral intensity of the harmonics also increases, which then saturates at relatively low prepulse intensities for the lower orders of harmonics, and at higher prepulse intensities for the higher orders of harmonics. When one continues to increase the prepulse intensity, the bandwidth of the harmonics widens more significantly for the lower orders, as compared with the higher orders. As a result, the number of harmonic photons increases monotonically with the growth of the prepulse intensity. Figures 5 and 6 justify this observation. In particular, Fig. 6 shows the spectral bandwidths of the 17th, 25th, and 43rd harmonics (full width at half maximum), as functions of the prepulse intensity. The shape of these curves clearly shows that the bandwidth of the lower harmonics increases with prepulse intensity, while those of higher harmonics does not.

To understand this phenomenon, we performed simulations using the hydrodynamic code HYADES [10]. We simulated the expansion of a silver slab interacting with a laser pulse and then determined the electron density, ionization level, and ion density as the functions of the prepulse intensity at a distance of 300 μm from the target surface. These data are presented in Table I.

The following conclusions can be drawn based on the analysis of the experimental spectra, the tendency of the harmonic bandwidth variation, as well as the results of the simulations shown in Table I. First, under our experimental conditions (silver target, 100 ns delay, main pulse intensity 1.1×10^{15} W cm^{-2}), for prepulse intensities lower than 0.9×10^{10} W cm^{-2}, the ionization level of the plasma remains lower than 1 but higher than 0.5. Consequently, the ablation contains singly ionized and neutral atoms, which generate the harmonic spectrum. Therefore, for this prepulse intensity range, as the prepulse intensity is increased with the main pulse relatively weak and constant, one increases the density

TABLE I. Results of the calculations of silver plasma characteristics at different prepulse intensities.

Prepulse intensity (10^{10} W cm^{-2})	0.3	0.59	0.9	1.3	1.95	2.6	3.25
Electron density (10^{17} cm^{-3})	0.89	2.56	3.2	4.0	5.79	6.33	8.0
Ionization level	0.5	0.7	1.01	1.17	1.3	1.39	1.58
Ion density (10^{17} cm^{-3})	0.89	2.56	3.17	3.41	4.45	4.55	5.06

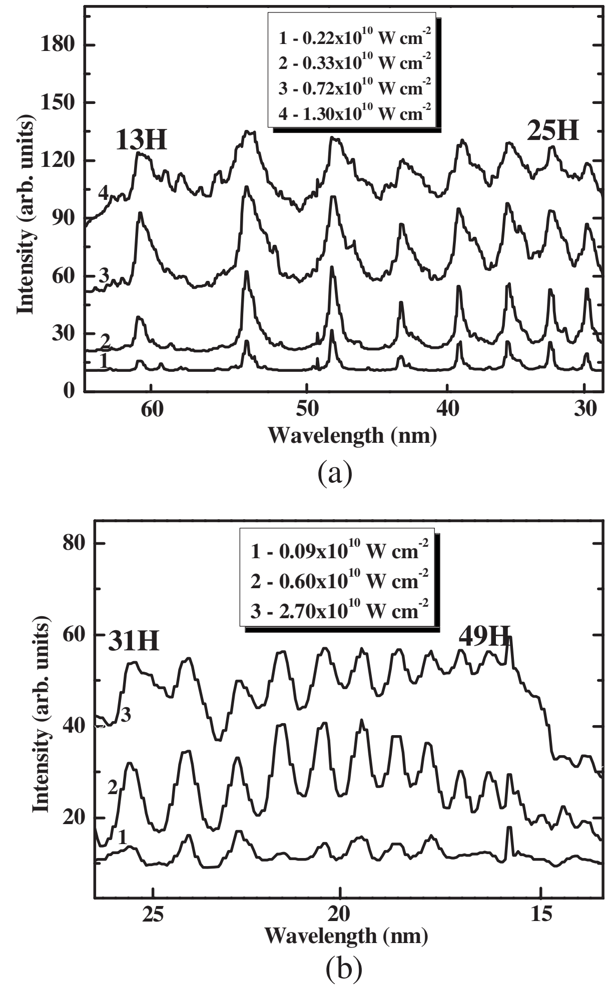

FIG. 5. Harmonic spectra obtained in silver target at different values of prepulse intensity. (a) Low-order and (b) high-order harmonic spectra.

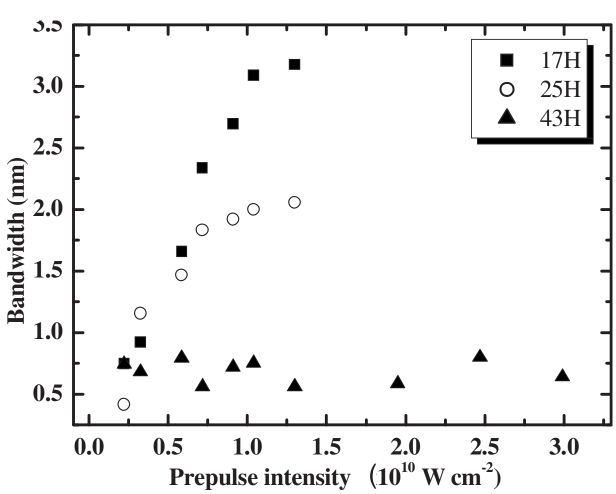

FIG. 6. Bandwidths of 17th, 25th, and 43rd harmonics as functions of prepulse intensity.

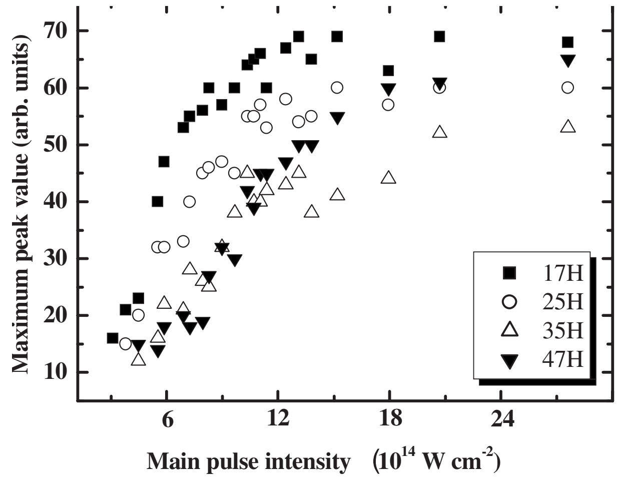

FIG. 7. Peak spectral intensities of the 17th, 25th, 35th, and 47th harmonics as functions of the main pulse intensity. Prepulse intensity is 0.6×10^{10} W cm^{-2} for 17th and 25th harmonics, and 1.69 $\times10^{10}$ W cm^{-2} for the 35th and 47th harmonics.

of singly charged ions, and consequently the number of harmonic photons. For prepulse intensity between 0.9×10^{10} and 3×10^{10} W cm^{-2}, the ionization level becomes higher than 1. The plasma in that case contains increasingly more singly ionized atoms and a small amount of doubly charged atoms. The harmonics are emitted from the singly charged ions as well as from the weak contribution of doubly charged ions, from which distinct spectral broadening is observed for the lower orders, while the higher-order harmonic spectrum continues to increase. For intensities exceeding 3 $\times10^{10}$ W cm^{-2}, the ionization level becomes higher than 1.5, thus creating equally both singly charged and doubly charged ions. The growth of free-electron concentration in this case prevents efficient harmonic generation due to self-defocusing and self-modulation of the femtosecond pulse.

B. Effect of the main pulse intensity on ablation harmonics

In this section, we present our studies of the influence of the main pulse intensity on the harmonic intensity. For this purpose, we fixed the intensity of the prepulse, and varied the main pulse intensity from 3×10^{14} to 3.2×10^{15} W cm^{-2}. Similar to the studies on varying the prepulse intensity, we did not observe any significant extension in the cutoff energy with an increase of the main pulse intensity. As soon as the main pulse and the prepulse intensity reach conditions adequate for generating high-order harmonics, a stable cutoff is reached.

The change in the harmonic intensity as a function of the main pulse intensity has a similar tendency to those reported in Sec. III A. The number of harmonic photons increases with the main pulse intensity, after which the peak spectral intensity for all the harmonic orders then saturates (Fig. 7). However, if one integrates within the harmonic bandwidth, the number of photons increases with the main pulse intensity, but one also notes a tendency to saturate for the higher orders (Fig. 8). These observations are also supported by the results presented in Figs. 9 and 10.

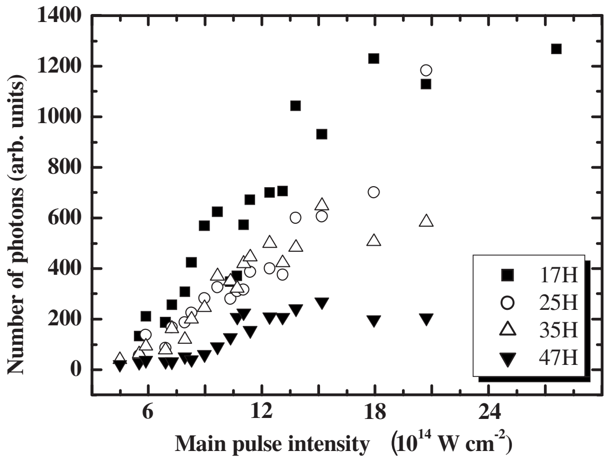

FIG. 8. Integrated harmonic intensities for the 17th, 25th, 35th, and 49th harmonics as functions of the main pulse energy. Prepulse intensity is 0.6×10^{10} W cm^{-2} for 17th and 25th harmonics, and 1.69×10^{10} W cm^{-2} for the 35th and 47th harmonics.

As in the case of the data presented in Sec. III A, a broadening of the spectra of low-order harmonics with an increase in the main pulse intensity was observed. However, the spectral broadening is less distinct, and is accompanied by a redshift of the harmonic spectrum. This phenomenon, which has also been observed in gas harmonics [11], is attributed to the shift of the spectrum of the main pulse due to the high level of ionization of the medium created by the main pulse itself. Indeed, the high electron density of the plasma involves a variation of the index of refraction for the propagation of the harmonics in plasma [12].

The question that one can now put to the test of this study is as follows: what is the best way to increase the number of harmonic photons, either increasing the energy of the prepulse or that of the main pulse? To answer this question we show in Fig. 11 the number of harmonic photons generated according to the energy variations of prepulse and main pulse. These dependences for low- (15th-) and high- (47th-) order harmonics enables us to conclude that, when one has weak laser pulse energy (less than 17 mJ), it is preferable to put more energy on the prepulse because that will produce more harmonic photons than the same energy in the main pulse. For example, the pair (12 mJ prepulse and 5 mJ main pulse) would be more efficient than (5 mJ prepulse and 12 mJ main pulse). But if one has much laser energy, it is enough to increase the energy of the main pulse (higher than 17 mJ) at a reasonable energy of the prepulse (approximately 7 mJ) to reach the point of saturation. It is also important to note that there is a limit in the increasing of the prepulse energy because a high energy of the prepulse produces high plasma emission. This also stops the harmonic generation because of the high electron density in plasma, which defocuses the main pulse [13], decreases its intensity, and creates the multi-ionized particles. This limit depends also on the main pulse intensity, i.e., if the main pulse intensity is high, then the limit is low.

The highly ionized medium, with higher electron density in the center than in the outer region, acts as a negative lens, leading to a defocusing of the laser beam in a plasma and

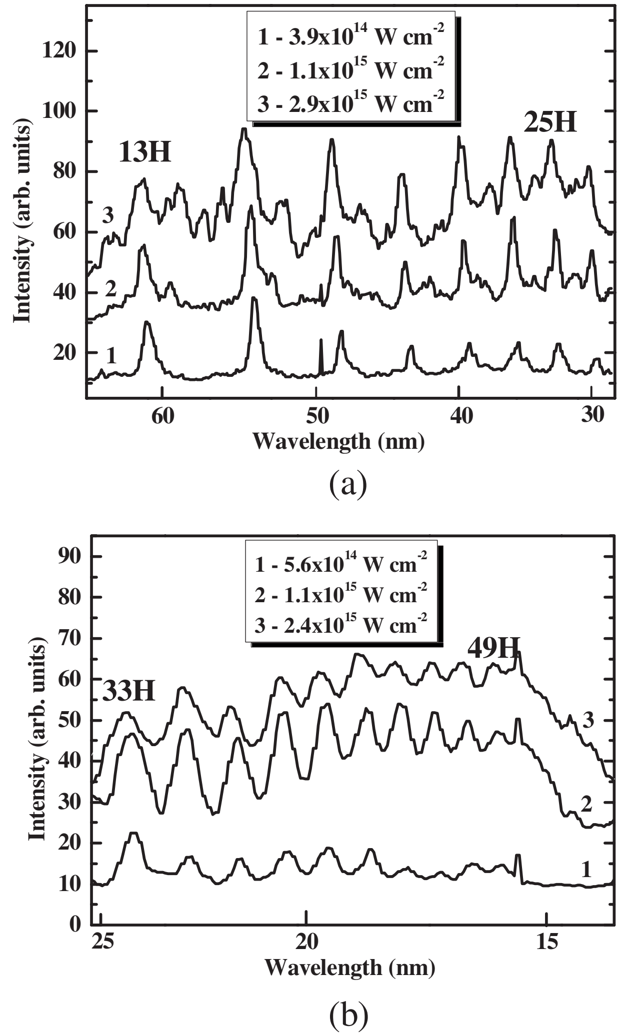

FIG. 9. Harmonic spectra obtained from silver plume at different values of main pulse intensity. (a) Low-order and (b) high-order harmonics.

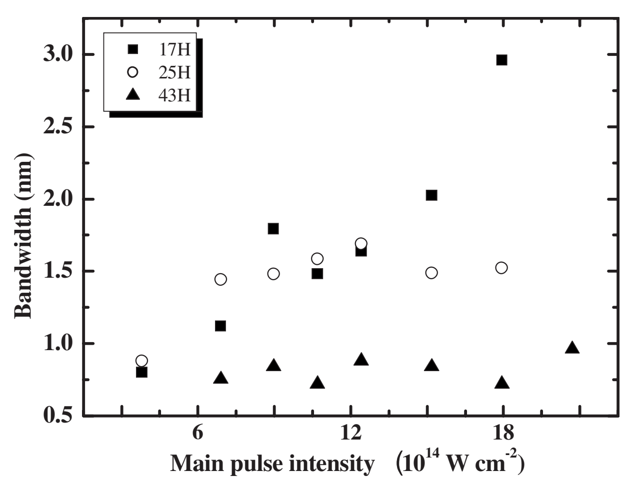

FIG. 10. Bandwidths of 17th, 25th, and 43rd harmonics as functions of main pulse intensity.

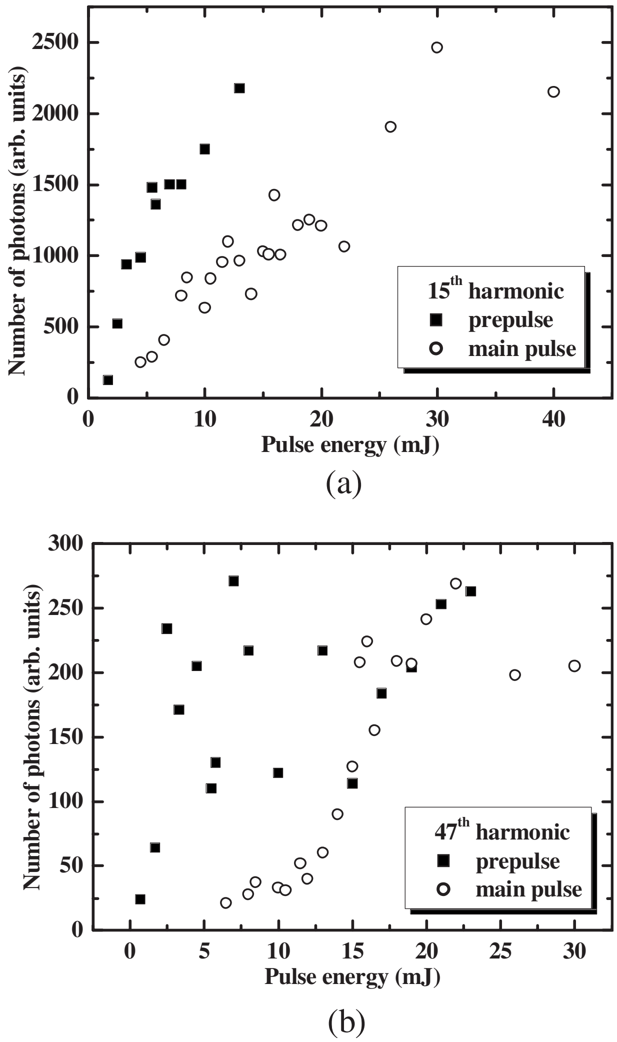

FIG. 11. Integrated harmonic intensities of the (a) 15th and (b) 47th harmonics as functions of the prepulse and main pulse energy.

hence to a reduction in the effective harmonic generation volume. In addition, the rapidly ionizing high-density medium modifies the temporal structure of the femtosecond laser pulse due to self-phase-modulation (SPM). We maintained the conditions when no significant ionization of the plasma by the main laser pulse took place, by keeping the laser intensity in the vicinity of the plume below the barrier suppression intensity of singly charged ions.

It was previously shown during laser–gas-jet studies that, at high laser intensities, the spectral structure of high-order harmonics contains chirp components, i.e., positive chirp due to the SPM of the driving laser pulse propagating through an ionizing medium [14] as well as a dynamically induced negative chirp [15]. Since the chirp of the driving radiation results in a broadening of the harmonic spectrum, it should be appropriately compensated for to achieve sharp harmonics and high brightness of the coherent xuv radiation.

We did not measure the absolute value of conversion efficiency of harmonic yield in the plateau range in the case of a Ag-containing plume. However, taking into account the past work [9] and comparing the spectrum obtained, we can conclude that conversion efficiencies of about 8×10^{-6}, similar to those obtained in past work, have been achieved in this work.

IV. DISCUSSION

Previously, Ag plasma has shown the highest conversion efficiency (8×10^{-6}) and cutoff [9] among other plasma samples. The question arises why such a strong harmonic efficiency was obtained in the plateau range in the case of Ag plasma, while for the plumes from other targets it was almost 10–80 times less. After defining the parameters that are responsible for efficient HHG conversion (atomic number, ionization potentials, etc.), there is one suggestion that can be helpful for understanding the difference between the results obtained in the cases of silver and other plumes.

The feature related with target plasma is a possible appearance of nanometer-sized fragments upon irradiation by a strong pulse. Such clusters were extensively studied with respect to their unique frequency conversion properties [16–18]. In some of these studies it has been shown that clusters can be used to reach conversion efficiencies comparable to those obtained in solid state systems. The efficient HHG in clustered media can be achieved at smaller pump intensities due to local-field-induced enhancement of nonlinear susceptibility, while a higher cutoff can be realized compared to monatomic plasma. The use of the surface plasmon resonance of nanosized particles can considerably enhance the nonlinear optical response of such systems. Our observations of high conversion efficiency in a silver plasma can be related to quasiresonance conditions between the second harmonic of the fundamental radiation (Ti:sapphire laser, 800 nm) and the surface plasmon resonance of silver nanoparticles (∼400 nm). Further studies are needed for the analysis of the influence of the properties of the clusters produced during plasma formation on HHG conversion efficiency.

The characteristic range of conversion efficiencies achieved in the plateau region in the case of HHG from gas jets did not much exceed 10^{-6} in early experiments. Since then, the conversion efficiency has been improved up to 10^{-5} by proper phase matching [19]. The HHG from initial laser plasma experiments shows conversion efficiency comparable with those achieved in the case of HHG from gas jets. However, plasma harmonics have demonstrated considerable increase in the efficiency of a single harmonic (as high as 10^{-4}), that exceeds those for the HHG from gases and even the HHG from laser-surface experiments in the xuv region [6,20,21]. Though we did not observe enhancement of a single harmonic in silver plasma, the conversion efficiency for silver was higher than many previously reported data on the HHG from gas jets.

In our recent work [22], we observed a correlation between the cutoff harmonics (H) generating from different targets and the ionization potentials (I_i) of the atoms participating in HHG, which was close to data previously reported in [23]. For most targets, a linear relation was observed between the second ionization potential and the cutoff energy, implying the important role of singly charged ions for harmonic generation from a plasma plume. We confirmed this relation by comparing the harmonic cutoffs using main pump Ti:sapphire lasers with different pulse duration (35, 48, and 150 fs). This dependence shows a linear relation between H and I_i for laser plumes where a harmonic plateau was observed. The dependence is represented by the empirical relation of H (harmonic order) $\approx 4I_i$ (eV)-32.1. From this relation, we can draw the next conclusion. When singly charged ions are strongly involved in the process of HHG from a plasma plume, maximum harmonics are generated with targets that have higher second ionization potentials. This means that the generation of additional free electrons by further increasing the main pump intensity, due to the ionization of singly charged ions, leads to the saturation of the $H(I_{fp})$ dependence, thus restricting the generation of higher-order harmonics.

The analysis of the role of neutrals and ions in the HHG from laser ablation is presented in [9]. Here we briefly repeat the discussion of the role of ions in this process. A three-step model does not explain the observed results with neutrals. The maximum harmonic order that can be achieved is defined by the atomic ionization potential, which is rather low ($I_1=7.58$ eV) for silver. Therefore the neutral Ag atoms can only generate harmonics with a maximum order of 7 ($I_1+3.17U_p=11$ eV; U_p is the ponderomotive potential that corresponds to the energy of free electron in the field of the electromagnetic wave). The calculations of the saturation intensity (4×10^{14} W cm^{-2}) for singly charged Ag ions (second ionization potential $I_{p2}=21.48$ eV), which was close to the observed saturation of the $H(I_{fp})$ dependence, confirm the consideration of the ions as a main source of harmonics in these experiments.

V. CONCLUSIONS

By using the 20 TW, 10 Hz output of the ALLS laser, we were able to study independently the influence of the prepulse and main pulse intensity on the harmonic spectrum obtained from silver ablation. We used silver ablation, since this target has previously shown high conversion efficiency for high-order harmonics. From this study we found that, at our experimental conditions, it is preferable to work with prepulse intensities below and in the range of 1×10^{10} W cm^{-2}. This value was estimated from Table I, showing the appearance of doubly charged ions in the laser plume at the prepulse intensities exceeding this level.

The study of the influence of the main pulse intensity on the harmonic spectra has shown that it is important to optimize this parameter for efficient harmonic generation. We have shown that the presence of doubly ionized atoms in the ablation, created either by a strong prepulse intensity or with the irradiation of the main pulse, is ineffective for the generation of harmonics. We identified the optimal conditions for the enhancement of low- and high-order harmonics at appropriate prepulse and main pulse intensities, and clarified the optimum method of maximizing the harmonic intensity for modest and high-energy pump lasers.

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