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Intense harmonic generation from silver ablation

T. OZAKI,[1] L.B. ELOUGA BOM,[1] R. GANEEV,[1] J.-C. KIEFFER,[1] M. SUZUKI,[2] AND H. KURODA[2]
[1]INRS-EMT, Université du Québec, Varennes, Québec, Canada
[2]The Institute for Solid State Physics, The University of Tokyo, Kashiwa, Chiba, Japan

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Abstract

We perform systematic investigation of a new method for generating high-order harmonics, using ablation as the nonlinear medium. We place emphasis on clarifying the pump laser conditions for optimum harmonic yield. For this purpose, we use 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 intensity. We show here that the presence of doubly ionized atoms in the ablation, created either by a strong prepulse intensity or by irradiating the main pulse, will suppress harmonic generation.

Keywords: Ablation; High harmonic generation

1. INTRODUCTION

High-order harmonic generation has become a unique technique for generating extreme radiation sources, with wavelength in the range of a few nanometers and pulse duration as short as 100s of attoseconds (Baeva *et al.*, 2007; Ozaki et al., 2006; Kuroda *et al.*, 2005). Conventional harmonic generation uses gas as the nonlinear medium, in which case the harmonic spectrum has been extended to wavelengths as short as 0.95 nm (Seres *et al.*, 2005), and pulses as short as 170 attoseconds have been measured (Lòpez-Martens *et al.*, 2005). However, the energy of typical gas harmonics is limited, because of low conversion efficiency. Consequently, few have been able to exploit its ability to generate nonlinear effects, with a few observations of multi-photon absorption processes (L'Huillier *et al.*, 2003).

An alternative method recently proposed is the use of ablation as a source of gaseous atoms. Ablation is used in various applications, such as the production of low-energy low-divergence pulsed indium atomic beams (Ali & Khare, 2006*a*), the generation of sculpted pulsed atomic beams of regular arrays in one and two-dimensions (Ali & Khare, 2006*b*), and measurements of surface impurities on metallic-foil targets (Fernandez *et al.*, 2005). We have proposed and demonstrated the use of ablation as the nonlinear medium for high-order harmonic generation. The advantage of this method compared with gas is that (1) it allows the use of any material that can be formed into solid targets, (2) it is capable of generating media with higher density, and (3) it can easily generate media with longer lengths and with variable conditions along the media. Such characteristics should provide a higher degree of freedom in the optimization of high-order harmonic generation (HHG). There has been several works in the past that have investigated this method (Akiyama *et al.*, 1992; Krushelnick *et al.*, 1997; Theobald *et al.*, 1995; Kubodera *et al.*, 1993; Wahlström *et al.*, 1995), in which a relatively low-intensity laser pulse was used to generate a plume medium on solid targets such as lithium, sodium, potassium, carbon, and aluminum. However, the highest harmonic order observed in these works was limited to the 27th (Wahlström *et al.*, 1995), which was generated by a 150 fs Ti:Sapphire laser pulse focused into either a sodium or potassium ablation plume. Interestingly, no plateau in the harmonics was observed in any of these works.

Recent investigations have shown that conversion efficiencies higher than that of gas harmonics can be achieved using solid ablations as the nonlinear medium (Ganeev *et al.*, 2005). Furthermore, we have discovered an interesting phenomenon with indium ablation harmonics, where quasi-monochromatic spectrum with a single harmonic order dominating the spectrum has been demonstrated (Ganeev *et al.*, 2006*a*). This phenomenon has been attributed to resonance effects with a strong radiative transition. The ablation harmonic technique uses two laser beams, the first picosecond long

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Address correspondence and reprint requests to: Tsuneyuki Ozaki, INRS-EMT, Université du Québec, Varennes, Québec, Canada. E-mail: ozaki@emt.inrs.ca

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pulse creates the ablation, and a second femtosecond short pulse, which is delayed relative to the first pulse, is focused inside the ablated plasma, and creates the harmonics. With this method, conversion efficiencies as high as 10^{-4} from the pump laser to the harmonics have been achieved (Ganeev *et al.*, 2006*a*, 2006*b*). However, these experiments uses 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 perform comprehensive investigations of silver ablation harmonics, by systematically varying the pump laser conditions and studying their effects on the harmonic intensity.

2. EXPERIMENTAL SETUP

A key point of this investigation is the use of two temporally synchronized laser beams from a high-power Ti:Sapphire laser. This allowed us to control independently the characteristics of the two pump lasers (prepulse and main pulse), and to study the conditions for efficient harmonic generation in ablation. For this, we used the 10 Hz, multi-TW, 25 fs beam line of the advanced laser light source (ALLS) facility at the Institut National de la Recherche Scientifique (INRS). The output of this beam line was configured into two beams before compression, with each beam having a maximum energy of 200 mJ, and pulse duration of 210 ps. The energy of each beam can be continuously and independently controlled.

In this experiment, one of the two uncompressed beams is sent to a vacuum-grating compressor, which compresses the 210 ps pulse to 25 fs with maximum energy of 150 mJ. In this paper, we name the beam line providing the uncompressed pulse as Line #1 (210 ps duration, 200 mJ maximum energy), and that providing the compressed beam as Line #2 (25 fs duration, 150 mJ maximum energy). The laser pulses from Line #1 is used as the prepulse, which is first focused on a solid target to create low-ionized ablation. A temporal delay line is introduced into Line #2 before compression, and we use this output as the main pulse, which is focused on to the ablation to generate the harmonics. The ablation contains mostly neutral and singly ionized atoms, which interacts with the high intensity electric field of the main pulse to generate the harmonics.

We show in Figure 1 a schematic diagram of the experimental setup. One of the beams is used without compression (200 ps pulse duration) as a prepulse for creating ablation, which is focused onto the solid target placed inside a vacuum. The second beam is used as the main pulse for harmonic generation. The main pulse is delayed in time relative to the prepulse by propagating through an optical delay line, and then sent through a pulse compressor. The compressed pulse has typical pulse duration of 30 fs full-width at half-maximum (FWHM), which is then focused onto the ablation medium using MgF_2 lens ($f = 680$ mm). Since the intensity of the prepulse ($\sim 10^{10}$ Wcm^{-2}) is near the threshold of ablation, the ablation depth of each shot was not large. Therefore, we were able to observe harmonics without translating the target for 3000 to 6000 shots. The maximum energy used in this experiment was 30 mJ, due to the relatively poor vacuum in our target chamber, which was at 10^{-5} torr. This prevented us from opening the gate valve between the compressor and the target chamber, which in turn limited the energy that can be passed through the glass window of the gate valve, due to white light generation.

The high-order harmonics generated from the ablations were spectrally resolved using a flat-field grazing-incidence extreme ultra-violet (XUV) spectrometer with a Hitachi 1200-grooves/mm grating. The XUV spectrum was detected by a micro-channel plate with phosphor screen and recorded by a charge coupled device (CCD) camera. Ablation harmonic experiments were performed with silver and indium targets. In our experiments, each harmonic spectrum was

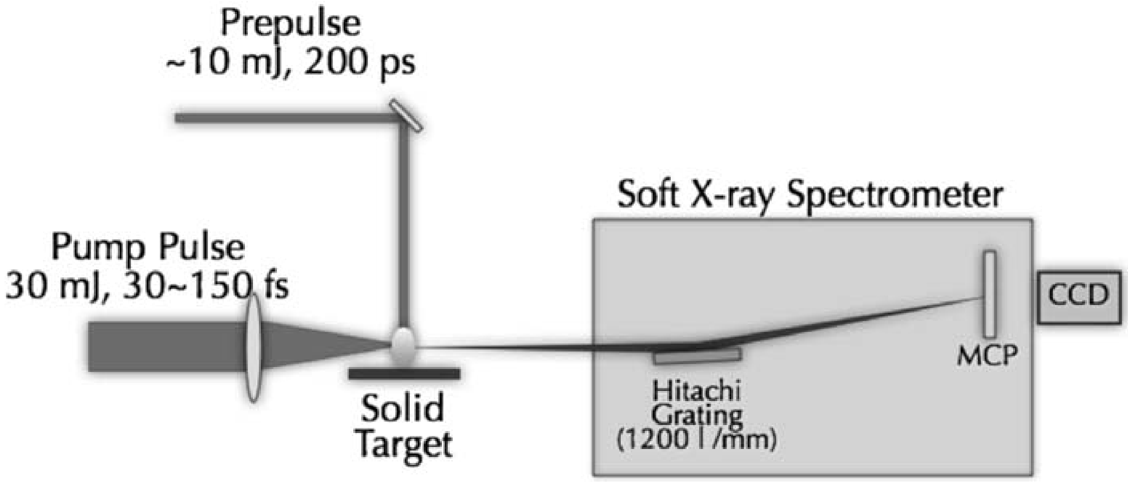

Fig. 1. Schematic diagram of the experimental setup for generating high-order harmonics from laser ablation. Mp: main pulse, Pp: prepulse, G: grating, MCP: micro-channel plate, CCD: charge-coupling device.

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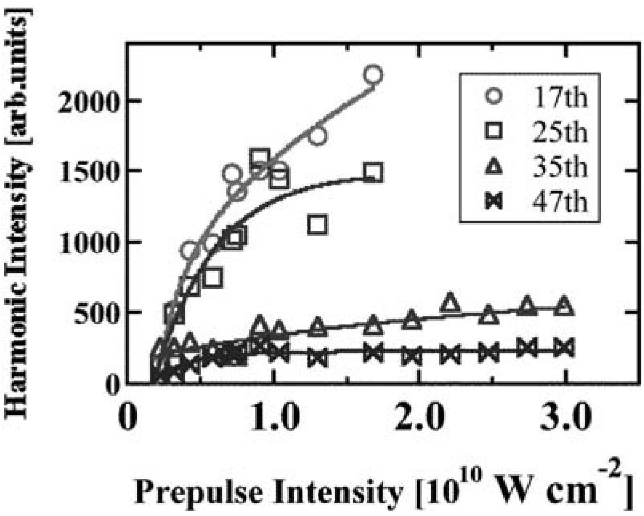

Fig. 2. Integrated harmonic intensities of the 17th, 23rd, 25th, 35th, and 47th harmonics as functions of the prepulse intensity at 100 ns delay between the main and prepulse. The main pulse intensity kept constant and was equal to 1.1×10^{15} Wcm^{-2}.

taken from a single shot, without the need for accumulation. Absolute conversion efficiency of the harmonics were measured by first comparing the third harmonics generated by a nonlinear crystal with those produced in air breakdown, measured using a vacuum monochromator (Acton VM-502) and photomultiplier tube with sodium salicylate plate used as the phosphor. This gave us the relation between the energy of the third harmonic and the photomultiplier tube signal. Next, the third to the 11th harmonics were calibrated based on the photomultiplier signal obtained using the vacuum monochromator. Finally, efficiency between the lower harmonics and the higher harmonics were calculated based on the spectral response of sodium salicylate to extreme ultraviolet radiation, and by comparing the photomultiplier signal and the MCP readout of the 11th harmonic.

3. RESULTS AND DISCUSSION

3.1. Effect of prepulse intensity

Typical harmonic spectrum generated in silver ablation can be obtained from a single shot. The ablation was produced by focusing a prepulse (2.5 mJ, 210 ps, and 800 nm wavelength) onto a silver slab target. The focal spot size was adjusted to 600 μm, corresponding to a prepulse intensity $I_{pp} = 0.32 \times 10^{10}$ Wcm^{-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. Typical beam waist of the focused main pulse is 200 μm, resulting in a main pulse intensity at the plasma position to be $I_{mp} = 1.1 \times 10^{15}$ Wcm^{-2}. 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 cut-off at the harmonic wavelength $\lambda = 13.5$ nm.

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 silver plume to maximize the harmonic efficiency, and cut-off 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 silver I and silver II lines. At tight focusing conditions and higher intensities of prepulse, the silver III silver 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 Silver plume. The important parameter here is the time delay between the prepulse and driving pulse.

Next, we investigated 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} Wcm^{-2} to 4×10^{10} Wcm^{-2}. Figure 2 shows the integrated spectral intensities of the 17th, 25th, 35th, and 47th harmonics, as a function of the prepulse intensity, with the main pulse intensity fixed at 1.1×10^{15} Wcm^{-2}. The curves in this figure show two tendencies: at low prepulse intensities (less than 0.8×10^{10} Wcm^{-2}), the harmonic spectral intensity increases gradually with the prepulse intensity, both for low-order and high-order harmonics. However, the harmonic spectral intensity shows saturation 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. We could not determine the saturation limit for lower-order harmonics because of the strong plasma emission at higher prepulse intensity, which made it difficult to identify the harmonic spectrum.

To understand this phenomenon, we performed simulations using the hydrodynamic code HYADES (Rubenchik *et al.*, 1998). We simulated the expansion of silver ablation produced by a laser pulse. From this simulation, we determined the electron density, ionization level, and ion density as a function of the prepulse intensity at 300 μm from the target surface. The results of this simulation are shown in Figure 3. From the results of this simulation, coupled with experimental data, the following conclusions could be drawn. First, under our experimental conditions (silver target, 100 ns delay, main pulse intensity 1.1×10^{15} Wcm^{-2}), for prepulse intensities lower than 0.9×10^{10} Wcm^{-2}, the ionization level of the plasma remains

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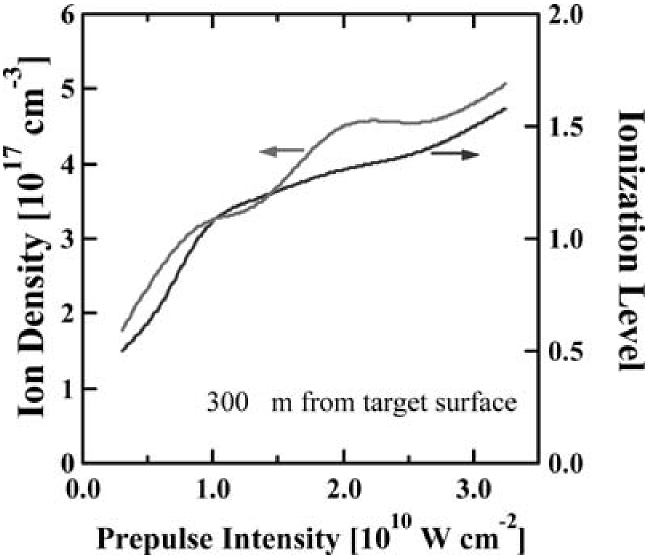

Fig. 3. Calculated ion density and ionization level of the silver ablation, as a function of the prepulse intensity.

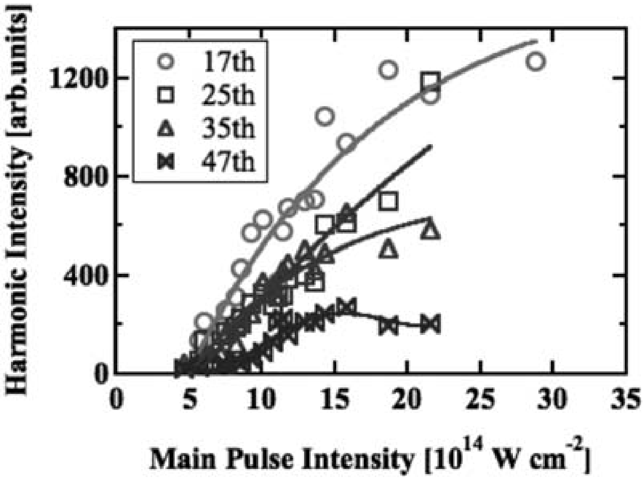

Fig. 4. Integrated harmonic intensities for the 17th, 25th, 35th, and 49th harmonics, as the functions of the main pulse energy. Prepulse intensity is 0.6×10^{10} Wcm^{-2} for 17th and 25th harmonics, and 1.69×10^{10} Wcm^{-2} for the 35th and 47th harmonics.

lower than 1 but higher than 0.5. Thus, 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 kept constant), one increases the density of singly charged ions, and thus the number of harmonic photons. For prepulse intensity between 0.9×10^{10} Wcm^{-2} and 3×10^{10} Wcm^{-2}, the ionization level becomes higher than 1. Thus the plasma contains more singly ionized atoms and small amount of doubly charged atoms. In this case, the harmonics are emitted from the singly charged ions as well as from the weak contribution of doubly charged ions. In this case, the lower-orders show distinct spectral broadening, while the higher-order harmonic spectrum continues to increase in peak spectral intensity. For intensities exceeding 3×10^{10} Wcm^{-2}, the ionization level becomes higher than 1.5, thus creating both singly charged as well as doubly charged ions. This condition results in the generation of intense plasma continuum for both low-order and high-order harmonics.

3.2. Effect of the main pulse energy

In this section, we present our studies on 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} Wcm^{-2} to 3.2×10^{15} Wcm^{-2}. The change in the harmonic intensity as a function of the main pulse intensity has a similar tendency with those reported in Section 3.1. The number of photons increases with the main pulse intensity, but one also notes a tendency to saturate for the higher-orders (Fig. 4). As with the data presented in Section 3.1, we observed a broadening of the spectra of low-order harmonics with an increase in the main pulse intensity. However, the spectral broadening is less distinct, and there is a red shift of the harmonic spectrum. This phenomenon, which has also been observed in gas harmonics (Altucci *et al.*, 2000), 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 (Wahlström *et al.*, 1995).

From the above results, we can evaluate if it is more beneficial to increase the energy of the prepulse or that of the main pulse. These dependences for low (15th) – and high (47th) -order harmonics on the prepulse and main pulse energy enables us to conclude the following. When one has limited laser pulse energy available (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, harmonic intensity would be higher for 12 mJ prepulse and 5 mJ main pulses, compared with that for 5 mJ prepulse and 12 mJ main pulses. However, if one has more pump laser energy available, it is enough to reach saturation in the harmonic intensity, by using prepulse energy of 7 mJ in combination with sufficient main pulse energy (higher than 17 mJ). It is also important to note that there is an upper limit in the prepulse energy, because a high prepulse energy produces high plasma emission. This also stops the harmonic generation because of the high electron density in plasma, which defocuses the main pulse (Ganeev *et al.*, 2006*b*), decreases its intensity, and creates the multi-ionized particles. This limit depends also on the main pulse intensity, which also produce unwanted ionization within the ablation.

If the ablation is highly ionized, with higher electron density in the center than in the outer region, the ablation acts as a negative lens. This in turn leads to a defocusing

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of the laser beam in plasma, and so a reduction in the effective harmonic generation volume. In addition, the rapidly ionizing high-density medium modifies the temporal structure of femtosecond laser pulse due to self-phase modulation. In our experiments, we took care so that the main pulse did not result in significant ionization of the plasma, by keeping the laser intensity near the barrier suppression intensity of singly charged ions.

4. CONCLUSIONS

By using the 20 TW, 10 Hz beam line of the advanced laser light source, 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 the range of 1×10^{10} Wcm^{-2}. Simulations show that this intensity is related to the threshold intensity for producing doubly charged ions in the ablation.

The study of the influence of the main pulse intensity on 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 the strong prepulse intensity or by irradiating the main pulse, is ineffective for the generation of harmonics. We identified the ideal conditions for enhancing low-order and high-order harmonics at proper 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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