Proceedings of the 3rd International Conference on the Frontiers of Plasma Physics and Technology (PC/5099)

Extending the Capabilities of Ablation Harmonics to Shorter Wavelengths and Higher Intensity

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Abstract: We study the generation of high-order harmonics from ablation plume, by using the 20 TW, 10 Hz laser of the Advanced Laser Light Source (ALLS). We perform detailed studies on enhancement of single high-order harmonics generated in laser plasma using the fundamental and second harmonic of the ALLS beam line. We observe quasi-monochromatic harmonics for Mn, Cr, Sb, Sn, and In plasmas. We identify most of the ionic/neutral transitions responsible for the enhancement, which all have strong oscillator strengths. We demonstrate intensity enhancements of the 13^{th}, 17^{th}, 21^{st}, 29^{th}, and 33^{rd} harmonics from these targets using the 800 nm pump laser and varying its chirp. We also observed harmonic enhancement from some targets for 400 nm pump laser. Using Mn plume, we demonstrated the highest harmonic photon energy (52.9 eV) at which enhancement has been observed (17^{th} order, $\lambda = 23.5$ nm).

1. Introduction

High-order harmonics is a distinct source of coherent soft x-rays, with unique capabilities to generate ultrashort pulses on the order of 100 attoseconds. However, conversion efficiency of high-order harmonics is a key issue when considering real applications. One successful approach to overcome this challenge has been to phase match the pump and the harmonics using gas-filled waveguides [1]. An alternative approach is the possibility to enhance harmonic generation using atomic resonances [2]. The challenge to achieve intensity enhancement of high-order harmonic generation (HHG) in gaseous media using atomic and ionic resonances has been studied, and both theoretical and experimental reports [3-8] have shown the perspectives of this approach. Intensity enhancement of some harmonic orders has been reported in laser-gas jet interaction. Further, by optimizing the laser pulse shape, Bartels et al [8] were able to increase the 27^{th} harmonic in Ar by more than an order of magnitude. Recently, generation of arbitrary shaped spectra of high harmonics by adaptive control of the pump laser pulse in laser-gas jet experiments was demonstrated [9]. However, in these studies, the intensity of neighboring harmonics was comparable with the enhanced harmonics.

Recent investigations of HHG from plasma plume imply an alternative approach. The method capitalizes on the efficient harmonic generation from low-ionized ions produced on the surfaces of various solid-state targets [10-12]. By using plumes produced from specific materials, coincidental overlap between the harmonic wavelength and a strong radiative transition of ions can lead to notable increase in the harmonic yield. By using solid target atoms for HHG, there is the possibility to explore resonance enhancements with materials that were not accessible in the past. Recently, we have reported observing intensity enhancement of a single harmonic in the plateau region [13-16]. In particular, the 80-times intensity enhancement of the 13^{th} harmonic ($\lambda = 61.2$ nm) of Ti:sapphire laser pump was demonstrated, using indium plasma as the nonlinear medium, and by varying the spectrum of the pump laser [13]. Plasma plume of GaAs and InSb also showed enhancement of single harmonics at different harmonic orders. Presently, the highest photon energy of intensity-enhanced harmonic was achieved in chromium plasma (29^{th} harmonic, $\lambda = 27.4$ nm, $E_{ph} = 45.4$ eV).

Currently, intensity enhancement of single harmonics has been limited to low- to middle-orders. Therefore, an important direction would be to further extend the photon energy at which such intensity enhancements can be realized. Such studies would pave the way for creating intense, quasi-monochromatic source of coherent extreme ultraviolet (XUV) radiation in the water-window, a spectral regime important for biomedical imaging. In this paper, we demonstrate the active control of intensity enhancement of single harmonics using plumes of various materials, by varying the chirp of the pump laser. We studied targets such as In, Sn, Sb, Cr, and Mn, and were able to demonstrate intensity enhancement of the 13^{th}, 17^{th}, 21^{st}, 29^{th}, and 33^{rd} harmonics of the 800 nm pump laser, respectively. Such enhancement always occurred when the wavelength of the harmonic was spectrally close to a strong radiative transition with large oscillator strengths. We compare our data with previously reported results of the studies of some of these samples. We also demonstrate harmonic enhancement for several targets using frequency-doubled pump lasers (400 nm wavelength). In this case, the maximum order at which intensity enhancement is observed is for the Mn plasma (17^{th} order, λ = 23.5 nm), which is also the highest photon energy (E_{ph} = 52.9 eV) at which intensity enhancement has been demonstrated.

2. Experimental setup

To create the ablation, we focused a prepulse from the uncompressed Ti:sapphire laser (210 ps, 800 nm, 10 Hz) on to a target placed in a vacuum chamber, by using a plano-convex lens (focal length f = 150 mm). The focal spot diameter of the prepulse beam on the target surface was adjusted to be about 600 μm. The intensity of this sub-nanosecond prepulse, I_{pp}, on the target surface was varied between 7×10^9 W cm^{-2} to 4×10^{10} W cm^{-2}. This range of prepulse intensity variations was defined from previous studies of different ablated targets. After a delay between 50 and 80 ns, part of the femtosecond main pulse (E = 8 to 25 mJ, t = 35 fs, λ = 800 nm central wavelength, 40 nm bandwidth FWHM) was focused on the plasma from the orthogonal direction by using a MgF_2 plano-convex lens (f = 680 mm). The maximum intensity of the femtosecond main pulse we used was I_{fp} = 2×10^{15} W cm^{-2}, above which the conditions for efficient HHG was destroyed.

The harmonics were spectrally dispersed by a homemade spectrometer with a flat-field grating (1200 lines/mm, Hitachi). The XUV spectrum was then detected by a micro-channel plate and finally recorded using a charge-coupled device (CCD). We also performed time-resolved plasma spectroscopy of ultraviolet (UV) emission from the laser plume, to study the best conditions for HHG. In this case, the UV spectra from the plasma plume were measured using a spectrometer (SpectraPro500i, Acton Research Corp.) and recorded by a time-resolved CCD camera (DH501-18F-01, Andor Technology).

We initially studied various targets to identify promising materials that demonstrate the enhancement of specific harmonics in the plateau region. Among them, In, Sb, Mn, Sn, and Cr, showed the highest enhancement of harmonics. These studies were performed by varying the chirp of the main pump laser pulse, to tune the harmonic wavelengths to the wavelength of the ionic transitions with strong oscillator strengths. We varied the chirp of the main laser pulse, by adjusting the separation between the two gratings of the pulse compressor. Reducing the grating separation from the chirp-less condition generates positively chirped pulses, and an increase provides negatively chirped pulses. Varying the laser chirp resulted in a notable change in the harmonic spectrum from the laser plasma. We also studied the harmonic yield for the same target plumes using 400 nm pump laser, which is the second harmonic of the Ti:sapphire laser generated in a KDP crystal.

3. Results

In the present work, the plumes are produced by loosely focusing the prepulse laser, with an intensity not exceeding 3×10^{10} W cm^{-2}. This produced low ionized plasma, which was necessary for efficient HHG. Under such plasma conditions, we could get maximum conversion efficiency and highest cutoff energy for the high-order harmonics.

The harmonic spectra from the Mn, Sb, Sn, Cr and In plumes showed a plateau-like pattern, with the several harmonic orders having nearly equal intensity. Various characteristics of the HHG were systematically studied to maximize the yield and harmonic cutoff from these plasmas. The influence of the time delay between the prepulse and the main pulse on the harmonic yield was also studied. The harmonic yield increased when we increased the delay from 10 ns to 40 ns, after which it remained roughly constant up to the maximum delay used in this work (140 ns). The focus position of the main pump laser on the plasma was adjusted to optimize the high harmonic yield. We noted a saturation of the high-order harmonics when the main pump laser intensity was high. The best incidence position of the main pump for harmonic generation was at the distance of 100 to 150 μm from the target surface, depending on the harmonic order.

The main pump laser was a chirp-free 35 fs duration pulse. All the targets used in these experiments showed intensity enhancement of a specific harmonic order under these conditions. One method of varying the harmonic spectrum distribution in the plateau is by tuning the central wavelength of the main pump laser [13,15]. However, this is not practical since adjusting the oscillator spectrum cannot be directly transferred to the final laser spectrum due to gain narrowing and gain saturation. We also need to readjust the stretcher and the compressor, making the whole alignment difficult and cumbersome. A much simpler approach to tune the harmonic wavelength without changing the driving laser spectrum is by controlling the chirp of the fundamental radiation [16,17].

The harmonic peaks shift to longer wavelengths for pump lasers with positive chirp, when the leading edge of the pulse consists of the red spectral component. This wavelength shift of the harmonics can be explained by the spectral component in the leading edge of the chirped pump laser. As the intensity of the pump laser increases at the leading edge, HHG efficiency also increases. However, ionization also occurs as the laser intensity increases, which eventually inhibits HHG. Thus there is an ideal pump laser intensity at which the ionization level is still low enough, but the intensity is still high enough to generate harmonics. This ideal intensity is reached at a specific time within the pulse, and so for chirped pulses, there is a specific spectral component associated with this ideal intensity. Therefore, for chirped pulses, the harmonics are odd orders of this spectral component at the leading edge of the pulse. The harmonics produced with positively chirped laser pulses were red shifted because the harmonics produced in the leading edge of the laser pulse come from the red component of the laser spectrum. The same can be said about the blue shifted harmonics produced by negatively chirped pulses. Below we present our studies of some peculiarities of HHG from several materials.

3.1. Manganese plasma

Initially, we observed harmonic generation from manganese plasma up to the cutoff order H of 29. This well coincides with the empirical rule $H \approx 4I_i$ [eV] – 32.1 [11], considering the second ionization potential of Mn (I_{2i} = 15.64 eV). The harmonic spectrum showed a conventional plateau pattern. The intensity of the sub-nanosecond prepulse that produces the plasma plume was $I_{pp} \approx 1\times10^{10}$ W cm^{-2}. However, by further increasing the sub-nanosecond prepulse intensity on the manganese target surface, we were able to observe a notable increase in the harmonic cutoff. Harmonics as high as the 101st order were clearly identified in this case, though the conversion efficiency for most harmonic orders were smaller compared with those for lower prepulse intensities. An interesting observation was the emergence of a plateau pattern at higher orders (from the 33rd to 93rd harmonic), which was followed by a steep drop of harmonic intensity up to the 101st order (7.9 nm). This second plateau appeared in place of a harmonic plateau between 15th to 29th orders, at moderate intensity of Mn target by the sub-nanosecond prepulse. The newly observed cutoff well coincided with the empirical $H(I_i)$ dependence, considering contribution from doubly charged ions and the third ionization potential of manganese (33.67 eV). We should note that the highest harmonic that we could observe was restricted by the spectral resolution of our spectrometer, as well as the continuum emission from the plasma in the range of 5 to 10 nm.

In HHG experiments with 800 nm pump laser, we did not observe any notable enhancement of a single harmonic, although we noticed slight increase of several harmonics between the 33rd to 41st orders. A different pattern was observed for 400 nm driving pulses. The maximum harmonic order (21st) in this case was lower than the case for 800 nm pump (101st), which well coincided with the $H \sim \lambda^2$ rule [18]. However, enhancement of a single harmonic was observed for this experimental configuration (Mn plasma pumped by 400 nm main pulse) (Fig. 1). The intensity of the 17th harmonic was more than 3 to 5 times more intense that those of neighboring harmonics. Interestingly, the wavelength of this harmonic (λ = 23.5 nm) was close to the wavelength of the 33rd harmonic (λ = 24.3 nm) of the 800 nm main pump, which also showed some enhancement, although much less pronounced.

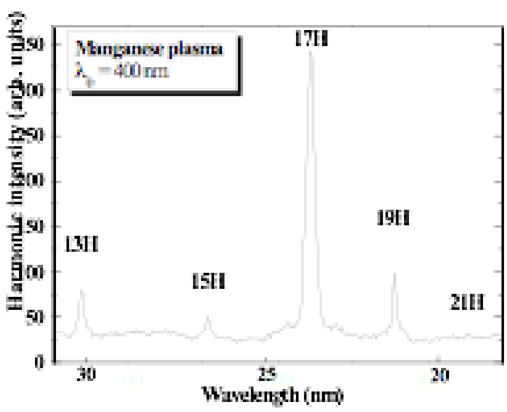

Fig. 1. Harmonic spectra from manganese plasma for 400 nm pump laser.

We tried to tune the harmonic wavelength in the case of 400 nm main pump, by varying the chirp of the 800 nm laser. However, the intensity of the 17th harmonic remained strong, and we were not able to detune the resonance. This behavior can be explained by the narrow bandwidth of the 400 nm pulses (~8 nm), which only allowed the tuning of the 17th harmonic within a narrow spectral range (0.25 nm). The result implies that this spectral tuning is inadequate to detune from the resonance line responsible for enhancing the 17th harmonic. Note that, for the 800 nm laser, varying the laser chirp allowed a notable change in enhancing specifics harmonic in previous works [13-16,19].

The enhancement of the 23.5 nm harmonic (E_{ph} = 52.9 eV) for Mn plasma is attributed to the effects of the presence of ionic lines with strong oscillator strengths. The Mn III and Mn II lines in the range of 51 to 52 nm were studied in past works and proved to have strong oscillator strengths [21, 22]. These results point out that the influence of some of these transitions led to the enhancement of 17th harmonic yield.

3.2. Chromium plasma

In previous studies, chromium plasma showed resonance-induced intensity enhancement and suppression properties for some harmonic order. In [23], a large decrease in the intensity of the 27th harmonic of 796 nm laser (λ = 29.5 nm, $E_{ph} \approx$ 42.2 eV) was reported, and attributed to several ionic transitions with strong absorption oscillator strengths. In the Cr harmonic spectrum, the ratio of the intensity of the 27th and that of the neighboring harmonics changed from almost zero to a value close to 1 by using harmonic tuning [16]. Past works have reported suppression of the 27th harmonic generated from Cr plume [23]. However, no spectral variations of the harmonic wavelength were performed in those experiments to confirm the important role of the resonance-induced variation of the 27th harmonic yield.

At the same time, for Cr, a strong 29th harmonic of the 795 nm laser (λ = 27.3 nm) has recently been reported [16]. This pattern was observed for chirp-free pulses. Varying the pump laser chirp led to the decrease of the 29th harmonic yield compared to the neighboring harmonics. The maximum ratio between the intensity of the 29th harmonic and the 31st harmonic was measured to be 23.

Our present studies using 800 nm, 35 fs pulses confirmed previously reported peculiarities of the harmonic spectra generated from chromium plasma and revealed new features of harmonics for 400 nm pump pulses. In particular, an increased yield of the 29th harmonic (λ = 27.6 nm, $E_{ph} \approx 45.1$ eV) was observed, which roughly coincided with the short-wavelength wing of the strong spectral band of the 3p → 3d transitions of Cr II ions. Moreover, the observed enhancement of the 15th harmonic of the 400 nm pump laser can also be attributed to the influence of the same spectral band, although not as pronounced as the 29th harmonic of the 800 nm laser.

Previous studies of photoabsorption and photoionization spectra of Cr plasma in the range of 41 to 42 eV have demonstrated the presence of strong transitions, which could be responsible for such a suppressed pattern of harmonic spectrum [24-26]. In particular, the region of the "giant" 3p→ 3d resonance of Cr II spectra was studied [26] and the strong transitions, which could both enhance or suppress the optical and nonlinear optical response of the plume were revealed. The neutral and ionized Cr spectra, previously believed to be different, were shown here to be similar. The role of the $3d^5(^6S)$ state in deciding the special position of Cr among the 3d elements was emphasized.

These and other studies used the photoabsorption/photoionization spectra to identify the areas of strong absorption. A great effort has been made to explain the spectral structure previously observed in the 3p excitation region of both neutral and ionized Cr, but the attempts have not been successful. In any case, the reported data explained the nonlinear optical response of Cr plasma for harmonic generation using 800 nm main pump laser, which was revealed in previous [16,23] and present studies.

In the first set of experiments with Cr plume, we observed both the suppression of the 27th harmonic and enhancement of the 29th harmonic of the 800 nm pump laser (Fig. 2). Varying the chirp of the main laser pulse led to further enhancement of the 29th harmonic yield. The ideal conditions, at which this harmonic showed maximum conversion efficiency, corresponded to positively chirped 135 fs pulses. In this case the harmonic yield was about two times stronger than the case for chirp-free pump laser. The enhancement factor of the 29th harmonic compared to the neighboring harmonics (18-times enhancement) was slightly less than that recently reported (23-times enhancement [16]). This is because of the broader spectrum of the pump laser for the former case (40 nm, compared with 20 nm in [16]).

Our experiments with the 400 nm pump laser showed similar enhancement of a single harmonic, whose wavelength was close to previously observed harmonics that showed intensity enhancement using the 800 nm pump. The enhanced 15th harmonic (5-times enhancement, λ = 26.7 nm, $E_{ph} \approx$ 46.7 eV) almost coincides with a broad spectral emission of singly charged Cr, which was also responsible for enhancing the 29th harmonic of the 800 nm pump laser in previous experiments. We tried to vary the chirp of the 400 nm laser by varying that of the 800 nm pump. However, as in the case with Mn plasma, this did not lead to the tuning of harmonic wavelength and thus did not show the relative change in the 15th harmonic intensity compared with its neighbors.

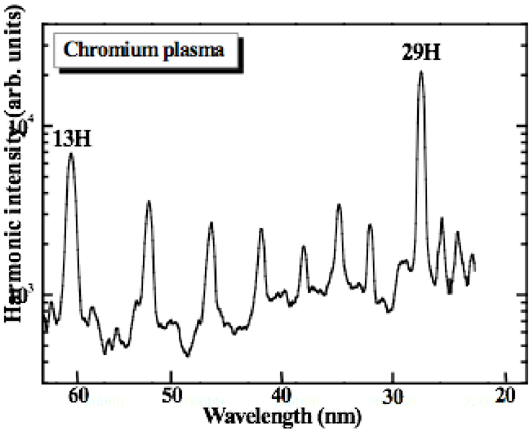

Fig. 2. Harmonic spectra from Cr plasma in the case of 800 nm, 35 fs chirp-free main pulses.

The calculations of the oscillator strength *gf* [24] clearly show a group of the transitions in the 44.5 - 44.8 eV region with strong oscillator strengths (with *gf* varying from 1 to 2.2). These oscillator strengths were larger than those of other transitions in the range. These transitions were assumed to be responsible for the observed enhancement of the 29th harmonic. At the same time, the strong photoabsorption lines within the 41-42 eV region reported in the above work could decrease the yield of 27th harmonic.

3.3. Antimony plasma

HHG of antimony ions has previously been studied using InSb plume [19], as well as with pure Sb plume generated on the surface of solid antimony target [27]. Past works have shown that for InSb plasma, with positively chirped laser pulses, the intensity of the 21st harmonic of 795 nm laser exceeded that of the neighboring harmonics [19]. For a positively chirped laser pulse of 140 fs duration, this enhancement factor was reported to be 10-times. On the other hand, for chirp-free and negative chirped laser pulses, the 21st harmonic intensity was only slightly higher than that of the neighboring ones. The role of Sb in 21st harmonic enhancement was confirmed by the studies of pure In plume, where no enhancement was observed for this harmonic. A confirmation of this conclusion was also obtained in [27]. The intensity enhancement of a single high-order harmonic at a wavelength of 37.67 nm was demonstrated using low ionized antimony laser-ablation plume. The conversion efficiency of this harmonic was reported to be 2.5×10^{-5} and the output energy was 0.3 μJ. Such an enhancement of single-harmonic was caused by the multiphoton resonance with the strong radiative transition of the Sb II ions. The intensity of the 21st harmonic at the wavelength of 37.67 nm was 20 times higher than that of the 23rd and the 19th harmonics.

The Sb I spectrum is dominated by two peaks, a broad peak centered near 31.24 eV with a bandwidth close to 1 eV (FWHM) and a narrower peak centered near 32.22 eV. The spectrum also contains a large contribution from Sb II, which results in peaks at 32.4 and 32.7 eV. However, the strongest transitions among these Sb I lines [28] was the $4d^{10}5s^25p^3\ ^2D_{5/2}$ – $4d^95s^25p^4\ (^3P)^2F_{7/2}$ transition ($E_{ph} \approx$ 31.5 eV). The *gf* value of this transition is calculated to be 1.54, which is few times higher than those of other transitions in this spectral range. At the same time, among the calculated *gf*-values for the $4d^{10}5s^25p^2$ - $4d^95s^25p^3$ transitions of Sb II ion, the $^3P_2 – (^2D)^3D_3$ transition ($E_{ph} \approx 32.8$, $gf = 1.36$) also shows a strong oscillator strength, which could influence the nonlinear optical response of plasma due to proximity of the harmonic wavelength with this transition.

In the present experiments, enhancement of the 21st harmonic generated from the antimony plume was observed using main pump lasers with different pulse durations and chirp. The maximum conversion efficiency is achieved with a negative chirp with pulse duration of 210 fs. For antimony, no enhanced harmonics were observed for the 400 nm pump. The difference in the results of the present work with those of Ref 19 is attributed to the different central wavelength and the spectral bandwidth of the pump laser. The enhancement factor for the 21st harmonics in these studies (8-times) was less than those of past works (10-times [19] and 20-times [27]). This could be explained by the different methods that were used for tuning the harmonic wavelength to the ionic/neutral transitions of Sb. In [27], wavelength tuning of the pump laser was realized by shifting the master oscillator wavelength close to the resonance lines of ions, while in [19] the chirp was varied, but using pump laser with a narrower bandwidth. These results imply that the use of pump laser with narrower bandwidth is preferable to increase the enhancement of the harmonics.

3.4. Tin plasma

We observed in the present study strong enhancement of the 17th harmonic of the 800 nm pump (47.1 nm, 26.5 eV) for the Sn plume (Fig. 3). We studied this phenomenon by shifting the harmonic wavelength in the range of ±0.5 nm relative to the chirp-free position of the harmonic wavelength. We find that there is variation of this harmonic yield, which was maximum for a negatively chirped 70 fs pump laser. The origin of this phenomenon is similar to previously presented data on resonance-induced enhancement of single harmonics from specific plumes. For tin plasma, the 15-times increase in the harmonic yield at specific chirp of the pump laser is attributed to the proximity of the 17th harmonic wavelength to ionic transitions with strong oscillator strength.

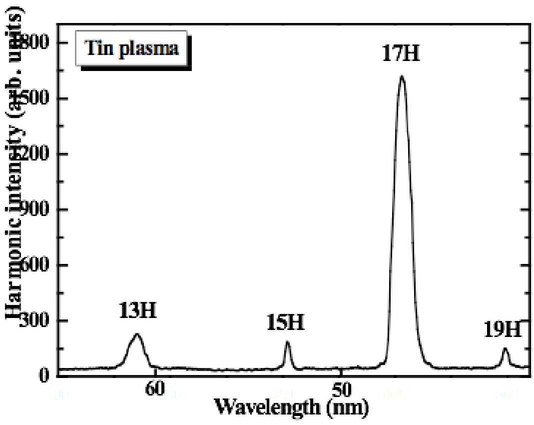

Fig. 3. Harmonic distribution in the case of Sn plume. λ = 800 nm.

Recently, such an enhancement was reported and optimized by tuning the central wavelength of the master oscillator of the laser [30]. In this work, the observation of strong single high-order harmonic generation at the wavelength of 46.76 nm by using tin laser-ablation plume was reported. The intensity of the 17th harmonic at the wavelength of 46.76 nm was 20 times higher than its neighboring harmonics. The energy of the 17th harmonic was measured to be 1.1 μJ. The origin of this enhancement was attributed to resonance with strong radiative transition of the Sn II ion, produced within the laser-ablated plume.

In past work, the Sn II ion has been shown to posses a strong transition $4d^{10}5s^25p\,^2P_{3/2}$ – $4d^95s^25p^2$ (^1D) $^2D_{5/2}$ at the wavelength of 47.20 nm ($E_{ph} = 26.24$ eV) [31]. The *gf* value of this transition is 1.52, and this value is five times larger than other transitions from the ground state of Sn II. Therefore the enhancement of the 17th harmonic with the 800 nm wavelength laser pulse can be explained as being due to resonance with this transition.

3.5. Indium plasma

Harmonic generation from indium plasma has recently been studied [19]. Here we present some additional studies on the peculiarities of single harmonic enhancement from this nonlinear medium. As in previous publications [13,19], strong 13^{th} harmonic (61.5 nm, 20.2 eV) appeared from In plasma upon irradiation of the 800 nm, 35 fs chirp-free laser pulse. At the same time, a notable suppression of the 15^{th} harmonic was observed under these conditions. Harmonics up to the 35^{th} cutoff were observed and showed a plateau-like harmonic spectrum. As in previous cases, we studied the effects of the main pulse chirp on the harmonic intensity. As a result, we observed a large increase in the intensity of the 13^{th} harmonic using negatively chirped pulses, compared with those generated by chirp-free pump. The enhancement factor of the 13^{th} harmonic compared to neighboring ones was 180, which is close to previously reported data (80-times [13] and 200-times [19]).

The indium plasma emission observed in the spectral range of interest is due to radiative transitions between the ground state ($4d^{10}\,5s^2\,{}^1S_0$) and the lowly lying state ($4d^{10}\,5s\,5p$) of In II. Among them, the transition at 19.92 eV (62.1 nm), corresponding to the $4d^{10}\,5s^2\,{}^1S_0 \rightarrow 4d^9\,5s^2\,5p\,({}^2D)\,{}^1P_1$ transition of In II, is exceptionally strong. The absorption oscillator strength *gf* of this transition is 1.11 [32], which is more than twelve times larger than other transitions from the ground state of In II. We attribute the intensity enhancement of the 13^{th} harmonic to resonance effects with this transition.

When we changed the wavelength of the pump laser from 800 to 400 nm, this strong enhancement disappeared. For the 400 nm pump, harmonics from indium plasma did not have any increased harmonic yield, since the wavelength of the 7^{th} harmonic of the 400 nm pump laser was too far from In transitions with strong oscillator strengths.

4. Discussion

In most of the resonance-related HHG work, the harmonic spectrum was studied as a function of the laser intensity to show the existence of enhancements for particular intensities [3-6,33]. This technique is distinct from the early approach where changing the pump laser spectrum tuned the harmonic wavelength and adjusted it to coincide with an atomic or ionic resonance in the nonlinear medium [2,34,35]. Our approach is close to the latter one, though we did not change the spectrum of the fundamental radiation but instead changed the spectral distribution inside the pulse by controlling the chirp of the laser radiation. As shown above, this leads to the tuning of the harmonic wavelength and thus allows one to achieve the resonance enhancement of the harmonic yield for some plumes.

There is a primary difference between our observation of enhanced single harmonics in the present work, using Sb-, Sn-, Cr-, Mn-, and In-containing plumes, with the previous work on resonance enhancement of harmonic generation. The latter includes those in alkali metals [4], where multi-harmonic enhancement was reported, and in rare gases [6,33], when the single harmonic intensity was enhanced a few times compared with those of neighboring harmonics. Chirp control of the pump laser allows one to achieve an ideal relation between the quasi-resonance conditions, re-absorption, and induced self-defocusing, leading to a large enhancement of the single harmonic yield.

The few demonstrations of resonance enhancement of the high-order harmonics in previous laser-gas HHG experiments can be understood by considering the difference between the excited spectra of atoms and ions of available solid targets and few rare gases. For plasma from various targets, there is a higher chance to find a proper target with multi-resonance conditions in XUV range, which can lead to the enhancement of the harmonic yield.

Resonance enhancement introduces a new possibility of increasing the conversion efficiency of a specific harmonic order by more than two-orders of magnitude. If this effect could be combined with phase matching or coherent control of HHG, one would be able to generate a spectrally pure coherent x-ray source with only a single-line in the spectrum, much like saturated x-ray lasers produced by ionic population inversions in highly ionized plasmas. The resulting source will, however, have superior spatial coherence, possibility of high (up to kHz) repetition-rate, and improved conversion efficiency. Such a unique radiation source will be ideal for accelerating its various applications in physics, chemistry and biology, and to explore new fields such as nonlinear x-ray optics and attosecond physics.

5. Conclusions

We presented the results of detailed studies on resonance-induced enhancement of single high-order harmonics generated in laser plasma, in different spectral ranges using femtosecond pump lasers with 800 and 400 nm central wavelength. For these purposes, the Mn, Cr, Sb, Sn, and In plasmas were identified as the suitable nonlinear media for efficient harmonic generation and single harmonic enhancement. Most of the ionic/neutral transitions responsible for the observed resonance-induced enhancement are identified, which all showed strong oscillator strengths. The enhancements of the 13^{th}, 17^{th}, 21^{st}, 29^{th}, and 33^{rd} harmonics from these targets were obtained and analyzed using the 800 nm femtosecond laser with various chirp. We also presented the observation of harmonic enhancement from some targets with a second harmonic pump laser (400 nm central wavelength). Using Mn plume, we demonstrated the highest harmonic photon energy (52.9 eV) at which enhancement has been observed (17^{th} order, $\lambda = 23.5$ nm).

Acknowledgements

This work was partially supported by the Research Foundation for Opto-science and Technology. The authors wish to thank J.-P. Moreau for the technical support. R. A. Ganeev gratefully acknowledges the support from the Fonds Québécois de la Recherche sur la Nature at les Technologies to carry out this work.

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