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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 plasma 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. Quasi-monochromatic harmonics are observed 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. Intensity enhancements of the 13^{th}, 17^{th}, 21^{st}, 29^{th}, and 33^{rd} harmonics from these targets are demonstrated 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 (17th order, $\lambda = 23.5$ nm).

Keywords: Ablation; High harmonic generation

INTRODUCTION

High-order harmonics is a distinct source of coherent soft X-rays (Ozaki *et al.*, 2006, 2007), with unique capabilities to produce ultrashort pulses on the order of 100 attoseconds. However, conversion efficiency of high-order harmonics is a key issue when considering real applications, such as diagnostics of dense plasma where penetrating radiation is required (Cao *et al.*, 2007; Nardi, 2007; Orlov *et al.*, 2007). One successful approach to overcome this challenge has been to phase match the pump and the harmonics using gas-filled waveguides (Kazmias *et al.*, 2003). An alternative approach is the possibility to enhance harmonic generation using atomic resonances (Reintjes, 1984). 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 (Figueira De Morisson Faria *et al.*, 2002; Gaarde & Schafer, 2001; Taïeb *et al.*, 2003; Toma *et al.*, 1999; Zeng *et al.*, 2002; Bartels *et al.*, 2000) 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.* (2000) 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 (Pfeifer *et al.*, 2005). However, in these studies, the intensity of neighboring harmonics was comparable to the enhanced harmonics.

As a method for generating intense harmonics, we have been exploring laser ablation media as the nonlinear medium. Laser ablation has been used in numerous applications, including laser induced plasma spectroscopy (Fang & Ahmad, 2007; Schade *et al.*, 2006), laser deposition (Veiko *et al.*, 2006; Wolowski *et al.*, 2007) and the production of nanoparticles (Wang *et al.*, 2007). Recent investigations of HHG from plasma plume imply an alternative approach for intense harmonic generation. The method capitalizes on the efficient harmonic generation from low-ionized ions produced on the surfaces of various solid-state targets (Ganeev, 2005*a*, 2005*b*, 2006*a*). 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 (Ganeev *et al.*, 2006*b*, 2006*c* 2007; Suzuki *et al.*,

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2006). In particular, 80-times intensity enhancement of the 13th harmonic ($\lambda = 61.2$ nm) of Ti:sapphire laser was demonstrated, using indium plasma as the nonlinear medium, and by varying the spectrum of the pump laser (Ganeev, 2006*b*). 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 (29th 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 near 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, $\lambda = 23.5$ nm), which is also the highest photon energy ($E_{ph} = 52.9$ eV) at which intensity enhancement has been demonstrated. We have also been able to identify an extension of the harmonic cut-off under proper conditions, using Mn as the plasma plume. High-order harmonics up to the 101^{st} order has been observed, which is the highest cut-off ever observed for harmonics from plasma plume. Simulations show that the reason for this unique behavior of Mn plasma lies in the relatively low electron density of the plume when the femtosecond pump laser arrives.

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 laser system used in this work is one of the beam lines of the Advanced Laser Light Source, a Canadian international user facility on ultrafast laser science (Ozaki, 2006). We adjusted the focal spot diameter of the prepulse beam on the target surface 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}. We chose this prepulse intensity range based on previous studies of various ablated targets. After a delay between 50 and 80 ns, we focused the femtosecond main pulse (8 to 25 mJ energy, 35 fs pulse duration, 800 nm central wavelength, 40 nm bandwidth full width half maximum (FWHM)) on the plasma from an orthogonal direction, by using an MgF_2 plano-convex lens ($f = 680$ mm). The maximum intensity of the femtosecond main pulse we used was $I_{fp} = 2 \times 10^{15}$ W cm^{-2}, above which we could not get efficient HHG.

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, we observed the UV spectra from the plasma plume using a spectrometer (SpectraPro500i, Acton Research Corp., Action, MA) and recorded by a time-resolved CCD camera (DH501-18F-01, Andor Technology, Belfast, UK).

We initially studied various targets to identify promising materials that show 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 was generated by frequency doubling the Ti:sapphire laser with a KDP crystal.

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 obtain the maximum conversion efficiency and the highest cut-off energy for the high-order harmonics.

The harmonic spectra from Mn, Sb, Sn, Cr, and In plumes showed a plateau-like pattern, with several harmonic orders having nearly equal intensity. Various characteristics of HHG from these plasmas were systematically studied to maximize the yield and harmonic cut-off. 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

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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 (Ganeev *et al.*, 2006*b*, Suzuki *et al.*, 2006). However, this is not practical since adjusting the oscillator spectrum cannot be directly transferred to the final laser spectrum because of 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 (Ganeev *et al.*, 2007; Kim *et al.*, 2004).

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.

Resonance Enhancement in Tin Harmonics

We first describe resonance enhancement of a single harmonic using the fundamental wavelength of the Ti:sapphire laser. 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. 1). We studied this phenomenon by shifting the harmonic wavelength in the range of $\pm$ 0.5 nm relative to the chirp-free position of the harmonic wavelength. We find that there is variation of this harmonic

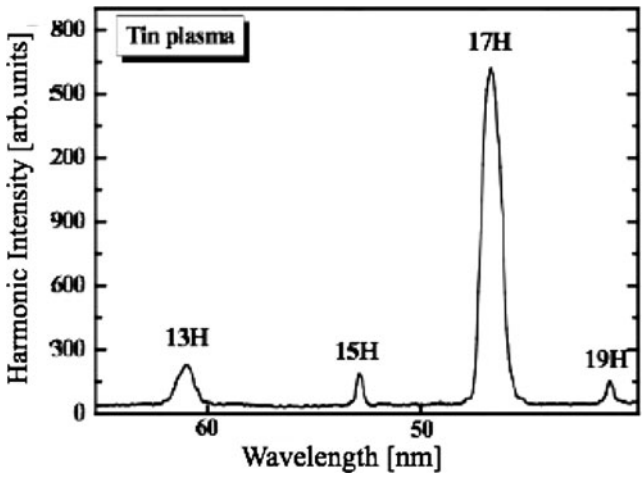

Fig. 1. Harmonic distribution for Sn plume. $\lambda = 800$ nm.

yield, which had maximum yield 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. 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.

Recently, such an enhancement was reported and optimized by tuning the central wavelength of the master oscillator of the laser (Suzuki *et al.*, 2006). This work reported observing strong single high-order harmonic generation at the wavelength of 46.76 nm by using tin laser-ablation plume. 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 $4d^{10}5s^25p^2P_{3/2} - 4d^95s^25p^2$ (^{1}D) $^2D_{5/2}$ at the wavelength of 47.20 nm ($E_{ph} = 26.24$ eV) (Duffy *et al.*, 2001). 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.

Resonance Enhancement in Manganese Harmonics

In HHG experiments using Mn plasma and 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 pump laser. The maximum harmonic order (21st) in this case was lower than that for 800 nm pump, which well coincided with the harmonic cutoff order $H{\sim}\lambda^2$ rule (Corkum, 1993). However, enhancement of a single harmonic was observed when we pumped

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the Mn plasma with a 400 nm main pulse (Fig. 2). The intensity of the 17th harmonic was more than three to five times more intense that those of neighboring harmonics. Interestingly, the wavelength of this harmonic ($\lambda =$ 23.5 nm) was close to the wavelength of the 33rd harmonic ($\lambda = 24.3$ nm) of the 800 nm main pump, which also showed some enhancement, although much less pronounced.

We tried to tune the harmonic wavelength for 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 tuning 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 (Ganeev *et al.*, 2006*b*, 2006*c*, 2006*d*, 2007; Suzuki *et al.*, 2006).

Enhanced 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 (Dolmatov, 1996; Kilbane *et al.*, 2005). These results point out that the influence of some of these transitions led to enhanced 17th harmonic yield.

Cut-off Extension in Manganese Harmonics

Although single harmonic enhancement could not be demonstrated for 800 nm pump lasers, this experimental condition showed a different but interesting behavior. As in previous cases, we observed harmonic generation from this material up to the maximum cut-off $H_{cut\text{-}off} = 29$. This is in good agreement with the empirical rule of $H_{cut\text{-}off} \approx 4I_i$ [eV] – 32.1, considering the second ionization potential of Mn

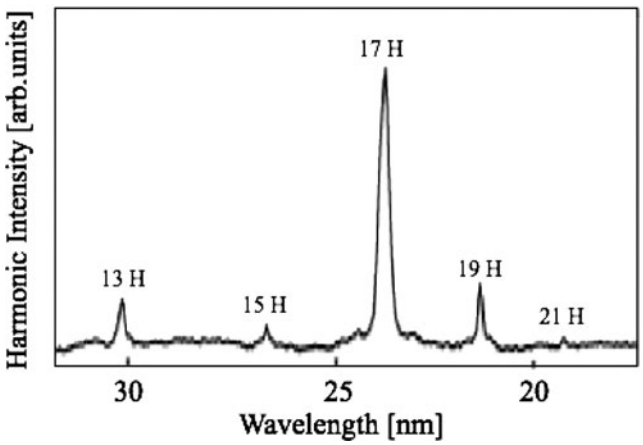

Fig. 2. Harmonic spectra from manganese plasma for 400 nm pump laser.

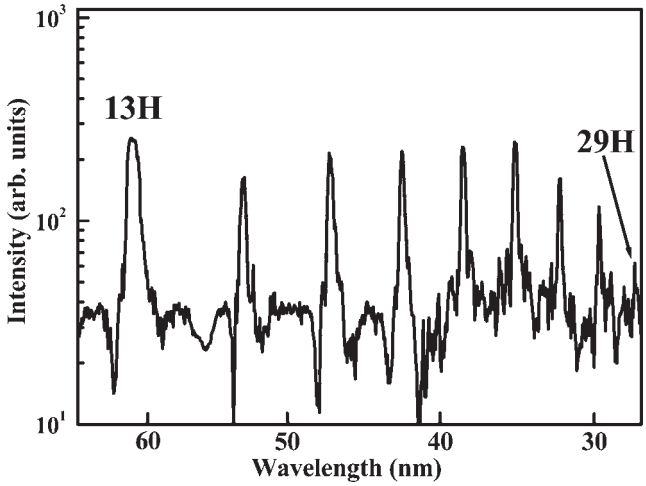

Fig. 3. Harmonic spectrum from manganese ablation obtained at $I_{fp} = 5 \times 10^{14}$ W cm^{-2} and $I_{pp} = 1 \times 10^{10}$ W cm^{-2}.

($I_{2i} = 15.64$ eV). The harmonic spectrum showed a conventional plateau pattern for high orders of harmonics (Fig. 3). The intensity of the sub-nanosecond prepulse that produces the plasma plume in this case was $I_{pp} \approx 1 \times 10^{10}$ W cm^{-2}. However, by further increasing the sub-nanosecond prepulse intensity on the Mn target surface, we were able to observe a notable increase in the harmonic cut-off. 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 the case of smaller prepulse intensities (Fig. 4). An interesting observation was the emergence of a plateau pattern at higher orders (from the 33rd to 93rd harmonic) with further steep drop of harmonic intensity up to 101st order (7.9 nm). This second plateau appeared in place of a harmonic plateau between 15th to 29th orders that were observed for moderate irradiation of Mn target by

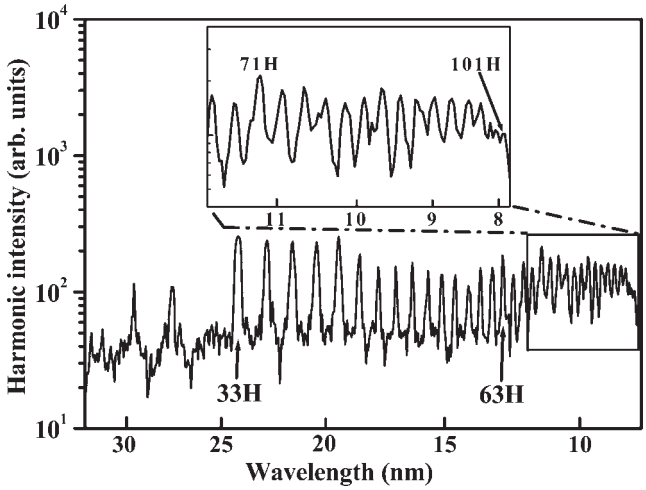

Fig. 4. A lineout of high-order harmonic spectrum obtained at $I_{fp} = 2 \times 10^{15}$ W cm^{-2} and $I_{pp} = 3 \times 10^{10}$ W cm^{-2}. Inset: A region of resolved harmonic distribution from 67th to 101st orders.

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the sub-picosecond prepulse. The newly observed cutoff well coincided with the empirical $H_{\text{cut-off}}$ rule, if one considers the involvement of doubly charged ions and the third ionization potential of manganese (33.67 eV).

We performed systematic investigations of this phenomenon, by measuring the cut-off as a function of the main pump intensity. However, we did not observe any saturation in the harmonic cutoff with an increase in the main pulse intensity (Fig. 5). This infers the possibility of obtaining even higher harmonics from manganese plasma plume, by using higher main pulse intensities. 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.

To understand this different behavior of the HHG process for different plasma materials, we performed hydrodynamic simulations using the HYADES code (Ganeev *et al.*, 2006*b*). We simulated the expansion of the manganese and gold targets irradiated by the prepulse laser. We determined the electron density, ionization level, and ion density of these plumes as a function of the prepulse intensity, at 300 μm from the target surface. Results of these calculations for 100 ns delay show that, already at 1.04×10^{10} W cm^{-2}, there is a notable difference in ionization states for Mn and Au plasma, which can lead to the difference in the nonlinear optical response. At this intensity, the ionization level of Au plume becomes higher than 1, which leads to increased free electron density, due to the ionization of singly charged ions. The ratio between the electron density and ion density in gold plasma continues to increase at higher prepulse intensities, and at $I_{\text{pp}} = 5 \times 10^{10}$ W cm^{-2}, their ratio becomes higher than 4. The increase in the free electron density for Au prevents efficient harmonic generation and the extension of harmonic cut-off, due to self-defocusing of the main pump laser and the growing phase mismatch between the harmonic and the pump laser.

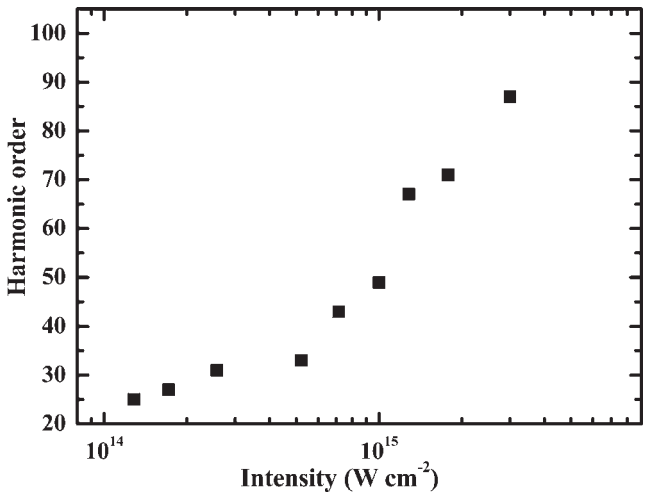

Fig. 5. Harmonic cut-off from Mn plasma as a function of main pulse intensity.

The characteristics of manganese plasma under the same conditions are different from those of Au. The ionization level of the Mn plume is considerably lower than that of Au under the same prepulse intensity, and doubly charged ions can only be expected at $I_{\text{pp}} = 5 \times 10^{10}$ W cm^{-2}. Therefore, the effect of free electrons on the main pump laser is smaller. What is especially important for HHG is that the ion density increases considerably with an increase in the prepulse intensity. As a result, the harmonics, especially near the cut-off, will increase nonlinearly in intensity, thus helping the detection of these harmonics. These combined features of relatively low electron density and high ion density in manganese plasma allowed the demonstration of the highest harmonics ever observed from plasma plume.

The results show that new findings in extending the harmonic cutoff toward the soft X-ray region may be possible, by searching for optimal HHG conditions of the plasma plume through plasma simulation and time-resolved UV spectroscopy. On the other hand, 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.

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. Tin demonstrates single harmonic enhancement of the 17th harmonic for 800 nm pump, while manganese shows similar enhancement of the 17th harmonic of the 400 nm pump. Further, we find that the cutoff is considerably extended.

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