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# Systematic investigation of resonance-induced single-harmonic enhancement in the extreme-ultraviolet range

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We demonstrate the intensity enhancement of single harmonics in high-order harmonic generation from laser plasma. We identified several targets (In, Sn, Sb, Cr, and Mn) that demonstrate resonance-induced enhancement of single harmonic, that are spectrally close to ionic transitions with strong oscillator strengths. We optimized and obtained enhancements of the 13th, 17th, 21st, 29th, and 33rd harmonics from the above targets, by varying the chirp of the 800 nm wavelength femtosecond laser. We also observe harmonic enhancement by using frequency-doubled pump laser (400 nm wavelength). For Mn plasma pumped by the 400 nm wavelength laser, the maximum order of the enhanced harmonic observed was the 17th order ($\lambda = 23.5$ nm), which corresponds to the highest photon energy (52.9 eV) reported for an enhanced single harmonic.

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## I. INTRODUCTION

Conversion efficiency of high-order harmonics is a key issue, when one starts to consider real applications of this unique source. One successful approach to overcome this challenge has been to phase match the pump and the harmonics using gas-filled waveguides [1]. Recently, there have also been considerable advances in using two-color excitation to enhance the intensity of multiple harmonics [2,3]. An alternative approach is the possibility to enhance harmonic generation using atomic resonances. The role of atomic resonances in harmonic generation was an important subject of discussion in early studies of low-order harmonic generation ([4], and references therein).

The challenge to achieve intensity enhancement of high-order harmonic generation (HHG) in gaseous media using atomic and ionic resonances has been investigated, and both theoretical and experimental reports [5–8] have shown the perspectives of this approach. Intensity enhancement of some harmonic orders has been reported in laser-gas jet interaction. For instance, roles of resonances and recollision in atoms have been discussed in Ref. [5] in terms of strong-field atomic phenomena. However, they have predicted harmonic intensity enhancement over a broad range of harmonics. Further, using an optimized laser pulse shape, Bartels *et al.* [8] were able to enhance the 27th 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 the above studies, the growth of intensity of neighboring harmonics occurred simultaneously. One should also note that there are also a limited number of available gases for HHG.

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

Currently, intensity enhancement of single harmonics has been limited to relatively 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 the creation of an intense, quasimonochromatic source of coherent extreme ultraviolet (XUV) radiation. 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 investigated targets such as In, Sn, Sb, Cr, and Mn, and were able to demonstrate intensity enhancement of the 13th, 17th, 21st, 29th, and 33rd harmonics of the 800 nm pump laser, respectively. Such enhancement always occurred when the wavelength of the harmonic was spectrally in the vicinity of a strong radiative transition with large oscillator strength. 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 enhance-

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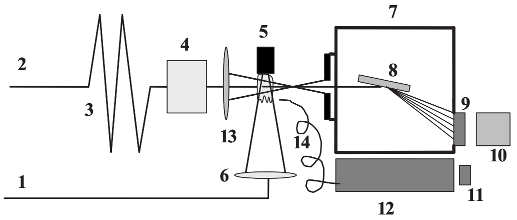

FIG. 1. Schematic of the HHG from laser plasma. 1: Subnanosecond radiation; 2: Main radiation; 3: Delay line; 4: Compressor; 5: Target; 6,13: Focusing lenses; 7: XUV spectrometer; 8: Grating; 9: Microchannel plate; 10,11: CCD; 12: UV spectrometer; 14: Fiber waveguide.

ment is observed is for the Mn plasma (17th order, $\lambda = 23.5$ nm), which also corresponds to the highest photon energy ($E_{\mathrm{ph}} = 52.9$ eV) at which intensity enhancement for single harmonics has been demonstrated.

## II. EXPERIMENTAL SETUP

A schematic diagram of the experimental setup is shown in Fig. 1. 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 approximately 600 $\mu$m. The intensity of this subnanosecond prepulse $I_{\mathrm{pp}}$ on the target surface was varied between $7 \times 10^{9}$ W cm$^{-2}$ and $4 \times 10^{10}$ 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 ($E = 8-25$ mJ, $t = 35$ fs, $\lambda = 800$ 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 $I_{\mathrm{fp}} = 2 \times 10^{15}$ W cm$^{-2}$, above which the conditions for the efficient HHG deteriorated.

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 microchannel 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 investigate the optimal 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 investigated 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 chirpless condition generates positively chirped pulses, and an increase provides negatively chirped pulses. Varying the laser chirp resulted in a considerable change in the harmonic spectrum generated from the laser plasma.

We also investigated the harmonic yield for the above target plumes using the 400 nm pump laser, which is the second harmonic of the Ti:sapphire laser generated in a KDP crystal. There are two ways of representing the harmonic output: in the first method, one takes the maximum value of the peak (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, since we observed a difference in the bandwidth of the harmonics under different conditions.

## III. RESULTS

In the present work, we used plumes that were generated under the conditions of loose laser focusing, with the intensity of the prepulse not exceeding $3 \times 10^{10}$ W cm$^{-2}$. This produced low ionized plasma, which was necessary for efficient HHG. Under such plasma conditions, we could obtain 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 plateaulike pattern, with the several harmonic orders having nearly equal intensity. Various characteristics of the HHG were systematically studied in order 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 investigated. The harmonic output increased considerably when the delay was increased from 10 ns to 40 ns, after which it remained approximately constant up to the maximum delay used in this work (140 ns). The focus position of the main pump laser relative to the plasma was adjusted to optimize the high harmonic output. We observed a saturation of the high-order harmonics when the main pump laser intensity was high. The optimum incidence position of the main pump for harmonic generation was at the distance of $100-150$ $\mu$m 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 some intensity enhancement (or reduction) of some 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 because the adjustment of the oscillator spectrum cannot be directly transferred to the final laser spectrum due to gain narrowing and gain saturation processes. We also need to readjust the stretcher and the compressor, making the whole alignment process very difficult and cumbersome. A much simpler approach to tune the harmonic wavelength without modifying the driving laser spectrum is by controlling the chirp of the fundamental radiation [16,17].

The harmonic peaks shift to longer wavelengths for positive chirp, when the leading edge of the pulse consists of the red spectral component of the pump laser. The 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 is also increased. However, ionization also occurs as the laser intensity is increased, which eventually destroys HHG. Thus there is an optimum pump laser intensity at which the ionization level is still low enough, but the intensity is still high enough to generate harmonics. This optimum intensity is reached at a specific time within the pulse, and so for chirped pulses, there is a specific spectral component associated with this optimum 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 redshifted 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 blueshifted harmonics produced by negatively chirped pulses. Below we present our studies of some peculiarities of HHG from several plumes.

### A. Manganese plasma

Initially, we observed the harmonic generation from manganese plasma up to the maximum cutoff $H=29$, which well coincides with the empirical rule $H \approx 4I_i$ [eV]$-32.1$ [11], taking into account the second ionization potential of Mn ($I_{2i}=15.64$ eV). The harmonic spectrum showed a conventional plateau pattern for higher orders of harmonics. The intensity of the subnanosecond prepulse that produces the plasma plume for this case was $I_{\mathrm{pp}} \approx 1 \times 10^{10}$ W cm$^{-2}$. However, by further increasing the subnanosecond prepulse intensity on the manganese target surface, we were able to observe a considerable 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 the 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 and 29th orders, which were observed for moderate irradiation of the Mn target by the subnanosecond prepulse. The newly observed cutoff well coincided with the empirical $H(I_i)$ dependence, taking into account the involvement of 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 5–10 nm.

In these HHG experiments with 800 nm driving radiation, no considerable enhancement of a specific single harmonic was observed, although a slight increase of several harmonics between the 33rd and 41st orders compared to other harmonics was clearly seen in harmonic spectra. A different pattern was observed in the case of 400 nm driving pulses. The maximum harmonic order (21st) in this case was considerably lower compared to the case of the 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. 2). The intensity of the 17th harmonic was 5× more intense than 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) in the case of the 800 nm main pump, which also showed some enhancement with respect to the neighbor harmonics, although much less pronounced.

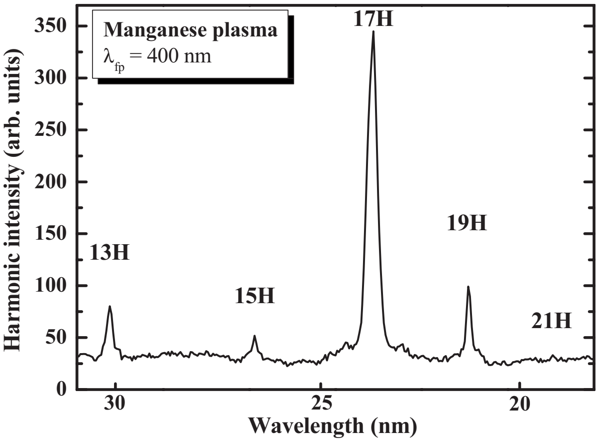

FIG. 2. Harmonic spectra from manganese plasma in the case of 400 nm driving radiation.

We attempted to tune the harmonic wavelength in the case of the 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). This level of spectral tuning seems to be insufficient to detune from the resonance line responsible for the enhancement of the 17th harmonic. Note that, in the case of the 800 nm laser, the variation of the laser chirp allowed a considerable change in the enhancement of specifics harmonic in previous works [14,16,19].

We performed time-resolved spectroscopy of the emission from the Mn plasma in the narrow UV range (253–263 nm). Figure 3 presents the dynamics of the change in such spectra during the first 150 ns, for a prepulse to main pulse delay of 100 ns. In these time-resolved spectra, the main pulse, which arrives 100 ns after the beginning of plasma formation, excites exclusively the ionic lines in the spectrum, while no increase in the intensity of the neutral lines was observed. These results show that, for conditions under which higher-order harmonics were generated, the main laser pulse interacted with the ions, up to the maximum delays used in these experiments. The time gate for each UV spectral measurement was 20 ns. Measurements were taken each 10 ns from the beginning of the irradiation of the Mn target, up to 150 ns. We performed these time-resolved measurements at both the optimal and nonoptimal conditions of HHG. We measured the change in the Mn plasma emission in the vi-

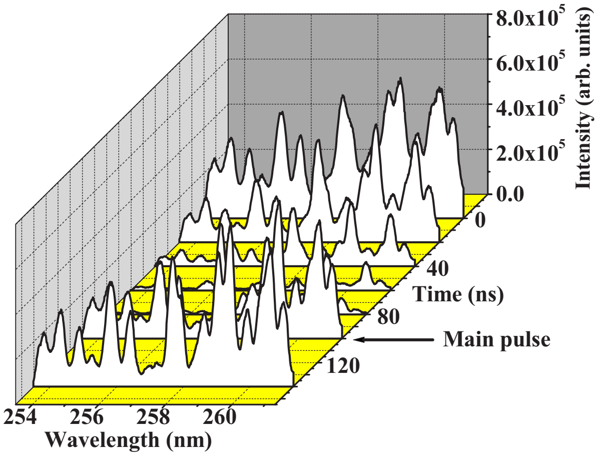

FIG. 3. (Color online). Time-resolved UV spectra of the "optimal" Mn plasma.

cinity of the UV spectral lines related with the excitation of singly charged ions.

The plasma characteristics were analyzed by simulations using the HYADES code [20]. We simulated the expansion of the manganese target interacting with the prepulse laser, and determined the electron density, ionization level, and ion density of this plume as a function of the prepulse intensity, at a distance 300 $\mu$m from the target surface. These simulations showed that, at $I_{pp} \approx (2-3) \times 10^{10}$ W cm$^{-2}$, the ionization level of the Mn plume achieves 1, and the concentration of singly charged ions and electrons becomes equal to 3.3 $\times 10^{17}$ cm$^{-3}$. What is especially important here for HHG is that the ion density increases considerably with an increase in the prepulse intensity. As a result, the harmonics, especially near the cutoff, will increase nonlinearly in intensity, thus helping the detection of these harmonics.

The enhancement of 23.5 nm radiation ($E_{ph}$=52.9 eV) in the case of Mn plasma could be associated with the resonance-induced growth of conversion efficiency in the presence of ionic lines with strong oscillator strengths. The Mn III and Mn II lines in the range of 51–52 nm were studied in past works and proved to possess strong oscillator strengths [21,22]. These results indicate that the influence of some of these transitions led to the growth of the 17th harmonic yield.

### B. Chromium plasma

In previous studies, chromium plasma showed resonance-induced intensity enhancement and suppression properties for some harmonic order. In [23], a considerable decrease of the 27th harmonic of 796 nm radiation ($\lambda$=29.5 nm, $E_{ph} \approx$42.2 eV) with regard to the neighboring ones was reported and attributed to some ionic transitions demonstrating strong absorption oscillator strength. 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]. Although the observation of the extinct 27th harmonic generated from the Cr plume has previously been reported in Ref. [23], no spectral variations of the harmonic wavelength were performed in those experiments to confirm the crucial role of the resonance-induced variation of the 27th harmonic yield.

At the same time, in the case of Cr, a strong 29th harmonic of the 795 nm laser ($\lambda$=27.3 nm) belonging to the midplateau region has recently been reported [16]. This pattern was observed in the case of chirp-free pulses. The variation of the chirp of the pump laser led to the decrease of the 29th harmonic yield compared to the neighboring harmonics. The maximum ratio of the intensity of the 29th harmonic compared to that of 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 generating from chromium plasma and revealed new features of harmonics in the case of 400 nm driving pulses. In particular, an enhanced 29th harmonic ($\lambda$=27.6 nm, $E_{ph} \approx$45.1 eV) approximately coincided with the short-wavelength wing of the strong spectral band of the $3p \rightarrow 3d$ transitions of Cr II ions. Moreover, the observed enhancement of the 15th harmonic of 400 nm driving radiation also can be attributed to the enhancement of nonlinear susceptibility of this harmonic induced by the influence of the same spectral band, though not so pronounced as in a case of the 29th harmonic of 800 nm radiation.

Previous studies of photoabsorption and photoionization spectra of Cr plasma in the range of 41–42 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" $3p \rightarrow 3d$ resonance of Cr II spectra was analyzed in Ref. [26] and the strong transitions, which could both enhance and diminish the optical and nonlinear optical response of the plume, were revealed. The neutral and ionized Cr spectra, previously believed to be completely different, were shown here to be rather similar. The role of the $3d^5(^6S)$ state in determining 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 considerable 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 entirely the nonlinear optical response of Cr plasma in the case of harmonic generation using the 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 extinction of the 27th harmonic and enhancement of the 29th harmonic of the 800 nm pump laser (Fig. 4). Variation of the chirp of the main laser pulse led to further enhancement of the 29th harmonic yield (Fig. 5). The optimal conditions, at which this harmonic showed maximum conversion efficiency, corresponded to positively chirped 135 fs pulses. In this case the harmonic output was approximately two times stronger compared to the case of the chirp-free pump laser. The enhancement factor of the 29th harmonic compared with the neighboring harmonics (18× enhancement) was slightly less than recently reported data (23× enhancement, [16]), probably due to the broader spectrum of

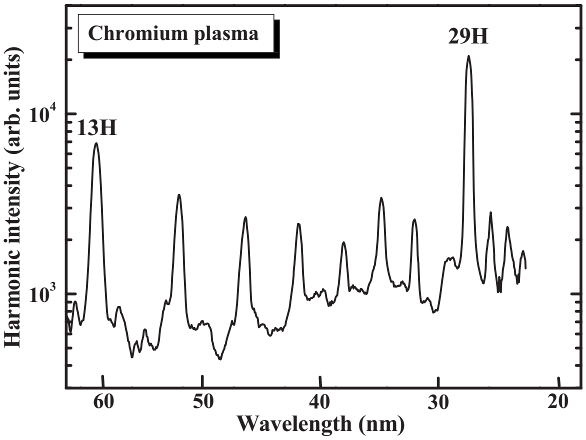

FIG. 4. Harmonic spectra from Cr plasma in the case of 800 nm, 35 fs chirp-free main pulses.

the pump laser for the former case (40 nm, as compared with 20 nm in [16]).

Our experiments with the 400 nm pump laser showed a similar enhancement of a single harmonic, whose wavelength was close to previously observed harmonics that showed intensity enhancement using the 800 nm pump. Figure 6 shows the harmonic spectrum obtained for the 400 nm main pump interacting with chromium plasma. The observed enhanced 15th harmonic (4× enhancement, $\lambda = 26.7$ nm, $E_{\mathrm{ph}} \approx 46.7$ eV) almost coincides with a broad spectral emission of singly charged Cr, which was also responsible for the enhancement of 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, but as in the case with Mn plasma, this did not lead to the tuning of harmonic wavelength and correspondingly did not show the relative change in the 15th harmonic intensity compared to its neighbors.

The calculations of $gf$ values in the photon energy range of 40–60 eV presented in [24] clearly show a group of the transitions in the 44.5–44.8 eV region possessing very strong oscillator strengths (with $gf$ varying between 1 and 2.2), which considerably exceeded those of other transitions in the range of 40–60 nm. 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 considerably decrease the yield of the 27th harmonic.

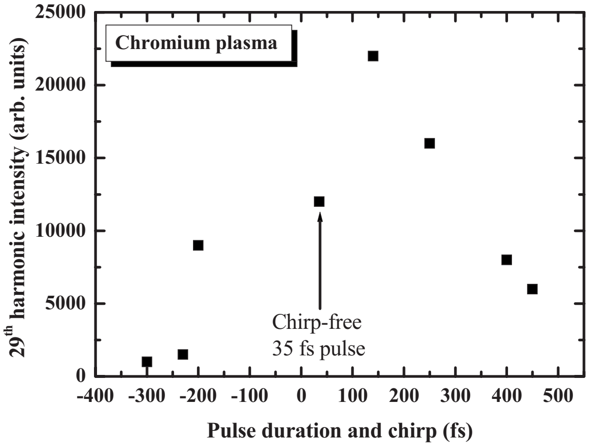

FIG. 5. Variation of the 29th harmonic intensity at different chirps of 800 nm radiation. Chromium plasma.

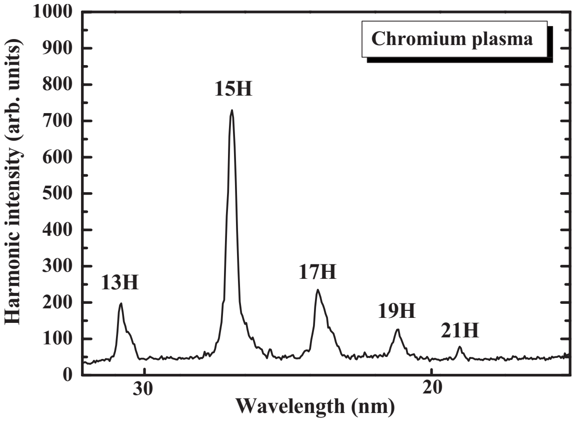

FIG. 6. Harmonic spectrum from Cr plasma in the case of 400 nm driving pulses.

### C. Antimony plasma

The HHG of the antimony atom has previously been investigated using the InSb plume [19], as well as with the pure Sb plume excited on the surface of the solid antimony target [27]. Past works have shown that for InSb plasma, in the case of positively chirped laser pulses, the intensity of the 21st harmonic of the 795 nm laser considerably 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×. 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 the 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 the low ionized antimony laser-ablation plume. The conversion efficiency of this harmonic was reported to be $2.5 \times 10^{-5}$ and the output energy was 0.3 $\mu$J. Such an enhancement of the 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 one centered near 31.24 eV with a bandwidth close to 1 eV (FWHM) and a narrower one centered near 32.22 eV. The spectrum also contains a large population of Sb II, which gives rise to peaks at 32.4 and 32.7 eV. However, the strongest transitions among these Sb I lines calculated and measured in [28], was the $4d^{10}5s^25p^3\,{}^2D_{5/2}$-$4d^95s^25p^4({}^3P)^2F_{7/2}$ transition ($E_{\mathrm{ph}} \approx 31.5$ eV). The $gf$ value of this transition is

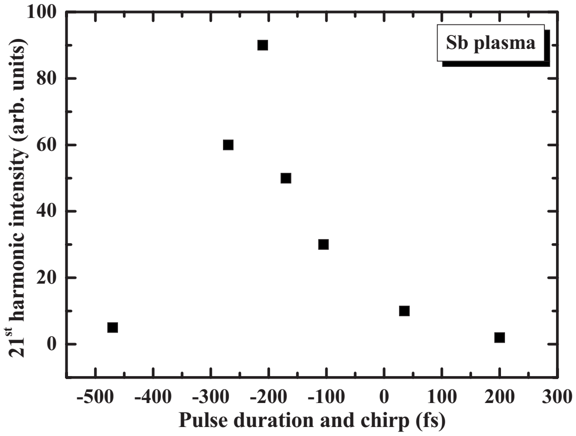

FIG. 7. Variations of the 21st harmonic yield from Sb plasma at different chirps of driving 800 nm radiation.

calculated to be 1.54, which is a 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 the Sb II ion, the $^3P_2$-$(^2D)^3D_3$ transition ($E_{\mathrm{ph}} \approx 32.8$ eV, $gf=1.36$) also shows a strong oscillator strength, which could influence the nonlinear optical response of plasma due to the proximity of the harmonic wavelength with this transition.

These transitions can be driven into resonance with the 21st harmonic (37.8 nm, 32.9 eV) by the ac Stark shift, thereby resonantly enhancing its intensity. It is difficult to calculate the ac Stark shift accurately, but according to Ref. [29] the ac Stark shifts can be several eV in magnitude.

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. Figure 7 presents the variations in the intensity of the 21st harmonic at different chirps of the main pump. One can see that the optimum conversion efficiency is achieved with a negative chirp and pulse duration of 210 fs. For antimony, no enhanced harmonics were observed for the 400 nm pump.

The difference in the results obtained in 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×) was less than those in the refereed works (10× [19] and 20× [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 the pump laser with a narrower bandwidth. These results infer that the use of the pump laser with narrower bandwidth is preferable to increasing the enhancement of the harmonics.

### D. 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. 8). We investigated this phenomenon by shifting the harmonic wavelength in the range of ±0.5 nm relative to the chirp-free position of the harmonic wavelength. The results of these experiments are presented in Fig. 9. One can see the variation of this harmonic yield, which was optimized 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. In the case of tin plasma, the 15× 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 possessing strong oscillator strength.

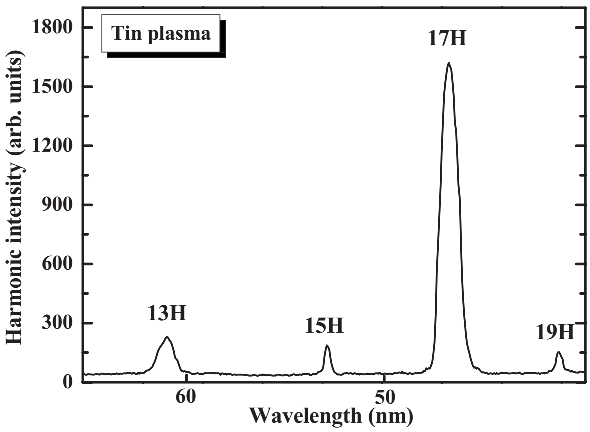

FIG. 8. Harmonic distribution in the case of the Sn plume. $\lambda = 800$ nm.

Recently, such an enhancement was reported and optimized by tuning the central wavelength of the master oscillator of the laser system [30]. In this work, the observation of strong single high-order harmonic generation at the wavelength of 46.76 nm by using a 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 output energy of the 17th harmonic was measured to be 1.1 $\mu$J. The origin of this enhancement was attributed to resonance with a strong radiative transition of the Sn II ion, produced within the laser-ablated plume.

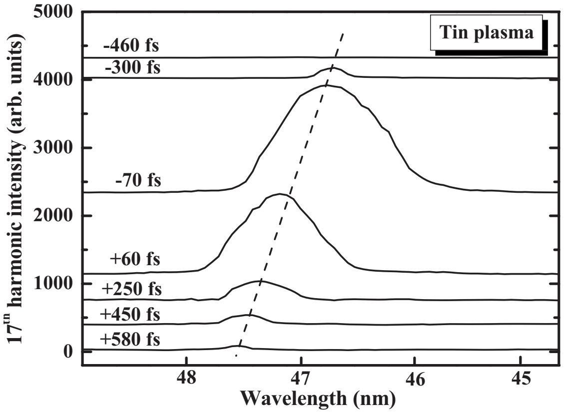

FIG. 9. Tuning of the 17th harmonic at different chirps of driving radiation. Tin plasma.

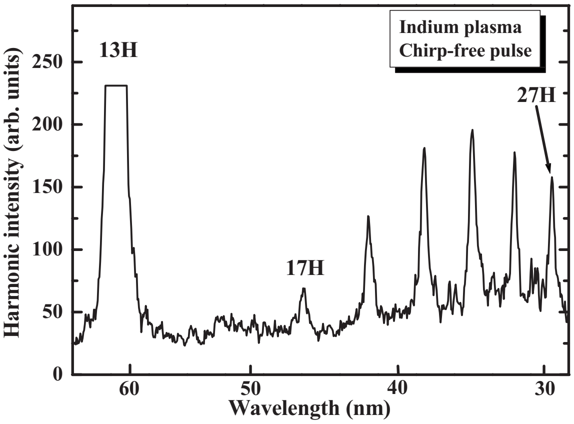

FIG. 10. Low-order harmonic distribution from In plasma by using the chirp-free driving pulses.

In past work, the Sn II ion has been shown to possess 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 ($E_{\mathrm{ph}}=26.24$ eV) [31]. The $gf$ value of this transition has been calculated to be 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.

### E. Indium plasma

Harmonic generation from indium plasma has recently been analyzed in Ref. [19]. Here we present some additional studies of the peculiarities of single harmonic enhancement from this nonlinear medium. As in previous publications [13,19], very strong 13th harmonic (61.5 nm, 20.2 eV) appeared from In plasma with the interaction of the 800 nm, 35 fs chirp-free laser pulses (Fig. 10). At the same time, a considerable suppression of the 15th harmonic was observed under these conditions. Harmonics up to the 35th cutoff were observed and showed a plateaulike harmonic spectrum. As in previous cases, we investigated the effects of chirp of the main pump on the 13th harmonic intensity. As a result, we observed a considerable increase in the intensity of this harmonic using negatively chirped pulses, compared to those generated by the chirp-free pump (Fig. 11). The enhancement factor of the 13th harmonic compared to neighboring ones was estimated to be 180, which is close to previously reported data (80× [13] and 200× [19]).

The indium plasma emission observed in the range of interest is due to radiative transitions between the ground state ($4d^{10}5s^2\,{}^1S_0$) and the low lying state ($4d^{10}5s^25p$) of In II (Fig. 12, curve 2). Among them, the transition at 19.92 eV (62.1 nm), corresponding to the $4d^{10}5s^2\,{}^1S_0 \rightarrow 4d^95s^25p(^2D)^1P_1$ transition of In II, is exceptionally strong. The absorption oscillator strength $gf$ of this transition has been calculated to be 1.11 [32], which is more than twelve times larger than other transitions from the ground state of In II. This transition can be driven into resonance with the 13th harmonics by the ac Stark shift, thereby resonantly enhancing its intensity.

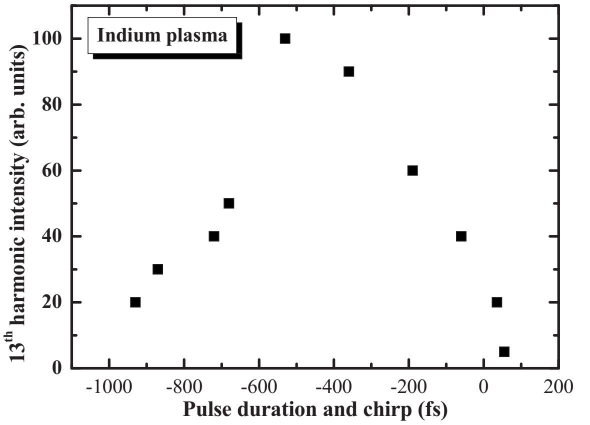

FIG. 11. Variation of the intensity of the 13th harmonic generated from In plasma at different chirps of 800 nm driving radiation.

When we changed the wavelength of the pump laser from 800 to 400 nm, this strong enhancement disappeared. In this case the harmonics from indium plasma demonstrated a featureless spectrum, without any enhanced harmonic, since the wavelength of the 7th harmonic of the 400 nm pump laser was too far from In transitions with strong oscillator strengths.

## IV. DISCUSSION

The photon fluxes of enhanced harmonics considerably differ from each other. Highest conversion efficiency ($\sim 10^{-4}$) was observed in the case of the 13th harmonic generated from the indium plume. The pulse energy of this radiation (61.5 nm) was estimated to be 2 $\mu$J. In the case of the 29th harmonic ($\lambda=27.6$ nm) produced from chromium plasma, the conversion efficiency was estimated to be $2\times 10^{-5}$. Thus the energy of 27.6 nm radiation was assumed to be 0.4 $\mu$J. These studies showed that the yield of an en-

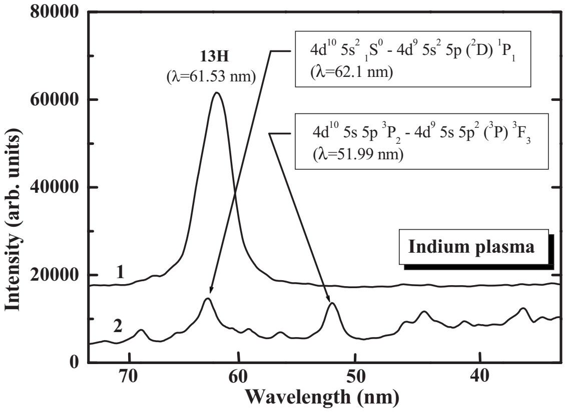

FIG. 12. Spectra of (1) the harmonics generated from In plasma and (2) indium plasma lines. No harmonics other than the 13th harmonic seen in the former spectrum, since the 13th harmonic considerably prevails (×180) over other harmonics in this spectral range.

hanced harmonic strongly depended on the tuning of harmonic wavelength.

The tuning of harmonics can be realized not only by the artificial change of the chirp of driving radiation, but also by the dynamically induced chirp during interaction of the strong laser field with the ionic medium. In particular, the multiphoton ionization of the medium occurring in the laser focus imposes a change of the refractive index due to the free electron contribution. In that case, the refractive index decreases, resulting in a blue shift of harmonics. This phenomenon was reported by different groups (see, for example, [33]). The self-induced redshift of harmonic wavelength was rarely observed in these studies. In particular, in Ref. [34] it was shown that the chirp-induced shifts of harmonics generated in the Xe gas jet in the case of relatively long (300 fs) laser pulses exhibit a trend either toward the blue or to the red, depending on particles density. The redshift was observed at low gas densities. This process might be related to Kerr effect, cluster formation in the gas jet, electron-ion recombination, and ion expansion. The former mechanism was found consistent with the experimental finding reported in Ref. [34]. The chirp-induced redshift and blueshift were explained by the model including both atoms and ions. The neutrals were assumed to be responsible for the blueshift, while the ions were the main reason for the redshift.

The ionic medium imposes both the phase and spatial change of the propagated radiation. Whether the refraction of the beam significant in the plasma depends on the intensity of radiation. Free electrons themselves do not lead to the self-defocusing when their concentration is insignificant and they distributed homogeneously in the area of laser-plasma interaction. The main peculiarity here is a spatial inhomogeneity of the focused radiation. The creation of additional free electrons along the propagation axis (due to considerably higher intensity compared to the wings of the beam) leads to the self-defocusing caused by the gradient of free electron concentration along the beam waist radius. This process also leads to further phase mismatch between the harmonic and fundamental waves. The studies of the plasma nonlinearities leading to the self-defocusing at the conditions of HHG are presented in Ref. [35].

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 [5,6,36]. This technique is considerably distinct from the early approach where the variation of the driving radiation spectrum tuned the harmonic wavelength and adjusted it to coincide with an atomic or ionic resonance in the nonlinear medium [37,38]. 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 the enhancement of 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 in alkali metals [39], where multiharmonic enhancement was reported, and in rare gases [6,36], when the single harmonic intensity was enhanced a few times compared to those of neighboring harmonics. Chirp control of the pump laser allows one to achieve an optimal relation between the quasiresonance conditions, reabsorption, and induced self-defocusing, leading to a considerable enhancement of the single harmonic yield.

The very 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. In the case of plasma from various targets, there is a higher probability to find a proper target for which the fulfillment of multiresonance conditions in the XUV range can lead to the enhancement of the harmonic yield.

It follows from the preceding results that the origin of the strong yield of the single harmonics in the plateau region is associated with the resonance-induced growth of nonlinear optical frequency conversion. Therefore, let us examine the resonance-induced growth mechanism in a little more detail. The role of atomic resonances in harmonic generation had been a main subject of discussion in the early studies of high harmonic generation [40].

Some experimental observations (in particular, the dependences of the harmonic yield on the beam waist position, plasma sizes, and laser radiation intensity) point out the effects related with a collective character of the HHG from laser plumes. Among the factors enhancing harmonic output are the effects related with the difference in the phase conditions for the different harmonics. The phase mismatch condition ($\Delta k = nk_1 - k_i$, where $k_i$ is the wave number of the $i$th harmonic) changes, due to ionization during the propagation of the driving pulse through the plume. According to the estimations, in the plateau region at the same particle concentration the phase mismatch caused by the influence of free electrons is about one to two orders higher than those caused by the influence of atoms and singly charged ions (these relations are the same for all high harmonics). At the resonance conditions, when the harmonic frequency is close to the frequency of the atomic transition, the variation of the wave number of the single harmonic could be considerable, and the influence of free electrons can be compensated by the atomic dispersion for specific harmonic order. In this case the improvement of the phase conditions for a single harmonic generation can be achieved.

The question arises as to why a multiphoton resonance with some excited state of ions leads to a pronounced resonance in the harmonic spectrum while a multiphoton resonance with other excited states does not? The numerical simulations of this problem were reported in Ref. [41]. One can understand that the competition between the reabsorption, phase matching (mismatching), and growth of harmonic intensity, as well as the population of the excited states and the transition lifetime have to be at "optimal" conditions to show the resonance-induced growth of the single harmonic in the plateau region. A few types of plumes can satisfy these conditions at a given condition of the pump laser. Further search of plumes demonstrating high enhancement of the single harmonic seems to be very important for extending the highest enhanced harmonic to the shorter-wavelength range.

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 effects and/or coherent control of HHG, one will 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, the possibility of high (up to kHz) repetition rate, and improved conversion efficiency. Such a unique radiation source will truly 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.

## V. 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 appropriate 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, in accordance with previous photoabsorption studies of the above plumes. The enhancement of the 13th, 17th, 21st, 29th, and 33rd harmonics from the above targets was obtained and analyzed using the 800 nm femtosecond laser with various chirp. We also presented the observation of harmonic enhancement from some targets in the case of a second harmonic pump laser (400 nm central wavelength). Using Mn plume, we demonstrated the highest harmonic photon energy (52.9 eV) at which single enhancement has been observed (17th order, $\lambda = 23.5$ nm).

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