High-order harmonic generation in laser-produced ions

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We have investigated the high-order harmonic generation of a subpicosecond KrF laser (248 nm) using nine ionic species in laser-produced plasmas. The highest order of the 21st harmonic (11.8 nm) has been observed in a lead plasma. The maximum observed harmonic orders in various ions and neutral rare gases are found to be proportional to the ionization potentials of these species. We have obtained superior results for the high-order harmonic generation by using ions compared to using neutral rare gases, and have demonstrated the advantages of using ions as nonlinear media to generate extreme ultraviolet radiation.

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I. INTRODUCTION

The recent progress of ultrashort, high-power lasers allows the generation of high-order harmonics in the extreme ultraviolet (XUV) and soft x-ray spectral regions [1–8]. Much effort has been done to obtain short-wavelength coherent light by the high-order harmonic generation. The practical limit of the high-order harmonic conversion, however, is the multiphoton ionization or tunneling ionization of a nonlinear medium exposed in a high-intensity laser field. There are several approaches in order to overcome such ionization processes for the high-order harmonic generation. Macklin, Kmetec, and Gordon [7] have recently observed the 109th harmonic of a 125-fs Ti: sapphire laser (806 nm) in Ne. An ultrashort pulse laser allows an atom to experience a high peak laser field prior to the onset of the ionization. Using a subpicosecond KrF laser (248 nm), Sarukura *et al.* [8] observed the 25th harmonic radiation (9.9 nm) in Ne. Short-wavelength lasers such as a KrF excimer, however, cause faster multiphoton ionization of a nonlinear medium compared to long-wavelength lasers, that eventually limits the conversion efficiency of the high-order harmonic radiation.

In all of the high-order harmonic experiments, nonlinear media used were neutral rare gases. In this paper, on the other hand, we have studied ionic species as nonlinear media for the high-order harmonic generation. The smaller polarizability of ions compared to that of atoms might be a drawback for the high-order harmonic generation. According to the experimental results using a Nd:glass laser by Lompré, L'Huillier, and Mainfray [4], however, they have pointed out that the maximum harmonic orders observed increased with the increase of the ionization potentials of rare gases. This result simply indicates that the use of new nonlinear media with higher ionization potentials should lead to the higher-order harmonic generation compared to the use of conventional rare gases despite the smaller polarizability of ions. In other words, the faster ionization rate of a nonlinear medium with a lower ionization potential would limit the high-order harmonic conversion. We have, therefore, proposed and successfully demonstrated the high-order harmonic generation by using nonlinear media with high ionization potentials such as alkali-metal ions in laser-produced plasmas [9].

The use of ions as nonlinear media for the short-wavelength generation was first proposed by Harris in 1973 [10]. No experimental demonstration, however, has been reported on the nonlinear frequency conversion using ions in laser-produced plasmas prior to our work [9].

Laser-produced plasmas to provide ions as nonlinear media have various advantages in terms of media preparation. The timing of a plasma-initiating laser pulse can be readily controlled with respect to the timing of a high intensity laser pulse by using two independently controlled lasers. This implies that a plasma with a certain ion number density with a certain charge state or a certain plasma temperature could be produced by optimizing the delay time between the two laser irradiations. Furthermore, by changing a focusing geometry of a plasma-initiating laser, a spatially localized medium can be produced that should control a phase-match condition and solve a window problem.

The success of alkali ions as nonlinear media [9] motivated us to study other ionic species in addition to three alkali metals (Li, Na, and K). Various elements with a wide variety of atomic numbers and electron configurations could provide important information on their roles for the high-order harmonic generation. We have thus chosen six solid elements as nonlinear media other than three alkali metals. They are boron, carbon, titanium, copper, tungsten, and lead.

In this paper, we present experimental investigations on the high-order harmonic generation of a high-power

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subpicosecond KrF excimer laser (248 nm) using several ionic species as nonlinear media. We successfully demonstrate the high-order harmonic generation using these ionic species and show the superiority of using such ions as nonlinear media for the generation of the extreme ultraviolet radiation.

II. EXPERIMENTAL ARRANGEMENT

Figure 1 shows the experimental apparatus for the high-order harmonic generation using laser-produced ions. Two KrF lasers (248 nm) were used in this setup: one for the plasma production to prepare ions as nonlinear media and the other as a fundamental radiation source for the high-order harmonics.

The plasma production was done by using a conventional KrF laser that was loosely focused onto a rotating solid target placed inside a vacuum chamber. The laser produced 200-mJ pulses with a pulse width of 20 ns. The focused laser intensity was adjusted around 10^9 W cm^{-2} in order to selectively produce rare-gas-like alkali ions and various metal ions with desired charge states. An estimated electron temperature in a plasma was approximately 1.5 eV and this temperature was high enough to produce ions predominantly in plasmas. The photon energy of the plasma-initiating laser (5 eV) was close to the first ionization potentials of alkali-metal atoms used in this experiment. This also excludes the possibility of existing neutrals in alkali-metal plasmas. Ions predominantly existed in such a plasma with its density one order of magnitude larger than that of neutrals even after a 100-ns delay time when a high intensity driving laser was irradiated in such a plasma. The plasmas produced by the laser adiabatically expanded into vacuum inside a vacuum chamber. The plasma length on the target could be varied as long as 2 mm.

A 5-mJ pulse energy of a 0.5-ps linearly-polarized KrF laser [11] was boosted to 50 mJ by using a KrF discharge amplifier (Lambda Physik, EMG 401). In order to minimize a prepulse caused in the amplifier, we installed a vacuum spatial filter consisting of a 100-μm pinhole and a pair of 60-cm focal length CaF_2 lenses. The prepulse contrast ratio of the focused subpicosecond laser intensity was more than 10^9.

After a certain delay from the 20-ns plasma-initiating laser pulse, the subpicosecond laser pulse was orthogonally focused into a plasma at 1 mm from a target surface. A maximum focused laser intensity by using a 30-cm focal length achromatic lens was estimated to be 10^{17} W cm^{-2}. The confocal parameter of this focusing geometry was thus 2 mm, that was comparable to a plasma length.

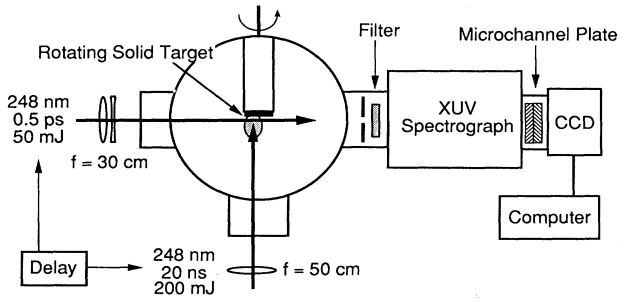

FIG. 1. Experimental setup for the high-order harmonic generation. A nonlinear medium is provided in a laser-produced plasma. CCD denotes a charge-coupled-device camera.

The harmonic signal was detected using either a flat-field normal incidence or a flat-field grazing incidence XUV spectrograph coupled with a two-stage microchannel plate (Hamamatsu Photonics, F2224-21PFFX). A spectral range between 3 and 120 nm was covered by using two spectrographs. The spectral image of the microchannel plate was monitored by a charge-coupled-device (CCD) camera. A width of an entrance slit was 100 μm and the typical spectral resolutions of these XUV spectrographs were 0.05 and 0.07 nm for the normal incidence and for the grazing incidence spectrograph, respectively. Either a 100-nm thickness carbon filter (Luxel) or a filter made of a 150-nm aluminum-silicon alloy overcoated with carbon (Luxel) was placed behind an entrance slit of each spectrograph in order to minimize the undesirable scattered light to the detector. The CCD camera was interfaced to a computer for further data analysis and processing.

In order to compare the harmonic conversion efficiency using ions to that using neutral rare gases, we measured the high-order harmonics using rare gases with a similar arrangement described in Fig. 1. In this case, a rotating target and a plasma-initiating laser were replaced by a pulsed supersonic valve (R. M. Jordan) with a backing pressure of 9 atm. We used He, Ne, and Ar as nonlinear media for the harmonic generation. The number densities of these rare gases were 3×10^{17} cm^{-3}.

III. EXPERIMENTAL RESULTS

Figure 2 shows a typical time-integrated spectrum obtained in a lead plasma at a laser intensity of 10^{17} W cm^{-2}. This spectrum was taken with accumulating 30 laser shots. The harmonics up to the 21st order were ob-

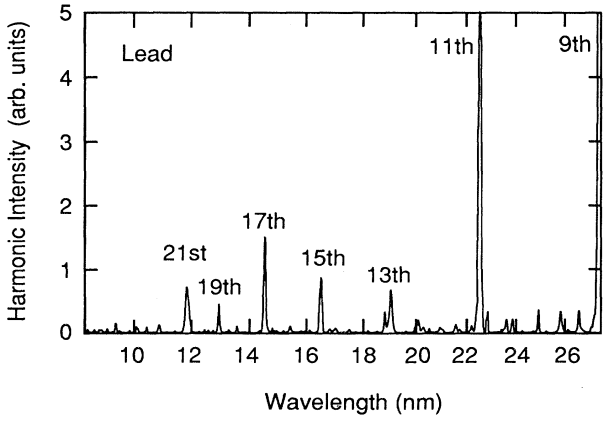

FIG. 2. Time-integrated high-order harmonic spectrum observed in a lead plasma. The spectral peaks are labeled by the harmonic orders. This spectrum has not been corrected for the spectral response of the detection system.

served, that were clearly separated from lead ionic lines.

It was necessary to estimate the number density of ions in a laser-produced plasma to make a quantitative comparison of the high-order harmonic generation using ions and neutral atoms. When a subpicosecond KrF laser was irradiated in Li^+, a series spectrum of He-like Li^+ ($np\,1s \rightarrow 1s^2$) was observed as well as the high-order harmonics. The intensity of the series spectrum was weaker compared to that of the high-order harmonics. From the series spectrum, the electron density could be estimated with a formula [12]: $\log_{10} n_e = 23.26 - 7.5\log_{10} n_s + 4.5\log_{10} z$, where n_e is the electron density to be evaluated, n_s the principal quantum number where a continuum starts (Inglis-Teller limit), and z the ionic charge.

Figure 3 shows the delay time dependence of the electron density in a lithium plasma calculated with this formula. The delay in Fig. 3 corresponds to the time of the subpicosecond laser irradiation after the nanosecond plasma-initiating laser pulse. The high-order harmonics were mainly observed at a delay time of approximately 100 ns, where the electron density was 5×10^{17} cm^{-3}. The He-like Li^+ density, therefore, was estimated to be around $(2-5)\times10^{17}$ cm^{-3}. This value is almost the same as the rare gas density provided by the supersonic jet. (See Sec. II.) The geometrical interaction lengths for using rare gases and alkali ions were also the same and the confocal parameter of the subpicosecond KrF laser was 2 mm for both cases.

Figures 4(a) and 4(b) show the harmonic intensity distributions for rare gases and alkali-metal ions, respectively. A relative intensity of each harmonic order was calibrated by taking account of the spectral response of the detection system. In this measurement, we observed no obvious contributions of rare gas ions for the high-order harmonic generation as pointed out by Xu, Tang, and Lambropoulos [13]. The theoretical calculation of the harmonic distribution in He in Ref. [13] well reproduces our experimental result of He.

The maximum harmonic orders observed in alkali-metal ions were the 17th (14.6 nm) for Li^+, the 19th (13.1 nm) for Na^+, and the 13th (19.1 nm) for K^+ [Fig. 4(b)]. This result shows a significant improvement compared to our previous result [9], where the maximum harmonic orders observed were the 9th for Li^+, the 11th for Na^+, and the 13th for K^+ at a laser intensity of two orders of magnitude lower. The laser intensity of 10^{15} $W\,cm^{-2}$ in our previous experiment was well below the saturation intensity for these nonlinear media with high ionization potentials [9]. The increase of the maximum harmonic order observed will be possible as the increase of the laser intensity as long as it is in the unsaturated regime that will be discussed in Sec. IV.

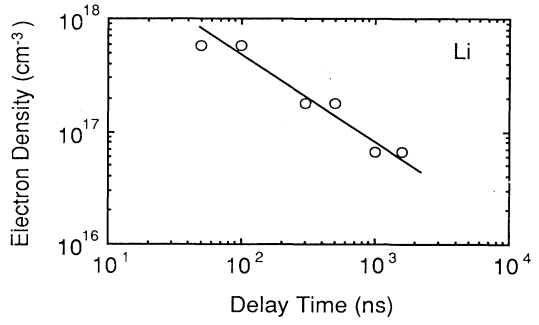

FIG. 3. Measured electron density in a lithium plasma as a function of the delay time between the nanosecond and subpicosecond KrF laser pulses. The electron density was evaluated from a series spectrum of He-like Li^+.

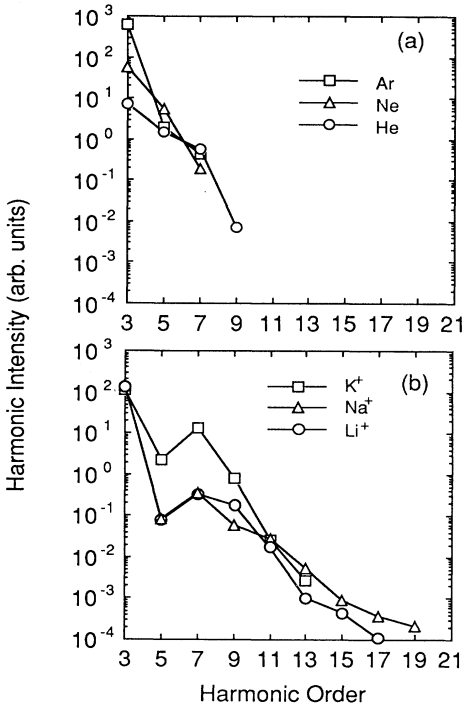

FIG. 4. High-order harmonic intensity distributions of (a) He, Ne, and Ar, and of (b) Li^+, Na^+, and K^+. The KrF laser intensity is 10^{17} $W\,cm^{-2}$. The number densities of three rare gases are 3×10^{17} cm^{-3}.

Since the number densities of the nonlinear media for ions and neutrals were almost the same and the interaction length was also similar for both cases, the results in Figs. 4(a) and 4(b) can be directly compared in terms of the maximum harmonic orders observed and their relative intensities. The comparison of the nonlinear media indicates that the alkali-metal ions are superior to rare gases for the high-order harmonic generation when a short wavelength fundamental laser (248 nm) is used.

The superiority of ions as nonlinear media for the high-order harmonic generation was further verified by using various metal ions. We expect that the use of various nonlinear media could provide important information on their roles for the high-order harmonic generation. Six solid target materials were chosen with various atomic numbers other than three alkali metals as discussed above, namely, boron ($Z=5$), carbon (6), titanium (22), copper (29), tungsten (74), and lead (82).

Figure 5 compiles the harmonic distributions of using these six elements. For each species a dominant charge state is evaluated using a collisional-radiative model

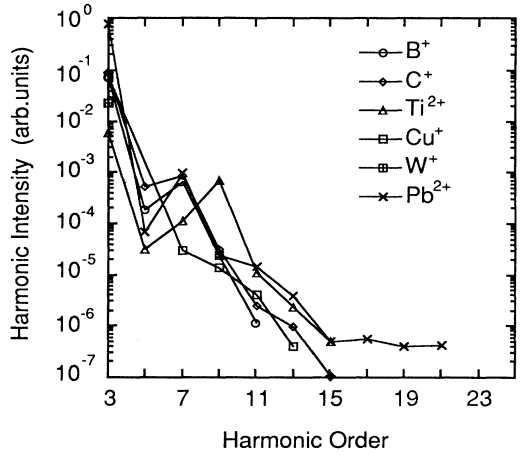

FIG. 5. High-order harmonic intensity distributions of six ionic species. The KrF laser intensity is 10^{17} W cm^{-2}. The dominant charge state of each species is indicated in the figure.

[9,14] and is shown in the figure. The maximum orders observed were the 11th (22.6 nm) for boron, the 15th (16.6 nm) for carbon and titanium, the 13th for copper, the 3rd (82.8 nm) for tungsten, and the 21st (11.8 nm) for lead. Although the maximum orders of harmonics observed vary from the third to the twenty-first, the overall harmonic distributions and their relative intensities are similar to each other. This suggests that the laser-produced plasmas were very effective in terms of preparing nonlinear media for the high-order harmonic generation regardless of the physical and chemical characteristics of the elements.

Figure 6 shows the comparison of harmonic distributions when lead and sodium ions were used as nonlinear media. The charge state estimate of these ions [9,14] shows that Na^+ and Pb^{2+} are dominant species in sodium and lead plasmas, respectively, when the nanosecond

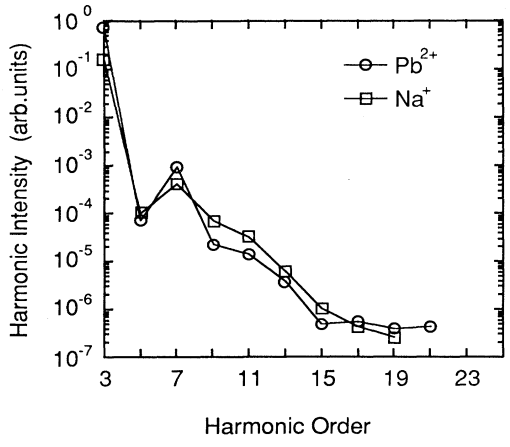

FIG. 6. Comparison of the harmonic intensity distributions of lead and sodium ions. In spite of a large difference of the atomic numbers, the harmonic intensity distributions are very similar to each other.

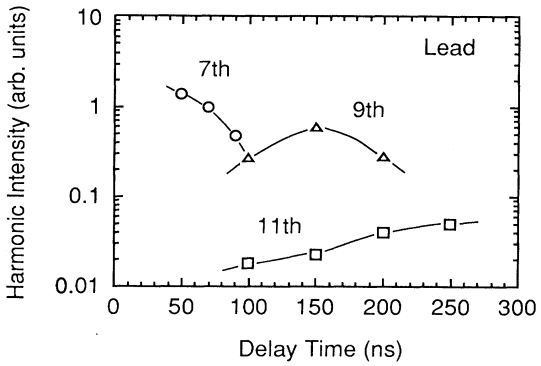

FIG. 7. Harmonic intensities of three neighboring orders observed in a lead plasma as a function of the delay time.

plasma-initiating laser intensity on the target was 10^9 W cm^{-2}. The maximum harmonic order observed using a lead target was the 21st; on the other hand, the sodium ions produced the 19th harmonic as described previously. Note that both harmonic distributions are very similar to each other. The large difference of the atomic numbers ($Z=11$ for sodium and 82 for lead) makes little difference on the harmonic distribution. The ionization potentials of Na^+ is 47.3 eV; on the other hand, Pb^{2+} has a 31.9-eV ionization potential. The electron configurations of the dominant ionic species are subshell closed ($2p^6$ for Na^+ and $6s^2$ for Pb^{2+}). It is thus conceivable that even the high-order harmonics come from the interaction between valence electrons and the driving laser field.

Since the delay time of the pulses from the plasma-initiating laser and high-intensity laser was readily controlled, the optimum conditions of certain orders of the harmonics were investigated. Figure 7 shows the delay time dependence of three neighboring harmonic intensities in a lead plasma. The increase of the delay time corresponds to a more tenuous lead ion density since the lead plasma hydrodynamically expands into vacuum. The 7th harmonic favors the shorter delay, namely, a denser plasma; on the other hand, the delay time dependence of the 11th harmonic intensity shows the opposite behavior. The 9th harmonic shows the intermediate behavior between the 7th and 11th harmonics.

IV. DISCUSSION

When using ions as nonlinear media, coexisting free electrons in a plasma would influence the harmonic beam propagation. For the high-order harmonics the phase mismatch in a plasma would be in general determined by free electrons and not by ions. The collective positive phase mismatch due to free electrons is expressed as $\Delta k=(q^2-1)\omega_p^2/2qc\omega$, where ω_p is the plasma frequency, q the order of harmonics, c the speed of light, and ω the laser frequency [15]. The expression of the phase mismatch for the high-order harmonic propagation becomes more and more ambiguous in the strong-field limit [16], nevertheless, we estimate the positive phase mismatch due to free electrons and compare the coher-

ence length defined by the phase mismatch with the confocal parameter of the fundamental laser radiation.

Figure 8 shows a calculated phase mismatch, Δk, induced by free electrons as a function of a harmonic wavelength. The free electron density of 10^{17} cm^{-3} is assumed based on the experimental measurement described in the preceding section. The confocal parameter of the subpicosecond laser pulse is 2 mm that should be comparable to the coherence length, $2\pi/\Delta k$. The acceptable phase mismatch, therefore, would be 30 cm^{-1}. For the wavelength of a KrF laser (248 nm), the phase mismatch would be acceptable up to the 25th harmonic or the wavelength longer than 10 nm; on the other hand, for the 1-μm laser, the phase match would be satisfied at the harmonic wavelength longer than 100 nm under the same electron density. The use of short-wavelength lasers is thus advantageous to satisfy the phase match condition when a laser-produced plasma is used as a nonlinear medium where free electrons exist. In order to verify this collective phase match condition, the high-order harmonic generation in ions using a high-power Ti:sapphire laser is under way in our laboratory.

Krause, Schafer, and Kulander [17] first pointed out that the ions could also produce harmonics comparable in strength to those obtained from neutral atoms and that emission extends to much higher order under a very intense laser field. Xu, Tang, and Lambropoulos [13] also presented the rare gas ion contribution to the high-order harmonic generation for the KrF laser radiation. Ionization of a nonlinear medium caused by a pump laser, however, leads to the decrease of an effective laser intensity at the focus [18]. When a nonlinear medium is ionized, created free electrons nonuniformly distribute in a localized plasma along the laser beam radius and spatially modulate the index of refraction of the medium. If the laser profile at the focus is Gaussian-like, the free-electron density distribution at the focus acts as a concave lens. Then the incident laser beam will be defocused and the effective laser intensity decreases. Ions produced by an intense laser field thus could not contribute to the high-order harmonic generation [6,7]. Our experimental results using rare gases indeed indicate no contributions of the ions for the high-order harmonic generation.

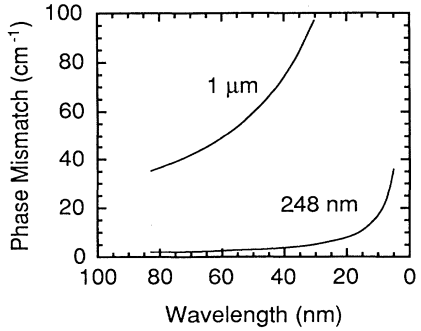

FIG. 8. Estimate of phase mismatch induced by free electrons in a plasma assuming an electron density of 10^{17} cm^{-3} as a function of the harmonic wavelength. Fundamental laser wavelengths are assumed to be 1 μm and 248 nm.

Our laser-plasma method has an advantage to reveal the contribution of ions for the high-order harmonic generation without being much affected by beam defocusing since ions are uniformly preformed in a plasma prior to the high-power laser radiation. Furthermore, for the alkali-metal ions, they have higher ionization potentials compared to rare gases and the other ions used in this measurement; therefore, the probability of the ionization would decrease, leading to less significant beam defocusing.

Among many ionic species used, we, however, have observed a signature of the beam defocusing in a lead plasma. Figure 7 shows the delay time dependence of the three neighboring harmonic order intensities in a lead plasma. The low-order harmonic (7th) favors the shorter delay time where a larger number density of the lead ions is expected in the plasma. On the other hand, the 11th harmonic intensity increases as the lead ion number density decreases. The higher-order harmonics should require higher laser intensities; therefore, it is conceivable that the effective laser intensities that the medium could experience should become higher when the delay time increases. This behavior could be explained by the decrease of the effective laser intensity when the delay time is short due to laser beam defocusing. In a separate experiment, we have observed a measurable beam defocusing in the lead plasma when the delay time was less than 100 ns.

Most of the experimental studies of the high-order harmonic generation have been supported by numerous theoretical investigations [13,15–17,19–22], where a many-atom response such as a phase matching is considered and a single-atom response is rigorously calculated. The several nonperturbative single-atom calculations show that a spectrum emitted from a single atom under strong laser fields agrees well with experimental results.

In order to support the advantage of the use of ions with high ionization potentials, we estimate the effective laser intensities of three alkali-metal ions for the high-order harmonics using a model proposed by Becker, Long, and McIver [19], where the single-atom response is analytically described using only ionization potentials of media. The intensity distributions of the harmonics were calculated by changing the subpicosecond 248-nm laser intensities as a fitting parameter. The best fits for the three alkali-metal ions give the effective intensities of 2.0×10^{15} W cm^{-2}, 6.0×10^{14} W cm^{-2}, and 1.6×10^{14} W cm^{-2} for Li^+, Na^+, and K^+, respectively. The difference of the evaluated laser intensities can be explained by the difference of the ionization potentials of the ions, since the higher ionization potentials make the ionization threshold shift to the higher laser intensities.

The estimated effective laser intensities are smaller by a factor of 4 to 5 compared to those evaluated assuming a tunneling ionization theory by Ammosov, Delone, and Krainov [23]. The tunneling ionization rate is a function of both the ionization potential and the charge state of a medium [23,24]; on the other hand, the multiphoton ionization is simply governed by the ionization potential of a medium [25]. The evaluated adiabaticity parameter [26] indicates that even if one takes the tunneling theory and estimates the effective laser intensities for the nonlinear

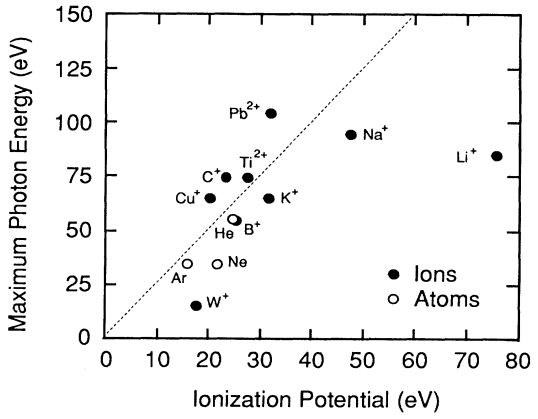

FIG. 9. Maximum photon energy observed ($\hbar\omega_{max}$) as a function of the ionization potential (I_p) of ionic and neutral nonlinear media used in this investigation. The dotted line shows an empirical relation of $\hbar\omega_{max}=2.5I_p$.

media using the KrF laser wavelength, the multiphoton ionization would be dominant compared to the tunneling ionization for all media except a lithium ion.

If the multiphoton ionization would limit the maximum harmonic order, the maximum order observed should be a simple function of the ionization potential only, and the difference of the charge state should have little effect for the maximum harmonic orders. Figure 9 summarizes the maximum observed harmonic orders as a function of the ionization potentials of ionic and neutral species. Except for the lithium ion, the maximum harmonic orders are found to be proportional to the ionization potentials of the nonlinear media as we expect, giving an empirical relation of $\hbar\omega_{max}=2.5I_p$, where $\hbar\omega_{max}$ is the maximum photon energy observed and I_p is the ionization potential of a medium. The rare gas data described in [4] follow this relation very well; however, a recent result using a Nd:glass laser shows a steeper slope and follows a theoretical relation [17] of $\hbar\omega_{max}=I_p+3.2U_p$, where U_p is the ponderomotive potential. This seems to be consistent since a short-wavelength laser and/or low intensity laser makes less significant ponderomotive energy shift. From Fig. 9, we have concluded that the multiphoton ionization limits the maximum harmonic orders of a short-wavelength KrF laser using the ionic and neutral species except lithium ions. This is a contrast to the results using long-wavelength lasers where the ionization process is dominant by tunneling ionization.

Since the lithium ion has a much higher ionization potential (75.6 eV) compared to the other species, the multiphoton ionization would be suppressed even for the KrF photon energy, and the tunneling ionization would be a limiting process for the high-order harmonics. Then the lithium ion could experience higher laser intensities than those currently available in our laboratory. The high ionization potential of the lithium ion also implies the low probability of the further ionization of the ion, resulting in less beam defocusing in the medium. The high-order harmonics could be realized by using the lithium ion as a nonlinear medium and higher laser intensities.

V. SUMMARY

We have used various ions in laser-produced plasmas as nonlinear media for the high-order harmonic generation in order to minimize the ionization processes, and have observed the high-order harmonics of a subpicosecond KrF excimer laser. The highest harmonic order observed was the 21st (11.8 nm) in lead ions. The maximum observed harmonic orders in various ions and neutral rare gases are found to be proportional to the ionization potentials of these species. Although initially prepared electrons in a laser-produced plasma could affect the phase-match conditions of the high-order harmonics, the phase-match condition was satisfied by using a short-wavelength KrF laser as a fundamental radiation source. The experimental compilation of the maximum harmonic orders and numerical calculations indicate that the higher-order harmonics would be plausible by using nonlinear media with higher ionization potentials.

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