Generation of High-Order Harmonics Using Laser-Produced Rare-Gas-Like Ions

Yasuhiro Akiyama,[a] Katsumi Midorikawa, Yuuki Matsunawa,[a] Yutaka Nagata,[a] Minoru Obara,[a]
Hideo Tashiro, and Koichi Toyoda
RIKEN, The Institute of Physical and Chemical Research, 2-1 Hirosawa, Wako-shi, Saitama 351-01, Japan
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We report the first observation of high-order harmonic waves from rare-gas-like ions excited by a subpicosecond KrF excimer laser. At an intensity of 10^{15} W/cm^2, the highest orders of observed harmonics are the ninth (27.6 nm) in Li^+, the eleventh (22.6 nm) in Na^+, and the thirteenth (19.1 nm) in K^+. The advantage of such ions as nonlinear media for extreme ultraviolet generation is discussed in relation to ionization potentials.

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The recent development of ultrashort, high-power lasers allows the generation of high-order harmonic waves in the extreme ultraviolet (XUV) and soft x-ray regions [1-6]. Using a Nd:glass laser, the high-order harmonics up to the 53rd (19.9 nm) were observed in Ne by Ferray and co-workers [2-4]. Miyazaki and Sakai [5] observed the harmonics up to the 41st (15.0 nm) with the use of a visible dye laser (616 nm). In these experiments, the appearance of a plateau in the intensity distribution among harmonic orders was found. On the contrary, such a plateau did not appear with the KrF-excimer-laser excitation (248 nm) by Sarukura *et al.* [6], which attained the shortest wavelength of harmonic generation, 9.9 nm at the 25th order.

Generally speaking, the use of shorter-wavelength lasers in experiments on harmonic generation has an advantage for XUV generation because of the reduction of harmonic orders [1,6]. For pumping with KrF or XeCl lasers, however, multiphoton ionization of the medium becomes serious and limits the maximum input power. According to the experiments using a Nd:glass laser by Lompré, L'Huillier, and Mainfray [4], there was a saturation in the highest order observed for increasing pumping power. They also found that the saturated order of the harmonics depended strongly on the ionization potential, so that the highest order of harmonics increased linearly with increasing ionization energy of rare gases. Therefore, much-higher-order harmonics would be expected if a nonlinear medium with a higher ionization potential were available. From this point of view, single-ionized alkali-metal ions such as Li^+, Na^+, and K^+ are chosen because they are isoelectric to neutral rare-gas atoms, and they have higher ionization potentials than those of rare-gas atoms. The ionization potentials of Li^+, Na^+, and K^+ are 75.6, 47.3, and 31.6 eV, respectively, while those of rare gas atoms are 24.6, 21.6, and 15.8 eV for He, Ne, and Ar, respectively. The increase of ionization potential, however, results in a decrease of the nonlinear response of the medium [7]. Therefore, high laser intensities and high number densities of the medium are necessary for media with high ionization potentials. In this Letter, we demonstrated harmonic generation with the rare-gas-like ions of Li^+, Na^+, and K^+, using a subpicosecond KrF excimer laser. Such ions are produced and utilized in laser plasmas. The effect of coexisting free electrons on phase matching is also discussed.

Figure 1 shows the experimental arrangement. Two KrF excimer lasers were used; one initiated a plasma to produce rare-gas-like ions, and the other was a subpicosecond KrF laser for the harmonic generation. The plasma-initiating laser produced a pulse of 200 mJ in 20 ns (FWHM), which was focused onto a rotating target of an alkali metal by using an $f=500$ mm lens. We optimized a focused intensity around 10^9 W/cm^2, so that all the ions in the plasma turned out to be rare-gas-like as described below. The subpicosecond laser delivered a pulse of 6 mJ in 500 fs [8]. The pulse width was measured by a multishot autocorrelator based on two-photon absorption of a 5-mm-thick BaF_2 plate [9]. By using an achromatic lens with $f=300$ mm, a subpicosecond pulse was focused into a plasma column at 1 mm from the surface of the rotating target. The laser intensity at the focus was 2×10^{15} W/cm^2. Two types of XUV spectrographs were used to cover the whole spectral region of high-order harmonics. In a spectral range from 3 to 30 nm, a flat-field grazing-incident XUV spectrograph with a varied-spacing concave grating (1200 lines/mm) was used. The lowest-order harmonic observed by this spectrograph was the ninth. To observe harmonics lower than the ninth, we used a flat-field normal-incident XUV spec-

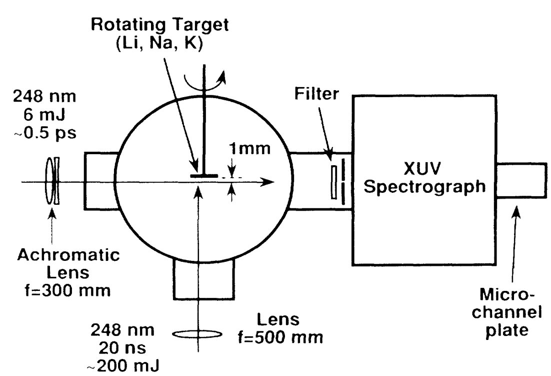

FIG. 1. Schematic diagram of the experimental setup.

trograph equipped with a varied-spacing concave grating blazed at 60 nm (1200 lines/mm), which covered the spectral range from 30 to 120 nm. A two-stage microchannel plate (Hamamatsu Photonics F2224-21PFFX) with a phosphor screen was used as a detector. The spectral image through a fiber-optic coupler was detected by a linear array detector. A thin-film filter made of an aluminum-silicon alloy overcoated with carbon (Acton Research) was inserted in front of the entrance slit of the spectrographs in order to minimize undesirable scattering light. We also experimented with rare gases instead of rare-gas-like ions in the same experimental apparatus, except for the nonlinear medium, to compare the efficiency of high-order harmonic generation. Rare gases were supplied by a pulsed supersonic valve (R. M. Jordan company) with backing pressure of 9 atm. A subpicosecond laser pulse was focused at 4 mm from the surface of the nozzle with a 0.1-mm-diam hole. The estimated gas density in the interaction region was 3×10^{17} cm^{-3}.

Neutral alkali metals have ionization potentials smaller than 5.4 eV, a value which is comparable to the photon energy of a KrF excimer laser (5 eV). Therefore such atoms will be ionized to rare-gas-like ions by the irradiation of a KrF laser for plasma production. We calculated the charge-state distribution in an alkali-metal plasma, using a collisional-radiative model [10]. The distributions calculated under the assumption of an electron density of 10^{16} cm^{-3} are shown in Fig. 2. It was found that singly ionized ions would be dominant over a wide range of the electron temperature of a plasma. The electron temperature was estimated to be approximately 1.5 eV [11], at a focused intensity of 10^9 W/cm^2. Little change is found in the calculated charge-state distributions even when the assumed electron densities are varied over a few orders of magnitude. Consequently, singly ionized alkali-metal ions will be dominant under our experimental conditions.

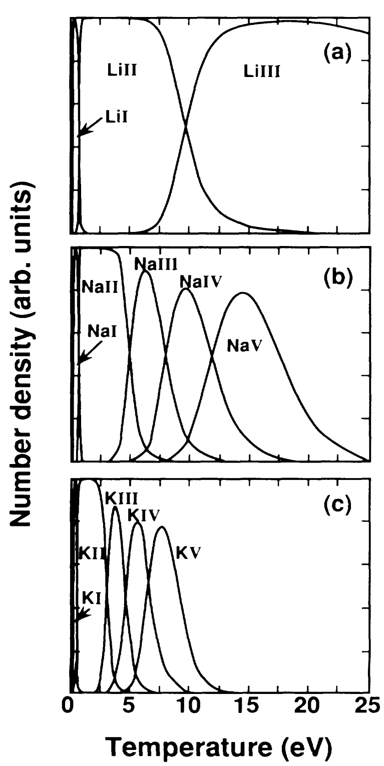

FIG. 2. State of charge density as a function of electron temperature for (a) lithium, (b) sodium, and (c) potassium.

The relative intensities of observed harmonics in both the rare gases and the rare-gas-like alkali-metal ions are shown in Fig. 3. The seventh and the ninth to thirteenth harmonics were observed by two different types of spectrographs as mentioned above. The intensities between the two spectrographs were not calibrated because of no overlapping signals. The relative intensities of the ninth to thirteenth harmonics were calibrated by taking account of the spectral response of the detection system. The highest order of harmonics observed was the thirteenth in K^+, while the harmonics up to the eleventh in Na^+, the ninth in Li^+ and He, and the seventh in Ar and Ne were also found. The wavelengths of the seventh, ninth, eleventh, and thirteenth harmonics were 35.5, 27.6, 22.6, and 19.1 nm, respectively. Among the nonlinear media used, the K^+ ion provided the highest intensity for all orders of harmonics.

We expected that the highest order of harmonics would have been observed in Li^+, which has the highest ionization potential. However, K^+ showed superior results to those of Na^+ and Li^+. As shown in Fig. 4, the output intensity of the ninth harmonic wave in K^+ is proportional to $I^{3.5}$, and is not saturated in this intensity region. Here I denotes the incident laser intensity. The conversion efficiency of harmonics is mainly limited by the nonlinear response of the medium under the unsaturated region. The nonlinearity of K^+ will be much larger than

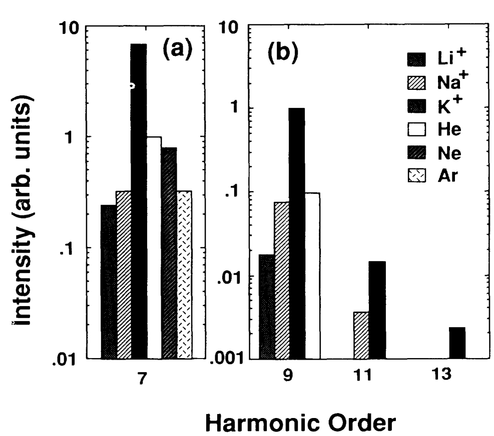

FIG. 3. Relative intensities of harmonics in the rare gases and the alkali-metal ions. The seventh and ninth to thirteenth order harmonics were observed by using normal- and grazing-incidence spectrographs, respectively.

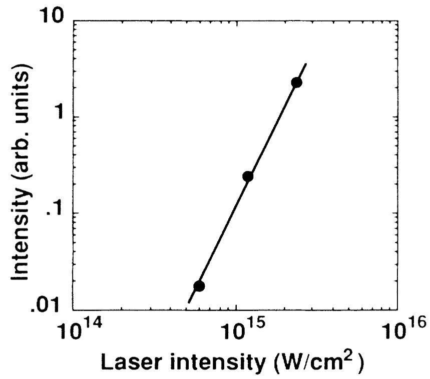

FIG. 4. Ninth harmonic output as a function of focused laser intensities.

that of the Li^+, because the ionization energy of K^+ is nearly a half of that of Li^+ [7,12]. To observe the highest orders in Li^+, higher pumping intensities will be required. Sarukura *et al.* [6], for example, observed higher-order harmonics in rare gases with higher ionization potentials; the 25th in Ne and 23rd in He compared with the 15th in Ar. They obtained this result in the intensity region from 2×10^{17} to 4×10^{17} W/cm^2, in which the intensity dependences of the seventh and thirteenth harmonics looked fully saturated.

One of the essential questions of the high-order harmonic generation is whether the conversion efficiency is governed by the single-atom response or by the collective response. The calculations by Kulander and Shore [13,14] and Eberly, Su, and Javanainen [15,16] showed that a spectrum emitted from a single atom under strong laser fields agreed well with experimental results. On the other hand, Lompré *et al.* [7] explained the major properties of high-order harmonic generation by phase matching, considering the many-atom response that depends on confocal parameters and the ionization of a medium. It is not yet clear whether the harmonic efficiency is governed by the phase matching or the nonlinear susceptibility of a single atom. However, it is important to estimate the phase mismatch due to free electrons to discuss the harmonic efficiencies in rare gases and rare-gas-like ions. The positive phase mismatch due to free electrons is expressed by the equation [17] $\Delta k_{fe}=(q^2-1)\omega_p^2/2qc\omega$, in which $\omega_p^2=4\pi N_e e^2/m$ is the plasma frequency, q the order of harmonics, c the speed of light, ω the angular frequency at 248 nm, e and m the charge and the mass of an electron, respectively, and N_e the electron density. The phase mismatch is directly proportional to the number density of electrons. Since Δk_{fe} is inversely proportional to the laser frequency, the use of short wavelength lasers is favored to reduce the effect of phase mismatch. Values of Δk_{fe} for a completely singly ionized medium with an electron density of 10^{17} cm^{-3} are listed in Table I. In the present experiment, the confocal parameter was approximately 2 mm, which was the same as the length of

TABLE I. Phase mismatch Δk_{fe} (cm^{-1}) due to free electrons with number density of $N=10^{17}$ cm^{-3}.

Harmonic order					
3	5	7	9	11	13
1.9	3.4	4.8	6.2	7.6	9.0

the laser-produced plasma. If only free electrons contribute the phase mismatch, the coherence length $(2\pi/\Delta k)$ of the thirteenth harmonic with an electron density of 10^{17} cm^{-3} would be 7 mm. In our separate experiment [18], the electron density in the alkali-metal plasma was estimated to be $4\pm2\times10^{17}$ cm^{-3} from the Inglis-Teller limit [19] in the He-like Li spectrum. Therefore, we concluded that free electrons did not influence the phase matching in our experimental conditions.

In summary, using a subpicosecond KrF excimer laser, we have demonstrated high-order harmonic generation in laser-produced rare-gas-like alkali-metal ions. At a focused laser intensity of 2×10^{15} W/cm^2, the highest harmonic observed was the thirteenth (19.1 nm) in K^+. To determine the limit of the highest order attainable, further increase of the focused intensity will be required.

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[a]Permanent address: Department of Electrical Engineering, Faculty of Science and Technology, Keio University, 3-14-1 Hiyoshi, Kohoku-ku, Yokohama 223, Japan.

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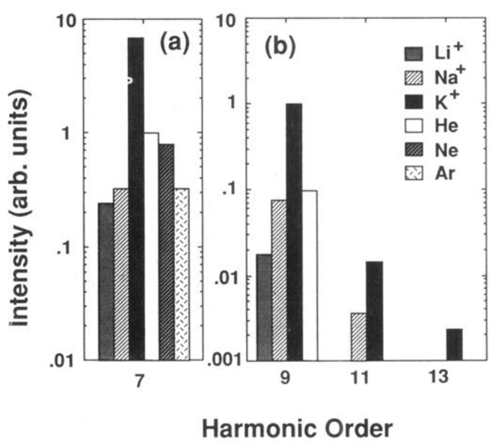

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