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Coherent soft x-ray generation by the harmonics of an ultrahigh-power KrF laser

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Harmonic generation is investigated at an intensity range of 10^{15}–10^{18} W/cm^2 by using a 280-fsec KrF laser. The highest-order harmonic observed with Ne as a nonlinear medium is the 25th (9.9 nm), which gives the shortest-wavelength coherent radiation ever obtained. The inversions or anomalous peaks of harmonic intensity are first found at the 19–21st in Ne, the 17–19th in He, and the 11–13th in Ar. The intensity dependence of the harmonics is measured over a range of 10^{15}–10^{18} W/cm^2. The observed distribution of harmonics is discussed and compared with the previous reports.

Recently there has been rapid progress in the study of multiphoton processes, which involve multiphoton ionization, above-threshold ionization[1] (ATI) and harmonic generation.[2] Of these processes, harmonic generation has attracted interest in the field of atomic physics and for practical use in a soft-x-ray region. Many efforts have been made to obtain the shortest wavelength with high-order harmonics by using the fourth harmonic (266 nm) of an yttrium-aluminum garnet (YAG) laser[3] or the fundamental of a KrF excimer laser (KrF).[4,5] McPherson *et al.* obtained up to the 17th harmonic in Ne by using a 1-psec KrF laser at an intensity range of 10^{15}–10^{16} W/cm^2. They found that there was a plateau in the harmonic distribution above the 11th, and also reported the highest-order harmonics to be the 13th in He, and the 7th in Ar. More recently, Ferray *et al.* clearly observed the plateau in Xe, Kr, and Ar, using a 36-psec Nd-YAG laser (1.06 μm) at an intensity range of $\sim 3\times10^{13}$ W/cm^2 (Ref. 6). They also observed a deviation from the perturbation theory at higher-order harmonics. The highest-order harmonics were the 33rd in Ar, the 29th in Kr, and the 21st in Xe. These results were analyzed using nonperturbative theories.[7–9] Eberly *et al.* clarified the close relationship between harmonic spectra and ATI electron peaks.

In this study, harmonic generation has been investigated using a 280-fsec KrF laser at an intensity range of 10^{15}–10^{18} W/cm^2. The observed highest-order harmonic was the 25th in Ne, which, to our knowledge, is the shortest-wavelength coherent radiation ever observed in a soft x-ray region. Some new anomalous peaks in a harmonic distribution were observed in Ne, He, and Ar. Intensity dependences of harmonics on laser intensity were observed well above 10^{15} W/cm^2. From these results, the observed distribution of harmonics is discussed and compared with the previous reports.

A 10-Hz terawatt KrF laser system using gated gain amplification (GGA) with polarization multiplexing was used in this experiment.[10] An output energy of 410 mJ in 280 fsec was obtained in two beams with different polarizations. The typical amplified spontaneous emission (ASE) energy level of this system was $\sim$10%. In the experiments the horizontally polarized beam was used. The system was operated at $\sim$1 Hz throughout the experiment. The energy in a subpicosecond pulse was measured for each shot, by subtracting the ASE energy. ASE energy was determined using a digital signal analyzer (Tektronics DSA602) for the integration of the temporal profile of the ASE background. The pulse width was measured to be 280 ± 35 fsec, assuming the sech^2 shape. These measurements were taken before the harmonic-generation experiment and employed the three-photon fluorescence method.[11] The spectral width was 0.4 nm, yielding a time and bandwidth product of 1.7 times the transform limit.

The beam was focused by an off-axis parabolic mirror with a 275-mm focal length. The spot size was measured for each shot using a UV video camera with a two-dimensional frame memory, by magnifying the focal image by a factor of 21 with a UV lens (Nikon UV micronikkor $f=105$ mm). The typical spot size for the full-size beam (5×3 cm^2) was 4.5×6.5 μm^2 at the full width at half maximum that corresponded to three times the diffraction limit.[12]

The experimental setup for harmonic generation was essentially the same as those reported previously.[5,6] The equipments consisted of two chambers, one for a gas jet and the other for a spectrograph, which were separated by a 100-μm-wide slit. The chambers were evacuated differentially with two turbo-molecular pumps, obtaining the high vacuum (5×10^{-7} Torr) required for the use of a microchannel plate (MCP).

The gas-jet chamber contained a focusing mirror and a pulsed gas source. An off-axis parabolic mirror with a 275-mm focal length was used to avoid spherical aberration and pulse-front distortion.[13] The confocal parameter (b) for the full-beam size was $\sim$0.5 mm, which was slightly short in relation to the interaction length of less than 1 mm. The pulsed gas jet (Lasertechnics) was operated synchronously with the laser system, and the laser beam was focused at a point a few hundred microns below the nozzle tip. From a typical backing pressure of 10 atm and a 1-mm-diam aperture of the nozzle, the gas concentration at the interaction region was estimated to be $\sim7\times10^{17}$ cm^{-3} in an $\sim$800-μsec-pulse width.[14]

A grazing incidence spectrometer with a flat-field grating coated with gold (Hitachi 1200 l/mm) was used in this experiment.[15] Because the spectral region covered by the spectrograph was from 5 to 40 nm, the lowest-order harmonic observed was the 7th. By the harmonics above the 9th were usually monitored at once because of the limited MCP size, although the 7th harmonic could be covered by translation of the MCP. The two-stage MCP (Hamamatsu photonics F2815-21P) was coated with CsI to enhance the sensitivity in the short-wavelength region. The thin-film filters made of boron or aluminum-silicon alloy (Acton Research) were inserted after the slit to prevent fundamental light scattering, as well as to cover many harmonics in the same sensitivity region by attenuation of lower-order harmonics. The spectrum image on the phosphorous screen of the MCP was observed by an optical-multichannel analyzer (OMA) outside the vacuum. The absolute wavelength was calibrated with the laser-produced plasma of aluminum.

As nonlinear media, He, Ne, and Ar were used. The spectrum of the harmonics using Ne is shown in Fig. 1. The 25th harmonic was observed in Ne, and the 27th harmonic also appeared. However, since the spectrum was overlapped by the background, the 27th harmonic was excluded from the quantitative analysis. The observed highest order in He was the 23rd. In Ar, the spikelike background due to the strong scattering from the laser-produced plasma prevented observation of higher-order harmonics above the 15th, and the spectrum distribution was not as reproducible as that of other spectra.

The relative intensity of the harmonics was calibrated from the spectral responses of the MCP, the diffraction efficiency of the grating,[16] and the transmittance of the boron filter. The spectral responses of the MCP and the transmittance of the filter were determined using the data from the manufacturers. The intensity observed in the OMA is proportional to the harmonic photon number. However, the estimation of the absolute harmonic photon number is difficult in the present system, because of the uncertainty of the collection efficiency of the slit and the relation between the photoelectron number in the MCP and the OMA signal. The shape of the relative intensity was reproducible in He and Ne. In He, the 13th and the 21st appeared slightly higher than the average shot. There were significant changes in shapes when different gases were used (Fig. 2). In each case, the inversions or anomalous peaks were observed. These inversions appeared at the 17–19th in He, the 11–13th and the 19–21st in Ne, and the 11–13th in Ar. The inversion at the 11–13th in Ne was first observed previously by McPherson *et al.*

Intensity dependences of the 7th, 13th, and 19th harmonics on laser intensity in Ne are shown in Fig. 3. The laser intensity was changed by introducing a fixed aperture along with an attenuator. First, the intensity was varied by introducing attenuators into a full-size beam. However, this resulted in a larger amount of scattered data points in the low-intensity region, probably due to the incompleteness of the spatial coherence over a full-size beam. Degrading spatial coherence (deviation from the plane wave) coincides roughly with the deviation from the diffraction-limited spot size. The measured spot sizes at the focal point were almost the diffraction limit for a 10-mm-diam beam, 1.6 times the diffraction limit for a 20- to 25-mm-diam beam, and 3 times for a full-size beam. Then, to observe the dependence in specified intensity regions, as small a beam aperture as possible was selected that was still capable of covering the intensity region. The apertures used are indicated in Fig. 3. Of course, one should be very careful to interpolate these different sets of data, because, strictly speaking, they are based on the different conditions. In the present work, we shall interpret and discuss the experimental results, being aware of this limitation. The 7th harmonic increased steeply and

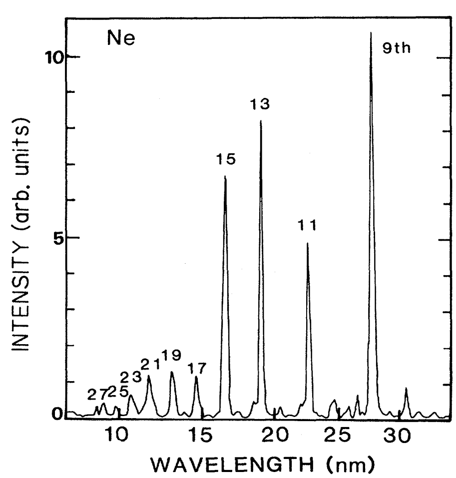

FIG. 1. Harmonic spectrum for Ne. A 100-nm-thick boron filter was used. The laser intensity was 4×10^{17} W/cm^2, at a gas density of $\sim7\times10^{17}$ cm^{-3}.

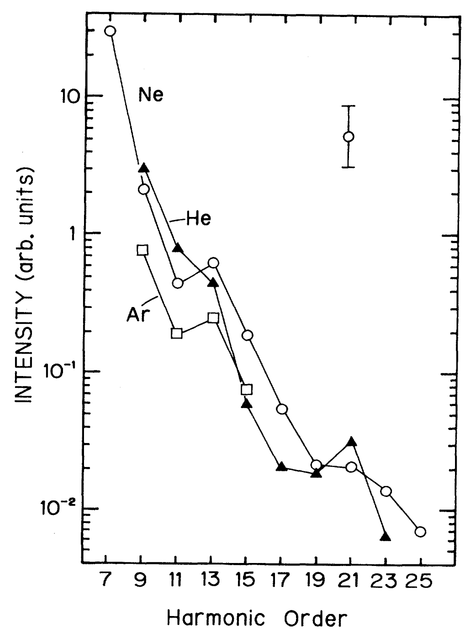

FIG. 2. Relative intensity of the harmonics taken in a single shot, except for the 7th harmonic, the 7th harmonic intensity was extrapolated from a similar shot. Laser intensities were 4×10^{17} W/cm^2 for Ne, 2×10^{17} W/cm^2 for He, and 3×10^{17} W/cm^2 for Ar. Gas densities were $\sim7\times10^{17}$ cm^{-3} for Ne and He, and $\sim2\times10^{17}$ cm^{-3} for Ar.

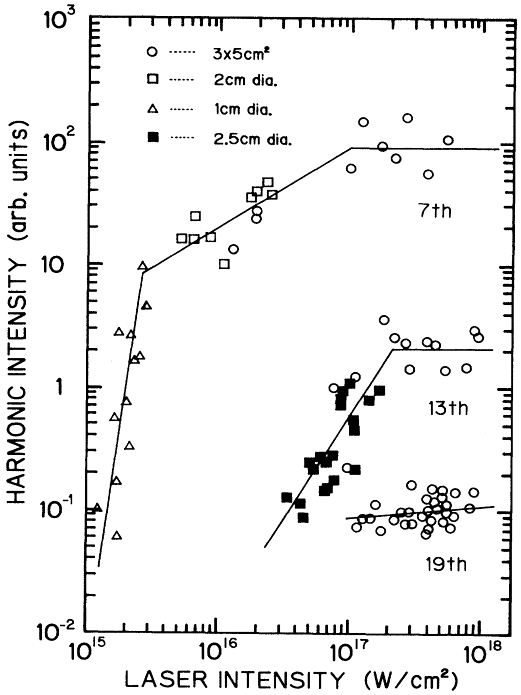

FIG. 3. Dependence of harmonic intensity on laser intensity in Ne. The gas density was $\sim 7\times10^{17}$ cm^{-3}.

saturated at $\sim 2.5\times10^{15}$ W/cm^2. The estimated saturation intensity has some uncertainty because there is a break of data points from a 1-cm beam to a 2-cm beam. The solid line indicated in Fig. 3 has the slope of 7 predicted by the perturbation theory, although the least-squares fit gave 5 ± 3. The slope above the saturation intensity rose slowly and then became flat above $\sim 1\times10^{17}$ W/cm^2. In the 13th, the harmonic intensity saturated at $\sim 1.5\times10^{17}$ W/cm^2, after a relatively gentle slope. The 11th harmonic showed almost the same tendency, although it is not shown in Fig. 3. The signal of the 19th harmonic could not be detected below $\sim 1\times10^{17}$ W/cm^2, which means the 19th harmonic saturated at this laser intensity. In the range of 2.5×10^{15}–10^{17} W/cm^2, the harmonics would saturate by the various effects associated with ionization, including the depletion of Ne atoms and additional phase mismatching due to free electrons. The values of the slopes in this region were 0.7 ± 0.5 for the 7th and 1.9 ± 0.7 for the 13th, if the data were fitted to the straight line. However, to clarify the mechanism of this saturation, the comparison with multiphoton ionization under similar conditions will be necessary. The spatial coherence would also affect in this region. The plateaus above $\sim 10^{17}$ W/cm^2 seem to correspond to degrading spatial coherence. In this region, harmonic intensities depend on spatial coherence of each shot rather than measured laser intensity.

Summarizing the experimental results, in addition to the achievement of the coherent radiation below 10 nm by harmonic generation, we found some new anomalous peaks in a harmonic distribution with He, Ne, and Ar. However, the distribution did not show a clear plateau as previously reported.[5,6] In our experiment the intensity of harmonics decreased by 2 orders of magnitude from the 11th to the 25th harmonic with Ne, while the harmonic intensity differed only within 1 order of magnitude from the 7th to the 29th harmonic of a YAG laser with Ar,[6] and an almost flat distribution was observed from the 11th to the 17th harmonic of a KrF laser with Ne.[5] In the present experiment, the intensity dependence was investigated in a region up to 10^{18} W/cm^2 and the intensity dependence above the 11th harmonic was observed.

Based on these facts, we discuss the harmonic distribution which includes inversions or anomalous peaks. There are many factors to be considered, such as the coincidence between ATI electron peaks and harmonic spectra,[9] the accidental resonances of atomic levels,[5,17] and phase matching.[6]

From the theory,[9] each higher-order harmonic intensity is proportional to the relevant ATI electron peak, and the ionization threshold is shifted by the ponderomotive potential. In Ne, the effective ionization potential at saturation intensity ($\sim 2.5\times10^{15}$ W/cm^2) is calculated to be 36 eV, where the first harmonic above the threshold corresponds to the 9th. On the other hand, the first anomalous peak appeared around the 11th and 13th harmonics.

McPherson *et al.* explained the onset of the plateau in Ne at the 11th harmonic by the inner-shell excitation, where the excitation energy was 45.5 eV.[5] However, this mechanism cannot explain the inversion in He at the 17–19th and the second inversion in Ne at the 19–21st. Perry, Szoke, and Kulander predicted the enhancement of harmonics above the 7th by the atomic-level resonance in He around an intensity region of $\sim 10^{14}$ W/cm^2 at an UV wavelength, from the ATI experiment in He.[17] However, we could observe no significant enhancement at such a low-order harmonic. This would be because of the difference in an effective interaction intensity. In the present experiment, the effective interaction intensity in He would be above that of Ne ($\sim 2.5\times10^{15}$ W/cm^2). Therefore, the present condition is far from the requirement for the predicted resonance.

Phase matching should also be considered. Under the assumption of a perturbative-intensity power law and soft focusing, the geometrical phase mismatch $2(q-1)/b$ (q: the harmonic order) for a focused Gaussian beam is estimated to be 1000 cm^{-1} at the 25th harmonic in our experimental condition,[6] while the phase mismatch in a completely ionized medium is estimated to be 120 cm^{-1}. To examine the contribution of phase matching to the shape of harmonic distribution, further experiments with different confocal parameters will be required.[18,19]

Within the framework of this experiment, the relative importance of these factors to the harmonic distribution cannot be determined.

In conclusion, we have generated the 25th harmonic of a KrF laser, which gives the shortest-wavelength (9.9-nm) coherent radiation that has been achieved to date in a soft x-ray region. Furthermore, many anomalous peaks in the harmonic spectra in He, Ne, and Ar were found. The intensity dependence of harmonics on laser intensity was observed in a range of 10^{15}–10^{18} W/cm^2. Finally, the observed distribution of harmonics is discussed and compared with the previous reports.

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