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High-order harmonic generation by ultrashort KrF and Ti:sapphire lasers

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Clear plateaus, up to the 71st harmonic in Ne and the 69th harmonic in He, were observed in a Ti:sapphire laser, while a gradual decrease to the 25th harmonic in Ne and the 23rd harmonic in He were observed in a KrF laser. High intensity, along with a shorter pulse, induced self-phase-modulation and/or beam breakup in gas media and resulted in the serious degradation or complete disappearance of higher-order harmonics. In the Ti:sapphire laser, the harmonic distribution did not depend on the pulse width of from 120 to 730 fs at a fixed peak intensity.

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Rapid progress in high-power laser technology has made it possible to expose an atom in the electric field of an intense light with a field strength greater than an atomic unit. Growing interest in multiphoton processes under such conditions has generated many investigations. One of the most interesting phenomena, harmonic generation, has been studied using various high-power lasers [1–5]. The most striking observation was a "plateau" followed by a sharp cutoff in the harmonic distribution curves. Recently, Sarukura *et al.* observed the 25th harmonic (9.9 nm) of a KrF laser in Ne [3]. Balcou *et al.* detected the 53rd harmonic (20 nm) of a picosecond Nd:glass laser [6]. Crane *et al.* observed the 45th harmonic (12 nm) using a frequency-doubled Nd:glass laser [7]. More recently, Macklin, Kmetec, and Gordon observed the 109th harmonic (7.4 nm) using a 125 fs Ti:sapphire laser [8]. Krause, Schafer, and Kulander pointed out that the maximum harmonic in a plateau is given by $E_{\max}=I_p+3U_p$, where I_p is the ionization potential, and U_p is the ponderomotive potential [9]. Their results suggest that the resonance effect is not very important to plateau behavior. They also pointed out that ions play an important role in the generation of higher-order harmonics in the KrF laser. However, the mechanism of high-order harmonic generation is not yet completely understood. We studied harmonic generation using ultrashort (~100 fs) KrF and Ti:sapphire lasers, focusing on the dependences of harmonic distribution on the laser wavelength, intensity, and pulse width.

In the KrF experiment, the experimental system was essentially the same as that used by Sarukura *et al.* [3], except for the pulse width. The pulse width of 100 fs was obtained using a prism pulse compressor [10]. Figure 1 shows the harmonic distributions for (a) 410-fs and (b) 100-fs pulses at a peak intensity of 6.1×10^{17} W/cm^2. The pulse width was changed with and without a pulse compressor. Thus, strictly speaking, the nature of light is different in the two pulses. The time-bandwidth products were larger than the transform limit by factors of 1.3 in 100 fs and 5.4 in 410 fs with the same spectral width of 0.86 nm. An absolute photon number was determined at the seventh harmonic by the total charge passing through a high-speed microchannel place (MCP) detector (Hamamatsu Model No. F3654-21s). The collection efficiency, the spectral response and quantum efficiency of the MCP, the diffraction efficiency of the grating and the transmittance of the filter were all taken into account. The target gas density was measured to be 1.9×10^{18} cm^{-3} using the four-photon fluorescence method [11]. Using a 100-fs pulse, harmonics above the 11th harmonic vanished completely, although harmonics up to the 21st harmonic were observed when a 410-fs pulse was used. These phenomena were reproducible. The spectra of KrF laser light transmitted through Ne were observed in the peripheral areas of the KrF laser beam. The insets in Fig.1 show the spectra for (a) 410-fs and (b) 100-fs pulses. Spectral broadening, with a red shift due to self-phase-modulation (SPM), was observed in the 100-fs pulse. The frequency sweep [$\Delta\omega$] due to SPM is proportional to

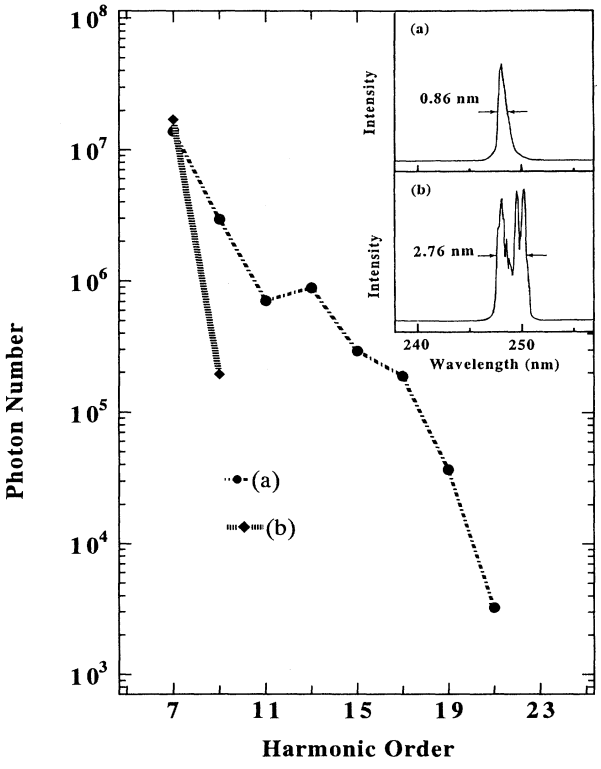

FIG. 1. Harmonic distributions in KrF with pulse widths of 410 fs (a), and 100 fs (b). Insets are the spectra for a KrF laser light passing through Ne. The peak laser intensity was 6.1×10^{17} W/cm^2 and the gas density was 1.9×10^{18} cm^{-3}.

$[-dI(t)/dt]$, where $I(t)$ is a temporal laser intensity. Thus inset (b) shows that SPM has taken place at the rising part of the laser pulse. SPM in Ne induces an increasing phase mismatch for higher-order harmonics. Before the experiment, we expected to obtain an extended harmonic order with shorter pulses, because shorter pulses give a higher effective intensity and are closer to the transform-limited pulse. However, the frequency sweep of the 410-fs pulse degraded harmonic generation very little in this experiment.

Obviously, a long-wavelength laser is better for producing a high SPM threshold and then the plateau behavior. Therefore we developed a 0.3-TW, 120-fs Ti:sapphire laser to investigate harmonic generation. The details of that laser system will be presented in another paper.

A Ti:sapphire laser pulse was focused into Ne with a 150-mm focal-length lens. The absolute photon number of the harmonics was estimated by comparing the results with those from the KrF experiment. The spot size at the focus was measured to be 42 μm in diameter [full width at half maximum (FWHM) in intensity]; the confocal parameter b was thus estimated to be 11 mm. The optimized focal position was very close to the top of the nozzle tip. Figure 2 shows the harmonic distributions for 120-fs pulse width peak intensities of 8.5×10^{15} and 3.1×10^{15} W/cm^2. The peak intensity was determined by $I_{\text{peak}}-0.61E_{\text{pulse}}/\pi r_0^2\tau_p$, where E_{pulse} is the laser energy, r_0 is the radius of the spot at half intensity, and τ_p is the pulse width (FWHM). The pulse width was estimated by assuming sech^2-shaped pulses. Degradation of harmonic distribution was observed at a higher intensity with a fixed pulse width (120 fs). Again, SPM and/or beam breakup suppressed higher-order harmonics, so successive experiments were performed under conditions where SPM was not significant. Figure 3 shows the dependencies of harmonic distributions on laser intensity and pulse width. All the curves show the plateaus and sharp cutoffs. The plateaus extended to the 47th, the 55th, and the 63rd harmonics for laser intensities of 6.1×10^{14}, 1.6×10^{15}, and 3.1×10^{15} W/cm^2, respectively. The saturation intensity of ionization for neutral Ne gas was measured to be 8.8×10^{14} W/cm^2. Thus the effective intensity, where neutral atoms interact effectively, is around this value. However, that effective intensity is affected by the peak laser intensity and pulse width. Even a slight increase in either would extend plateaus to higher orders. The curves in Fig. 3 show that the intensity of effective interaction with neutral atoms increased as the laser intensity increased, resulting in high-order harmonics. This agrees well with the findings of Crane *et al.* [7]. We also compared the harmonic distributions for pulse widths of 120 and 730 fs at a fixed peak intensity of 6.1×10^{14} W/cm^2, as shown in Fig. 3. A pulse width of 730 fs was obtained by adjusting the grating separation in the pulse compressor. The time-bandwidth products were larger than the transform limit by factors of 2.0 for the 120-fs pulse and 12 for 730 fs with the same spectral width of 11 nm. No significant difference was observed between the two curves. This is contrary to our expectation, as in the KrF experiment. The rather large difference in frequency sweep between the two pulses had little influence on harmonic generation. At a high intensity level as in this experiment, factors other than the effective intensity and the frequency sweep, for example

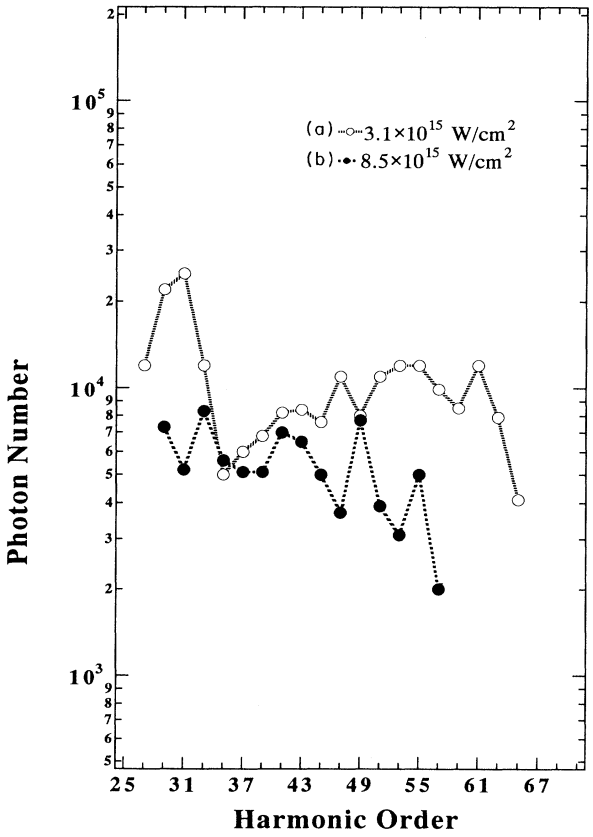

FIG. 2. Harmonic distributions in Ti:sapphire with a pulse width of 120 fs. The laser intensities were 3.1×10^{15} W/cm^2 for (a), and 8.5×10^{15} W/cm^2 for (b). The Ne gas density was 1.9×10^{18} cm^{-3}.

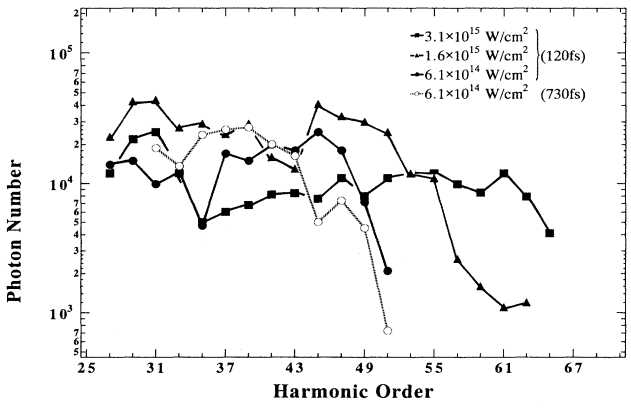

FIG. 3. Harmonic distributions in Ti:sapphire. The solid curves were obtained using 120-fs pulses. The laser intensities were 6.1×10^{14} W/cm^2 for the solid circles, 1.6×10^{15} W/cm^2 for the solid triangles, and 3.1×10^{15} W/cm^2 for the solid squares. The dotted curve connecting the open circles was obtained using a 730-fs pulse, where the laser intensity was 6.1×10^{14} W/cm^2. The gas density was 1.9×10^{18} cm^{-3}.

SPM, might determine the harmonic distribution. In the experiment Balcou *et al.* [6] reported, apparent pulse width dependence was observed in Xe using 1.06-μm pulses. The highest-order harmonic was the 29th for 1.2 ps at 2×10^{14} W/cm^2, and the 21st for 36 ps at 4×10^{13} W/cm^2. However, since they changed the intensity along with the pulse width, it was possible to differentiate the dependence on pulse width from the dependence on laser intensity.

Harmonic distribution was also observed for three different values of confocal parameter b: 4.8, 11, and 63 mm, at a fixed laser intensity. Unlike the Balcou *et al.* [6] case, the harmonic intensity was nonproportional to b^3. But this can be explained simply by the different experimental condition. The present experiments were usually performed above the saturation intensity of ionization, where the phase mismatch in higher-order harmonics would be determined by free electrons and not by the confocal parameter.

Harmonic distributions for the KrF and Ti:sapphire lasers are plotted in Fig. 4. The photon number for KrF was much higher than that for the Ti:sapphire laser at lower harmonic orders. On the other hand, the clear plateaus in the Ti:sapphire laser extended up to higher orders, resulting in the same photon number as for the KrF laser at above 100 eV. For the Ti:sapphire laser, the harmonic distribution with He was lower than that with Ne at all harmonic orders. Usually, the 100-μm-wide slit of the spectrometer was placed 185 mm from the pulsed gas jet. But in He, harmonics were observed by setting the slit position at the pulse gas jet to improve the collection efficiency. Also, in Ne, the maximum harmonic order, the 71st, was observed with the 500-μm-wide slit although it is not shown in Fig. 4. The maximum harmonic orders observed were the 17th harmonic in Xe, the 29th in Kr, and the 33rd in Ar, although the harmonic distributions in Xe and Kr are not shown in Fig. 4. If we express the maximum harmonic orders by $E_{max}=I_p+kU_p$, the values of k were 0.92 for He, 1.7 for Ne, 1.47 for Ar, 1.8 for Kr, and 1.2 for Xe. The measured saturation intensities of ionization were 1.5×10^{15} W/cm^2 in He, 8.8×10^{14} W/cm^2 in Ne, 4.4×10^{14} W/cm^2 in Ar, 2.9×10^{14} W/cm^2 in Kr, and 2.0×10^{14} W/cm^2 in Xe. In He, Xu, Tang, and Lambropoulos explained clearly the harmonic distribution of the KrF laser in Fig. 4 by the contribution of He ion [12].

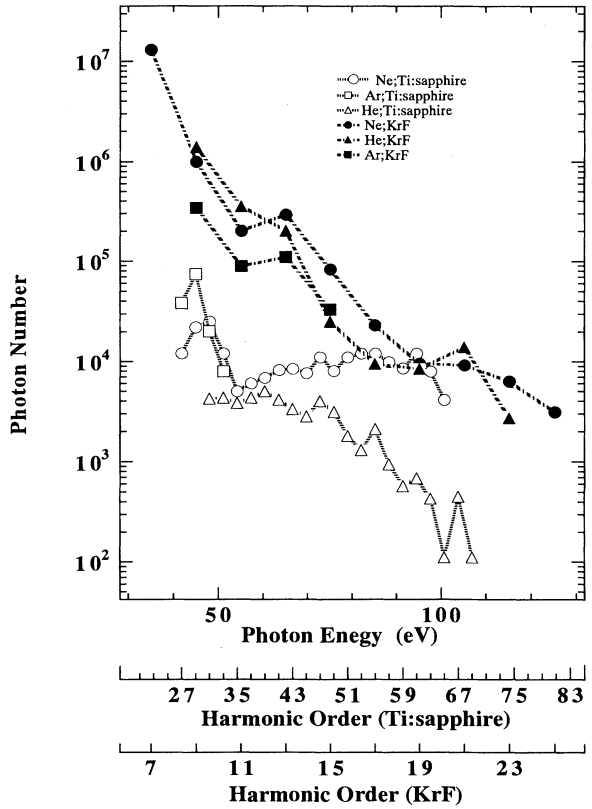

FIG. 4. Harmonic distributions for various target gases (He, triangles; Ne, circles; and Ar, squares) in a 280 fs KrF laser and a 120 fs Ti:sapphire laser. The KrF data, except for the absolute photon number, were taken from Sarukura *et al.* [3]. In the Ti:sapphire laser, the peak intensities were 3.2×10^{15} W/cm^2 for He, 3.1×10^{15} W/cm^2 for Ne, and 3.3×10^{15} W/cm^2 for Ar. The gas densities were 1.9×10^{18} cm^{-3} for Ne and He, and 7.9×10^{17} cm^{-3} for Ar.

We compared our results with some recent experiments. Crane *et al.* investigated the harmonic generation in He at an intensity level well above the saturation intensity using a frequency-doubled Nd:glass laser (527 nm) with a pulse width of 650 fs. Our results for intensity dependence at intensity levels above saturation are in good agreement with theirs. However, the pulse width dependence we observed contradicts their predictions. When using a pulse width of 100 fs at a high intensity, factors other than the saturation intensity, SPM and beam breakup for example, would become important.

Balcou *et al.* summarized the harmonic generation for a wide range of laser wavelengths, pulse widths and intensities. They investigated mainly at an intensity level below or around the saturation intensity where the contribution of ions and free electrons is less important. A pulse width range of 1.2 to 36 ps is one order of magnitude larger than in our experiments.

Macklin, Kmetec, and Gordon reported a 109th harmonic using a Ti:sapphire laser. But, in our experiments the highest order of harmonics observed using a Ti:sapphire laser was the 71st in Ne and the 69th in He. We usually performed our experiments with high laser intensity and with a high gas density to obtain a large harmonic signal. Under these conditions, the phase mismatch became larger for each higher harmonic order. At the laser intensity and gas density where Macklin, Kmetec, and Gordon observed a 109th harmonic, the harmonic signals were below the detection limit of our system. The resolution of our spectrometer was not sufficient to observe such a high order.

In summary, clear plateaus up to the 71st harmonic in Ne and the 69th harmonic in He were observed in a Ti:sapphire laser, while a gradual decrease, to the 25th harmonic in Ne and the 23rd harmonic in He, was observed in a KrF laser. Harmonic distribution dependence on laser intensity, pulse width, and confocal parameter were observed in the Ti:sapphire laser. The plateaus ex-

tended to higher orders as the peak intensity increased. But, intensity above a certain threshold, along with a shorter pulse, induced SPM and/or beam breakup in gas media, resulting in a degraded harmonic distribution at higher orders. In the KrF laser, higher-order harmonics disappeared completely above the 11th with 100-fs pulses. Below the SPM limit, harmonic distribution in the Ti:sapphire laser did not depend on the pulse width, when the intensity was fixed, despite a large frequency sweep between the two pulses 120 and 730 fs.

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