Flat-top phase-matched high-order harmonics in gas-filled cell

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Phase-matched high-order harmonic generation in Ar gas-filled cell is investigated experimentally. We obtain phase-matched 27th order harmonic driven by a commercially available solid-state femtosecond laser system at 0.55 mJ/pulse energy level and 1-kHz repetition rate. Moreover, we find that the spatial distribution of intensity of high-order harmonics is flat-top profile other than a Gaussian one under the condition of optimized conversion efficiency in the static gas cell.

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High-order harmonics are generated by coherent interaction of an intense laser and atoms or molecules[1]. With the development of the intense ultrashort pulsed laser, the research of high-order harmonic generation has reached the water window in spectral region[2] and subfemtosecond in time domain[3]. Especially, the generation and application of subfemtosecond pulse led the study of high-order harmonic generation into a completely new world[4,5]. It has made the study of ultrafast science from the femtochemistry to the attophysics. Presently, high-order harmonics can be easily generated on a tabletop laser system. Because of their high spatial and temporal coherence and low divergence, high-order harmonics can be used in the fields such as atomic and molecular spectroscopy and nonlinear optics as an excellent extreme ultraviolet (EUV) or soft X-ray source, and can be used in lithography and holography[6].

However, because of the phase mismatching, the conversion efficiency of high-order harmonic generation is still very low. The main reason of phase mismatching is the generation of free electrons ionized from atoms by the high-intensity focused laser. In the past years, high-order harmonics in different gas species under different gas densities, focus positions, laser intensities, laser profiles, laser chirp conditions, and those in different gas cells or hollow fibers have been studied systematically. The optimal conditions to reach phase-matching were found in both experiments and theories (for a review, see Ref. [1] as an example). Among these experiments, the highest harmonics conversion efficiency is 6.4×10^{-5} for the 15th harmonic generated in xenon gas-filled hollow fiber using a 35-fs, 16-mJ, 810-nm pulse at 10-Hz repetition rate[7].

The experimental setup used in the present work is illustrated in Fig. 1. We used a commercially available all-solid-state femtosecond laser system (Spectra Physics Spitfire 50fs) with 1-kHz repetition rate, 800-nm wavelength, 50-fs (FWHM) pulse duration, and 0.55-mJ pulse energy. The spatial profile of the output laser beam is nearly a Gaussian distribution. Focused by a fused silica lens ($f/34$), the Gaussian laser beam fought its way through the 0.1-mm-thick stainless steel seals of the glass cell which were filled with noble gases. The cell was fixed in a vacuum chamber. A precision valve outside the chamber controlled the gas pressure in the cell. The high-order harmonics were emitted just in the glass cell. As the laser propagated forwards, two 400-nm-thick aluminum foils blocked it, but the highly collimated coherence harmonic radiation propagated through them. A flat-field X-ray spectrograph[8] was used to obtain the spectrum of high-order harmonics in the EUV and soft X-ray ranges. The spectrograph consists of a gold-coated spherical mirror, a cylindrical mirror, a slit, a Hitachi flat-field grating (1200 groove/mm), and a soft X-ray CCD (Princeton Instruments, 1340×400 imaging array PI-SX: 400). The harmonic spectra is spatially resolved in horizontal axis and spectrally resolved in vertical axis.

The length and diameter of the cell used in the experiment are 20 and 10 mm, respectively. We found the optimal focus position of the laser is at the rear half of the cell. Under this condition, the laser-gas interaction length is less than 10 mm. Moreover, the size of the focus spot is approximately 40 μm under our experimental conditions. Because the holes of the cell, drilled by the focused intense laser beam, are about 100 μm in diameter, the noble gas in the cell can be considered static during the experiment.

In the experiment, the intensity of the driving laser on the focus spot was about 2×10^{14} W/cm^2. The signal from 21st to 33rd order harmonics were obtained in Ar gas, and a typical spectrum of high-order harmonics in Ar

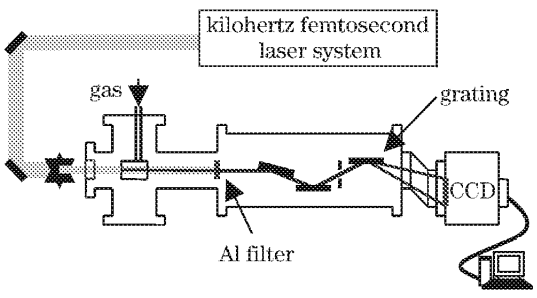

Fig. 1. Experimental setup for high-order harmonic generation in noble gas-filled cell.

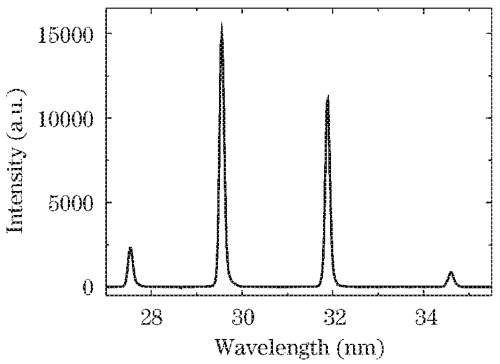

Fig. 2. Typical experimental spectrum of high-order harmonics in 44-Torr Ar gas (1 Torr = 133 Pa), harmonics from 23rd to 29th can be seen from right to left.

is shown in Fig. 2. The highest conversion efficiency of 27th order harmonic was about 1×10^{-7} experimentally in the optimal phase-matching condition of low driving laser energy.

We observed that the source sizes (full-width at half-maximum (FWHW) measured in the CCD plane) of high-order harmonics were enhanced under phase-matching conditions, which was quite different from the experimental results of hollow fibers[7] and loosely focused ($f/71$) laser driven static cells[9].

The source size and the intensity for different order harmonics in different gas density conditions are presented in Fig. 3. In the experiment, the measurement was one-dimensional (1D) spatially resolved. The harmonic source was not sharply imaged in the CCD plane of the spectrograph. However, it would not influence the interpretation in this paper. The source size was confirmed by the spatial distribution in the CCD plane. For the phase-matched 27th- and 29th-order harmonics, the source sizes were augmented about 30% and 20% respectively under their phase-matching conditions.

In the experiment, the spatial distribution in the CCD plane is directly related to the spatial distribution of the intensity of high-order harmonic generated in the interaction region of the driving laser with Ar gas. Therefore, the variations of the source size and the spatial distribution reflect the change in the spatial distribution of high-order harmonic generation. The distributions of high-order harmonics mainly rely on the distributions of the driving laser intensity and the free electron density. The distribution of the laser intensity was nearly a Gauss distribution in the experiment, and the distribution of the free electron density was straight determined by that of the driving laser intensity. Under ordinary conditions, Ref. [10] as an example, the distribution of high-order harmonic in the CCD plane was nearly a Gaussian distribution, like the intensity distribution of the driving laser.

However, we observed that the spatial distribution in the CCD plane was not always a Gaussian distribution under the phase-matching condition in the experiment. In Fig. 4, it is shown that the spatial distribution of the intensity of phase-matched 27th order harmonic generation in 55.0-Torr Ar gas is truncate, which is similar to the result of the saturated case. In fact, the counts in the CCD plane were not saturated, and the high-order harmonic generation was not saturated under the intensity of the driving laser. Furthermore, the spatial distribution of the intensity of 27th order harmonic generation in 26.0-Torr gas cell without phase-matching is quite different, also shown in Fig. 4.

Because the generation of high-order harmonics and the phase-matching conditions are sensitively related to

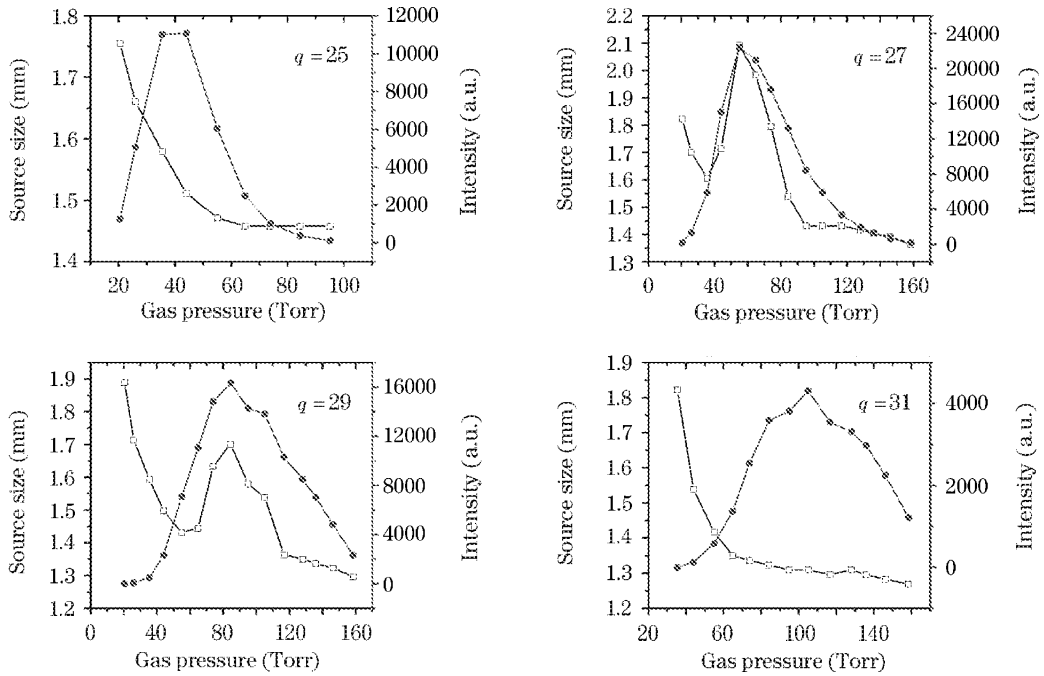

Fig. 3. The source size (lines with hollow squares) recorded in the CCD plane and the intensity (lines with filled circles) for different order harmonics in different gas pressure conditions. q represents the harmonic order.

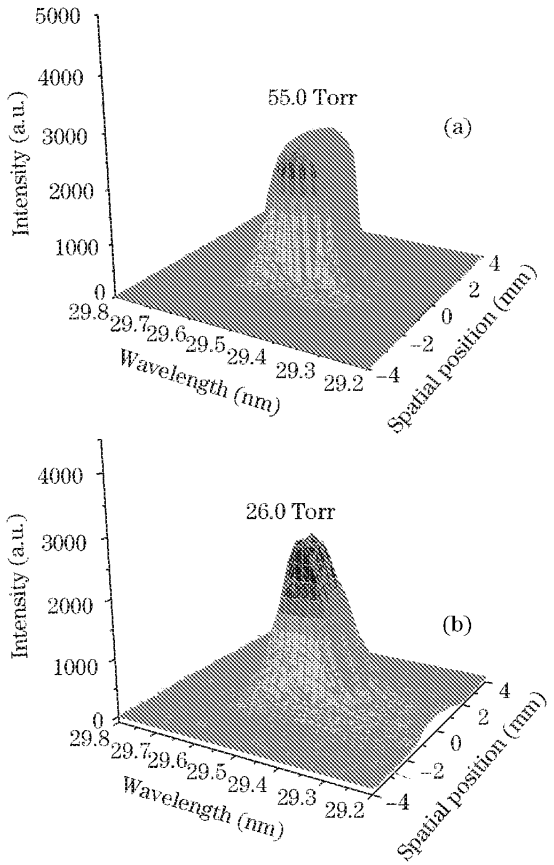

Fig. 4. 1D spatial resolution and spectral resolution profile of the 27th order harmonic. (a) The well phase-matched case when the gas pressure is 55.0 Torr; (b) non-phase-matched case when the gas pressure is 26.0 Torr.

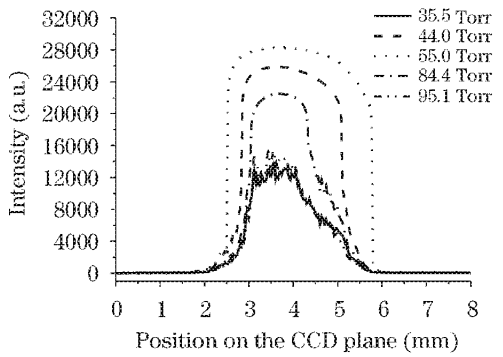

Fig. 5. The spatial distribution of the intensity of the 27th order harmonic under different Ar gas pressures.

the driving laser intensity[11,12], the spatial distribution of high-order harmonics sensitively relies on the distribution of the laser intensity in the interaction region. Because of the Gaussian distribution of the laser intensity around the focus, the interaction length of off-axis is shorter than that of on-axis, and then the gas density condition of optimizing phase-matching is different for the off-axis one from that for the on-axis one. For the shorter interaction length case, one needs shorter absorption length to achieve the optimizing phase-matching[13]. Therefore, the optimized gas density for the off-axis is higher than that of the on-axis one, which is presented in the results of the experiment. As the gas density increases, the on-axis emission achieves optimal phase-matching before the off-axis one, and there is an optimized gas density for the highest conversion efficiency. As a result, the spatial distribution of high-order harmonics will not be a Gaussian distribution, but the distribution shown in Fig. 4.

The spatial distributions of the intensities of phase-matched 27th order harmonic generation in different gas densities are presented in Fig. 5, which shows that the enhancement of the source size of high-order harmonics under phase-matching condition is mainly related to the intensity increase in the off-axis interaction region.

Moreover, in Figs. 4 and 5, it is also shown that the spatial contours of phase-matched harmonics are quite smooth and those of harmonics without phase-matching are fluctuant. We contribute the difference to the influence of the variation of dipole phases of high-order harmonics from different spatial positions in the interaction region. Without phase-matching, the dipole phases of high-order harmonics from different positions are quite different and will not coincide with each other after the propagation in Ar gas. The superposition of the dipoles with different phases will make the spatial contour very complex. Therefore, the spatial distributions of the phase-mismatched high-order harmonics will be fluctuant[11]. In contrast, for the phase-matched high-order harmonics, only the dipoles with the distinct phase can be enhanced by the superposition, and then the high-order harmonics with the same phase can be enhanced by the coherent superposition through the propagation in Ar gas, so the spatial distribution will be smooth. Furthermore, the phase-matching condition is sensitive to the laser intensity, so the spatial distribution has sharp edge, as shown in Fig. 5.

In addition, in Fig. 3, the enhancement of source size is not observed for the 25th order harmonic. This is because that the absorption edge of Ar gas is around the wavelength of this harmonic. Because of the ionization by driving laser, the absorption in the center is less than that in the outer annulus. So the increasing intensity in the outer annulus is stronger absorbed by Ar atoms than that in the center. On the other hand, for the higher order harmonics, the influence of absorption difference is not obvious. For the 31st order harmonic, see also in Fig. 3, there is no enhancement in the size because this harmonic is not well phase-matched.

Furthermore, as an excellent coherent and low divergence source, the high-order harmonic radiation can be used in the EUV or soft X-ray lithography. With the smooth, flat-top, and sharp-edge profile output, the phase-matched high-order harmonics generated from a commercially available compact laser system may be used in the EUV or soft X-ray lithography without further beam shaping.

In conclusion, we obtained phase-matched high-order harmonic generation in static Ar gas-filled cell driven by a low energy (0.55 mJ/pulse) femtosecond laser pulse. Moreover, we found that the source size and the spatial distribution of the intensity of high-order harmonics are quite different under the phase-matching condition from those of the phase-mismatching case, which is mainly related to the spatial distribution of the intensity of driving

laser, and leads to the flat-top output of high-order harmonics when the conversion efficiency is optimized.

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