Nonlinear multiphoton processes with soft x-ray photons and its application to autocorrelation measurement

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ABSTRACT

We have observed multi-photon ionization processes such as two-photon double ionization and above threshold ionization in He at 42 eV using intense soft X-ray pulses produced by high-order harmonics. Using the two-photon double ionization, the pulse width of the 27th (42 eV) harmonic was measured by an autocorrelation technique, and found it to be 8 ns. The high-intensity soft X-ray radiation achieved by phase-matched high-order harmonics enables the investigation of these nonlinear optical processes, which were beyond the reach of conventional light sources.

Keywords: Nonlinear optics, Soft x-ray, Multiphoton process, Ultrashort Pulse

1. INTRODUCTION

The observation of nonlinear optical processes in the soft X-ray region has been a very attractive and challenging area of research in quantum electronics since the first observation of second-harmonic generation and two-photon excitation in the visible range in 1961 [1]. No observation of nonlinear optical process by soft X-ray photons, however, had been reported because of a lack of intense short wavelength light sources. This situation has changed recently due to the advent of an intense coherent soft X-ray source based on high-order harmonics. We have produced the highest peak photon flux (~10^{26} photons/mm^2/mrad^2/s) in the soft X-ray region (the 27th harmonic of Ti:sapphire laser; a wavelength of 29.6 nm and a photon energy of 41.8 eV) by high order harmonics [2]. When such a harmonic pulse was focused with an off-axis parabolic multilayer mirror, the focused intensity attained 1 x 10^{14} W/cm^2 [3], which is considered to be sufficient for inducing nonlinear optical phenomena in the soft X-ray region [4,5].

In this paper, we report on the generation of strong optical filed in the soft x-ray region by high harmonics and its application to the study of nonlinear multiphoton ionization processes. Two-photon double ionization and above threshold ionization were observed in He with 42 eV photons [6]. Using the two-photon double ionization, the pulse width of the 27th (42 eV) harmonic was measured by an autocorrelation technique in the soft x-ray region. The energy of 42 eV is the maximum energy of the photons of which duration is measured by the autocorrelation technique [7]. Intense soft X-ray photons at 41.8 eV generated by phase-matched high harmonics enable the observation of these nonlinear optical processes.

2. HIGH-POWER, HIGH-ORDER HARMONIC GENERATION

We have already reported the investigation of the energy scaling of high-order harmonics under the phase-matched condition using a long interaction length and a loosely focused pumping geometry. Figure 1 shows the experimental setup for high-order harmonic generation using a long focusing pumping geometry. Details are described elsewhere [1, 8]. Experiments for Xe and Ar gases were carried out with a 10 Hz Ti:sapphire laser producing an output of 200 mJ with a pulse width of 35 fs. The wavelength was centered at 800 nm. The pump pulse was loosely focused with a fused silica lens, and delivered into the target chamber through a CaF_2 window. We set the focus at the entrance pinhole of the interaction cell. The interaction cell had two pinholes on each end surface of the bellow arms. These pinholes isolated the vacuum and gas-filled regions. The interaction length was variable from 0 to 150 mm in the interaction cell. Rare gases were statically filled in the interaction cell. With this experimental setup, we have generated peak powers of 130 MW at 62.3 nm in Xe [8] and 10 MW at 29.6 nm in Ar [1], assuming the same pulse width as the pump pulse of 35 fs.

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Recently, we have also applied this harmonic energy scaling procedure for Ne and successfully generated a peak power of 1 MW at 13 nm [9].

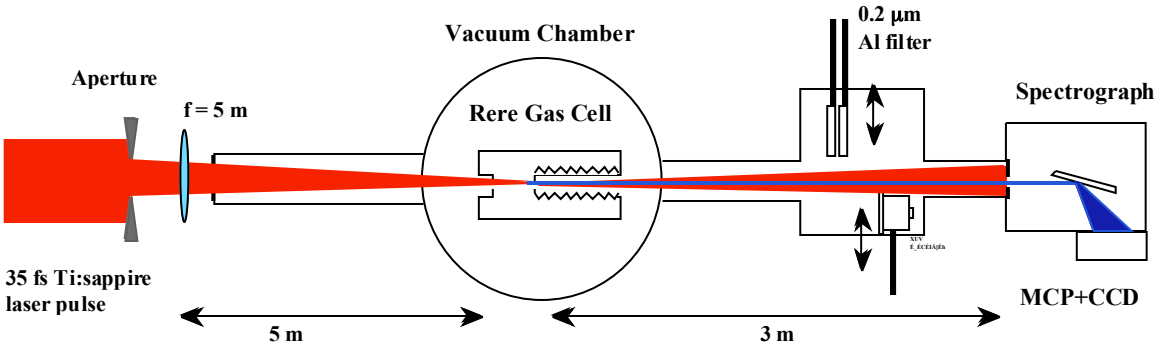

Figure 1: Experimental setup for high-power, high-order harmonic generation

3. FOCUSING PROPERTY OF HIGH-HARMONICS

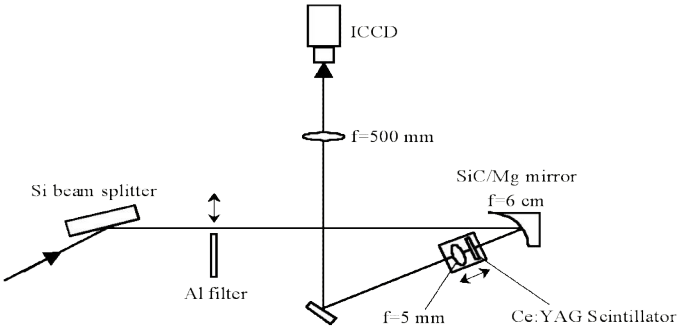

Figure 2: Experimental setup for imaging the focused profiles of high-order harmonics.

Although the advent of high harmonics having excellent beam quality and spatial coherence, together with high output energy, opens up the possibility for focusing XUV beams to micron-order spot size with intensities of greater than 10^{14} W/cm^2, reported intensities have been lower than 10^{12} W/cm^2. Here, we report a technique for focusing an intense coherent soft x-ray beam to a 1 μm spot size with a peak intensity as high as 10^{14} W/cm^2 [3].

The experiment was carried out using a Ti:sapphire laser producing an output of 14 mJ, with a full-width at half maximum (FWHM) pulsewidth of 22 fs at a repetition rate of 10 Hz and a center wavelength of 785 nm. At the exit of the interaction cell, the generated energy was estimated to be 320 nJ with a conversion efficiency of 2.3×10^{-5}. Typical measured energies for the 27th harmonic wave with and without an Al filter were 23 nJ and 160 nJ respectively.

We used an imaging system as shown in Figure 2 to measure the focal spot of the soft x-ray beams. The 27th harmonic wave was selected and focused by an off-axis parabolic mirror (f=6 cm, θ=24º) with a SiC/Mg multilayer coating. The peak-to-valley surface accuracy of the mirror substrate over an area within a 1-inch diameter was $\lambda/8$ at 633 nm corresponding to 2.7λ at 29.6 nm. The RMS surface roughness was 0.7 nm, which is good enough to support the quality of the multilayer coating. The reflectivity of the multilayer coated mirror was 40% at 29.6 nm with a FWHM bandwidth of 2 nm. We estimated that the diameter of the beam on the mirror surface was 2 mm from the MCP spectrograph. In order to visualize the focal spot profile, the 27th harmonic wave was converted to visible light at 550 nm using a 100-μm-thick Ce:YAG scintillator placed at the focal point [10]. A set of image relay optics were transferred the visible light image onto an image-intensified CCD camera. The resolution of the imaging system was 0.5 μm, which was limited by the NA value of the objective lens.

Figure 3 shows the $1/e^2$ beam spot size of the 27th harmonic beam as a function of distance from the off-axis parabolic mirror. The open circles and the filled squares correspond to the horizontal and vertical spot sizes, respectively. There is little difference between the horizontal and vertical diameters, showing that circularity is maintained during focusing. The solid line is the best-fit curve based on the equation, $\omega(z)$ =

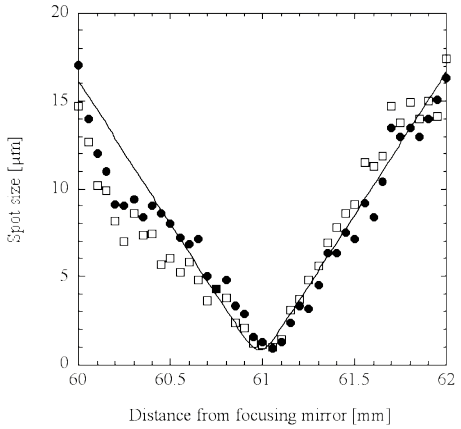

Figure 3: Focused beam spot size of the 27th harmonic wave as a function of distance from the focusing mirror. The open circles and filled squares correspond to the hori-zontal and vertical directions respectively.

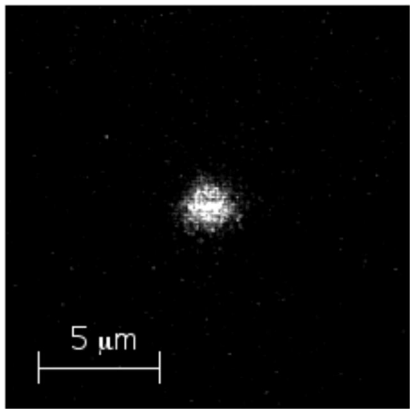

Figure 4: Typical focal spot image for the 27th harmonic beam

$\omega_o[(1+z/R)^2+(M^2\lambda z/\pi\omega_0^2)^2]^{1/2}$, where ω_0 and $\omega(z)$ are the beam radii on the parabolic mirror and at a distance z from the mirror, respectively, R is the wavefront curvature of the soft x-ray beam just after reflection, λ is the wavelength and M^2 is the beam quality parameter. Consequently, the best-fit curve provided a figure of M^2=1.4.

Figure 4 shows a typical focused image taken in a single shot. The dimensions of the spot in the vertical and horizontal directions are 1.0 μm and 0.9 μm respectively, both of which are less than 1.7 times the diffraction-limited value. Because the high-order harmonics have excellent spatial coherence and beam quality, the focusability might be dominated by the performance of the mirror. We have also measured the spot size from an ablation pattern produced on a gold-coated mirror placed at the focal position. The patter was produced by single shot irradiation of the focused 27th harmonic pulse [3]. The diameter of the circular hole is approximately 2 μm, which agrees well with the scintillator experiment.

Finally, we estimated the focused intensity of the 27th harmonic wave from the energy, the spot size, and the pulse width. Assuming that the pulse width of the 27th harmonic wave was the same as the fundamental pulse, the maximum focused intensities with and without the Al filter were 1.4×10^{13} W/cm^2 and 1.0×10^{14} W/cm^2, respectively.

4. MULTIPHOTON IONIZATION OF HE AT 42 EV

In this session, we report on the observation of two-photon ionization processes using 41.8 eV soft x-ray photons including two-photon double ionization [5, 11-14] and above-threshold ionization [6, 7]. The relevant energy diagram of He, He^+, and He^{2+} and ionization pathways using 41,8 eV photons is shown in Figure 5 when two-photon absorption occurs. There are several pathways for the production of doubly ionized He as shown in the figure. It should be noted that nonsequential two-photon absorption is required for the production of doubly ionized helium ions. Therefore, the observation of the doubly charged ion of He provides clear evidence for the nonlinear interaction in soft x-ray spectral region. The ATI process also occurs by two-photon absorption. However, since the final charge state of this process is single ionized one, it is impossible to detect it separately from the single ionized ions produced by one-photon absorption. Therefore, we used time of flight (TOF) electron energy spectrometer for detection of the ATI process instead of ion TOF mass spectrometer.

In the ion experiment, we measured the mass spectra of ions with a conventional TOF mass spectrometer. Several harmonics around the 27th harmonic generated with a femtosecond pulse of a Ti:sapphire laser (a pulse width of 23 fs, a central wavelength of 800nm, a pulse energy of 20 mJ/pulse and a repetition rate of 10 Hz) were separated from the intense fundamental pulse with beam splitter(s) of Si or SiC [15], then sent to a spherical mirror multilayered coat of SiC/Mg with a radius of curvature of 100 mm in the interaction chamber. The energy of the 27th harmonic was

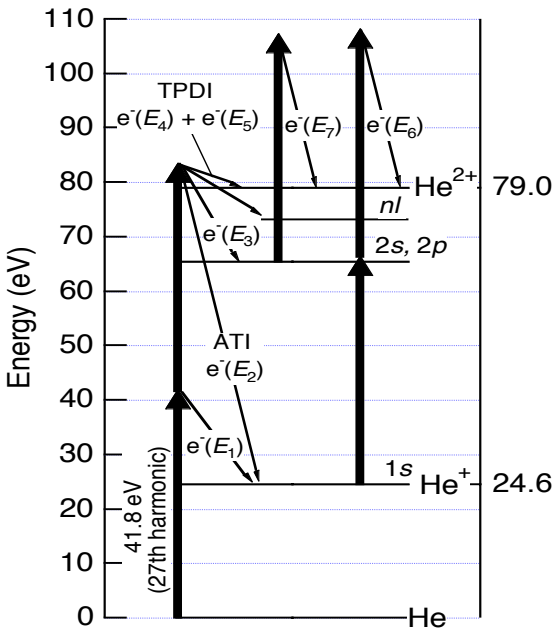

Figure 5: Relevant energy diagram of He, He^+, and He^{2+} and ionization pathways of 41.8 eVphoton.

estimated to be 24 nJ/pulse in the interaction region. The spot size was estimated to be ω_0 = 3.1 μm from a separate experiment [16]. Thus, the intensity of the 27th harmonic was estimated to be $I = 7 \times 10^{12}$ W/cm^2. Here, we regard the pulse width of the 27th harmonic to be the same as that of the fundamental. The confocal parameter was also determined to be 250 μm from the same experiment.

The TOF spectra were recorded on an oscilloscope and/or a computer, then averaged to assign the strong peaks (of the spectra) which originated from the single-photon process and also processed for counting the signals which exceed the threshold voltage to find the weak signals in the spectra. The isotope ^{3}He was used as the sample gas in this experiment to avoid the overlap of signals between $^4He^{2+}$ and H_2^+, the latter of which originated from residual water in the vacuum chamber (the estimated density of the sample gas in the interaction region, 3.2×10^{12} cm^{-3}). The number of shots accumulated is 10000.

Figure 6 shows the ion spectrum produced by the interaction between the focused harmonic pulses and ^{3}He. The production of $^3He^+$ results dominantly from the one-photon absorption of ^{3}He. The strongest peak of 1.8 μs can be assigned to $^3He^{2+}$. The signal of $^3He^{2+}$ clearly appears between the H^+ signal and H_2^+ signal. The generation of doubly charged He^{2+} confirms the observation of a nonlinear optical process (two-photon absorption) in the soft X-ray region. The ionization mechanism is clarified by the observation of the dependence of the He^{2+} yield on the 27th harmonic intensity because the two-photon double ionization process absorbs two net photons and other sequential ionization processes absorb three net photons. In order to simultaneously measure the relative intensities of the 27th-harmonic pulse and the corresponding yields of doubly charged helium ions at those intensities, we utilize the yields of singly charged helium ions as indicators of the intensities. We can assume that the yield of the singly charged ions is proportional to the intensity of the 27th harmonic, because there is no saturation in their production. The result of this experiment is shown in Figure 7. The yields of the doubly charged ions in relation to those of the corresponding singly charged ions are well

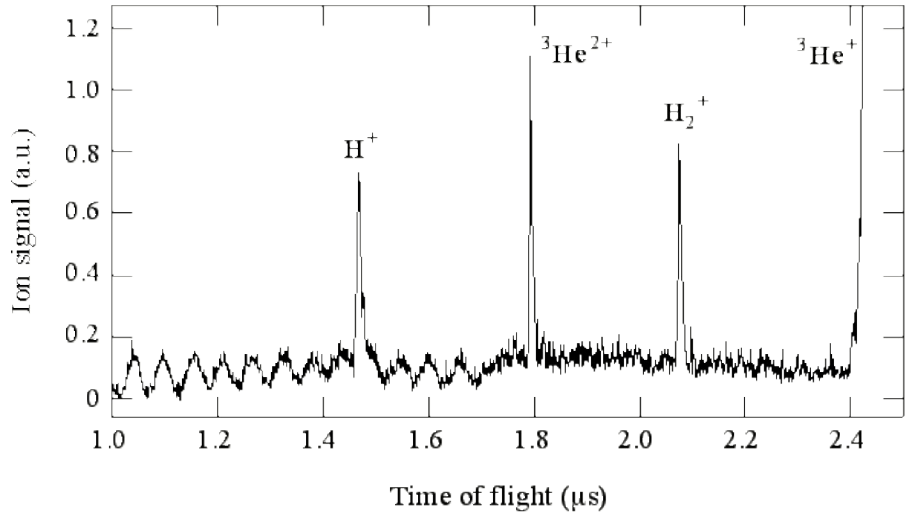

Figure 6: The expanded view of the TOF mass spectra of ^{3}He measured by ion counting.

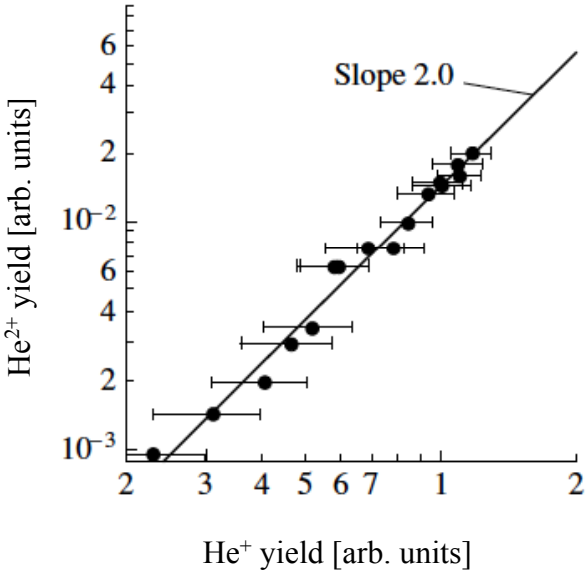

Figure 7: Yield of He^{2+} ions versus He^{+} ions

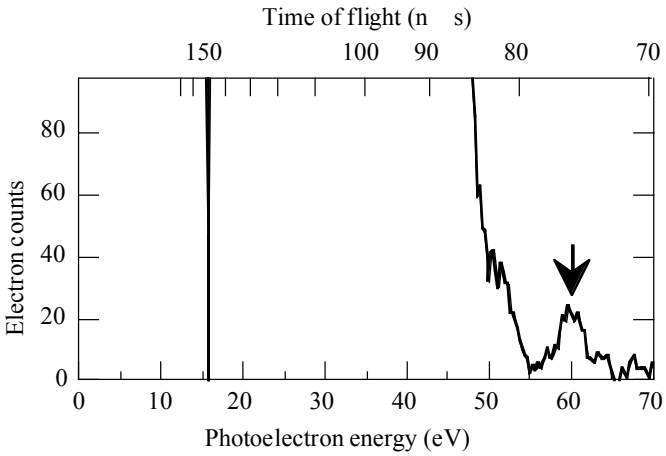

Figure 8: An expanded view of photoelectron spectra of He measured by electron counting. The bottom axis represents photoelectron energy. The top axis corresponds to the TOF of electrons.

located on the fitted line having slop of two. The slope of the intensity dependence is considered to be two when two-photon double ionization is the dominant process. On the other hand, a slope value of three is expected when the sequential processes are dominant. Therefore, we concluded that two-photon double ionization is the most dominant process in this experiment.

We carried out a subsequent experiment to measure the photoelectron spectra so that we could estimate the contribution of two-photon double ionization to the ionization process responsible for the production of doubly charged ions. Unfortunately, a few harmonics other than the 27th were partially reflected by the SiC/Mg mirror, thus disturbing the photoelectron spectra in the energy range of 0 eV to 4.6 eV, in which two-photon double ionization electrons are expected to appear, due to the large signals of the single-photon ionization processes. Accordingly, while we could not observe electrons from the two-photon double ionization process, we succeeded in detecting the above threshold ionization peak. With the counting measurement of the spectrum, we found the remarkable feature of photoelectron at the TOF of 76 ns as shown in figure 8 indicated with an arrow. By converting the TOF to kinetic energy using the assigned peaks, this fast (high-energy) peak was ascribed to photoelectrons with an energy of 59 eV produced via the above threshold ionization of the 27th harmonic. This result is more evidence of the nonlinear optical process.

5. PULSE WIDTH MEASUREMENT BY AUTOCORRELATION

Next we report on the application of the two-photon double ionization in He for the measurement of pulse duration of the 27th harmonic pulse by means of an autocorrelation method. For this purpose, we developed a split beam separator to make a delayed harmonic pulse pair. The pair of pulses is produced by spatially dividing the harmonic beam with two beam separators of SiC that are aligned with each other with a separation of approximately 100 mm (the photograph of the split beam separator is shown in Figure 9(a)). These separators are put in the place of the harmonic separator used in the previous experiment. The measurement principle using this split beam separator is the same as an all-reflective interferometric autocorrelator [17]. Each split pulse was relatively delayed by changing the position of one of two beam separators. Since the autocorrelation measurement requires splitting a pulse into equally two pulses and spatially overlapping the separated two delayed pulses at the focus point, we have checked these condition by following methods. For the beam separation, we have directly confirmed the spatially distribution of the harmonics with a 2D imaging spectrograph as shown in Figure 9(b). For the overlap of the two focused harmonic pulses, we have inspected this by monitoring a fringe pattern of the SHG signal of the fundamental pulse. Then the autocorrelation measurement of the

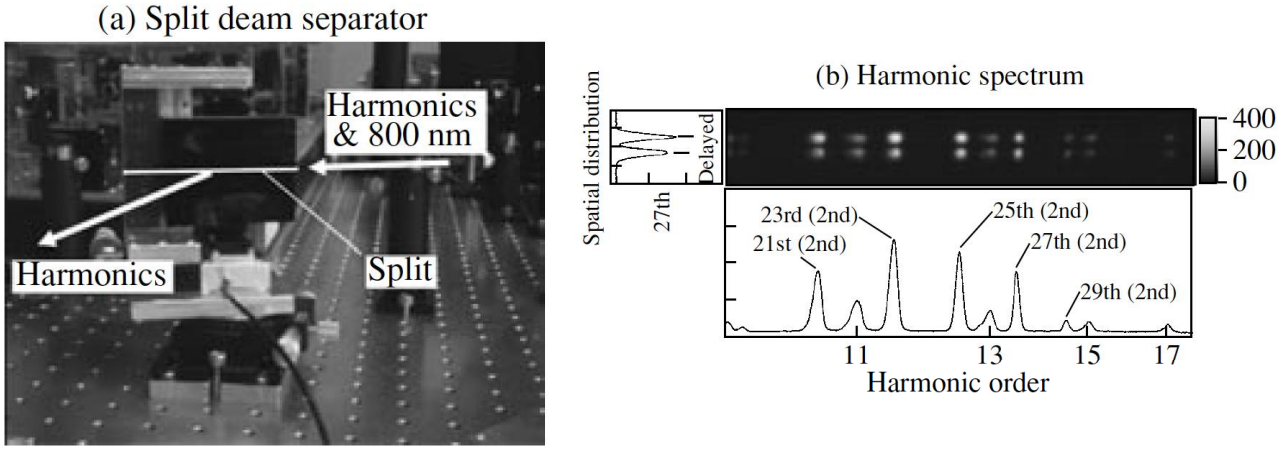

Figure 9: (a) Photograph of the split beam separator. (b) The spectral and spatial (27th) distributions of the harmonics. The double peak in the spatial distribution is relatively delayed.

fundamental pulse by the ionization signal of Ar was used to further confirm spatial and temporal overlap of the two delayed harmonic pulses.

The yield of He^{2+} ions produced by two-photon double ionization as a function of delay time is shown in Figure 10. We can see a typical autocorrelation race in this figure, while the pedestals, which are inherited from those of the fundamental pulse, appear at a delay of ~ 40 fs. As was calculated by Tzallas *et al.* [18], the ratio of the peak height to the background in the autocorrelation signal integrated to a certain volume of the interaction region should be approximately three. The contrast ratio in the autocorrelation trace obtained in our experiment also became three as how in the figure. A pulse width of 8 fs at the full width at half maximum was derived from the fitting curve assuming a Gaussian temporal profile traced as a solid curve in Fig. 10. It is not easy to conclude whether the pulse duration of the 27th-harmonic pulse is reasonable or not with respect to the theory of harmonic generation, because the pulse durations of the harmonics at high orders far from the perturbative regime depend on both the intensity of the fundamental harmonic and the phase-matching conditions, and thus a quantum-mechanical calculation of the induced dipole moments is required, with the propagation effect obeying Maxwell's equations. Practical use of the power law of the intensity dependence of the fundamental pulse, however, is feasible for estimating the pulse duration of the harmonic pulse. Under the condition that the induced dipoles for the high-order harmonics are proportional to the fifth power of the intensity of the fundamental harmonic, which is a reasonably good approximation to the result obtained under quantum theory [19, 20], the durations of the high-order harmonics are roughly estimated to become $1/\sqrt{2\times 5}$ of that of the fundamental pulse. The resultant duration of 7.3 fs agree well with the measured one. The 27th harmonic with a photon energy of 42 eV is the shortest wavelength which is measured by autocorrelation measurement [5].

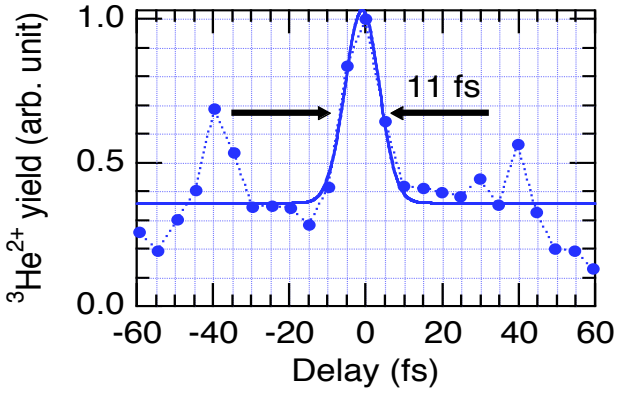

Figure 10: Autocorrelation trace (solid circles with dotted line) of the 27th harmonic pulse. The solid line is obtained by fitting a Gaussian shape to the trace.

6. CONCLUSION

The nonlinear multiphoton processes of helium were induced using intense phase-matched high-order harmonics in the soft X-ray region ($\lambda < 30$ nm). The measurement of the dependence of the He^{2+} yield on the 27th harmonic revealed that the two-photon double ionization dominates He^{2+} production under our experimental conditions. The use of the two-photon double ionization process in He enables us to directly measure the pulse duration of the 27th-harmonic pulse by an autocorrelation technique, and we found it to be 8 fs. The energy of 42 eV is the maximum energy of the photons in the femtosecond pulse whose duration is measured by an autocorrelation technique.

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