Production of Doubly Charged Helium Ions by Two-Photon Absorption of an Intense Sub-10-fs Soft X-Ray Pulse at 42 eV Photon Energy

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We report on the observation of doubly charged helium ions produced by a nonlinear interaction between a helium atom and photons with a photon energy of 42 eV which are generated with the 27th harmonic of a femtosecond pulse from a Ti:sapphire laser. The number of ions is proportional to the square of the intensity of the 27th harmonic pulse, and thus two-photon double ionization should be dominantly induced as compared with other nonlinear processes accompanying sequential ionization via a singly charged ion. This phenomenon is utilized to measure the pulse duration of the 27th harmonic pulse by using an autocorrelation technique, for the first time to our knowledge, and as a result a duration of 8 fs is found.

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The nonlinear transition of electrons induced with an intense light field in the soft x-ray region is an attractive phenomenon because it can be expected to reveal new features of the interaction between electrons and photons, due to the energies of the photons being sufficiently high to singly ionize an atom with one photon. However, experimental research into this phenomena has been slow to progress, except for some superior works reported recently [1,2], because of the lack of intense ultrafast light sources in this wavelength region.

A free electron laser (FEL) is a promising apparatus for generating coherent short-wavelength light, although the highest photon energy in the FEL light is still much lower than that necessary for the ionization of helium [3].

The most favorable alternative to the FEL is the high-order harmonics generated with femtosecond laser pulses. The first demonstration of the nonlinear interaction of the high-order harmonics with an atom was the sideband generation in photoelectron spectra from a rare gas atom dressed with an intense visible laser field, which is the so-called two-color above-threshold ionization (two-color ATI) [4,5]. Application of the two-color ATI for characterization of the pulse of the harmonics [4,6–8] has been feasible. From the viewpoint of the nonlinear interaction, however, the sideband generation is only due to the high intensity of the visible laser pulse. The intensities of the high-order harmonics have been too low to induce the nonlinear effects, so that the two-color ATI remains a process with a single-photon in the harmonics.

Thus, the nonlinear transitions induced with the high-order harmonics have themselves attracted growing interest since the observation of two-photon ionization from an atom to a singly charged helium ion with the ninth harmonic of a Ti:sapphire laser pulse and the measurement of the pulse duration of this harmonic by autocorrelation [9]. This type of autocorrelation technique has been studied in detail [10,11] and recently applied in order to find the attosecond bunching generated with the Fourier synthesis of multiple phase-locked harmonics [2]. The spectrum of the electrons detached with two-photon absorption reflects the self-convolved spectrum of the harmonic(s), so that it can be interpreted as a frequency-resolved optical gating (FROG) signal in the autocorrelation measurement [12].

Miyamoto *et al.* have recently reported on the ATI of rare gas atoms induced by the harmonic pulse itself, the photon energy of which exceeds the ionization energy for a singly charged ion [1]. The physical content of this phenomena is similar to that of two-photon ionization in the sense that they can both be described as a one-electron system perturbated with two or more photons, although the ATI is very important for the engineering of the harmonics [13].

The double ionization of a helium atom or another kind of rare gas atom differs from the ionization process mentioned above because it may reveal the correlated energies and the momenta between the two detached electrons. The first observation of the double ionization of helium excited by many photons in a visible femtosecond laser field appeared to be a discrepancy of the doubly charged ion yield from the theoretical model, based on sequential ionization via the singly charged ion [14,15]. Weckenbrock *et al.* have now clarified the full characteristics of the momenta of two electrons leaving a doubly charged neon ion [16] after many investigations into this phenomenon over the course of a decade. Although there have also been many studies of the single-photon double ionization of helium [17–19], which is the simplest process for producing a correlated electron pair with a photon, by using extreme ultraviolet (XUV) light from a synchrotron source, we have not yet obtained experimental proof of the next simplest process: the two-photon double ionization (TPDI) of helium.

In this Letter, we report on the observation of doubly charged helium ions produced by the nonlinear interaction of helium atoms and photons in the 27th harmonic of a femtosecond Ti:sapphire laser pulse. A photon energy of

42 eV for the harmonic may ionize the singly charged ion to the doubly charged one through two-photon absorption, which is another nonlinear process distinguished from TPDI, because the potential energy for the ground state of the electron in the singly charged ion is 54.4 eV lower than that for the free state. The fact that the yield of the doubly charged ion depends quadratically on the intensity of the harmonic pulse in our experiment, however, suggests that two electrons escape directly from a helium atom climbing over the 79 eV potential barrier to achieve the double ionization with two-photon absorption. The second order nonlinearity of the ion yield enables us to measure the pulse duration of the 27th harmonic with an autocorrelation technique, and as a result, a duration of 8 fs is found.

The laser system used in the experiment is based on Ti:sapphire and delivers up to 70 mJ pulses at a 10 Hz repetition rate. The duration of the laser pulse is estimated to be 23 fs, with some subpeaks which may be due to the imperfect compensation of high-order dispersion through the laser amplifier chain. It has already been reported in Refs. [20,21] how the phase-matched harmonics can be generated and separated from the fundamental pulse. The reflected pulses of the 27th and other orders of the harmonics from a harmonic separator made of siliconcarbide, being from the seventh to 35th, are sent into a vacuum chamber for the measurement of the time-of-flight of ions for which they are passed through a 1-mm diameter hole bored at the center of two thin plates of stainless steel for acceleration.

The beam of the harmonics passes 5 mm away from the center of the 1 mm hole in the TOF plate in order to prevent a microchannel plate (MCP), which is placed 385 mm away from the hole, from detecting ions produced with the unfocused beam. A multilayer coat of siliconcarbide and magnesium on the surface of a concave substrate made of fused silica with a radius of curvature of 100 mm reflects 24% of the 42 eV photons with a bandwidth of 2.2 eV, which means that almost only the 27th harmonic pulse in the harmonics can be reflected by this concave mirror, and is focused in front of the hole. The spectrum of the harmonics can be monitored with a spectrometer set behind the TOF chamber by removing the concave mirror. We truncate the undesired edge of the harmonic beam by placing an aperture with a diameter of 3 mm between the beam separator and the TOF chamber, while the central part of the harmonic beam including approximately 70% of the harmonic energy can pass through this aperture due to the small divergence (~ 0.7 mm radians) of the phase-matched harmonics. This aperture also eliminates $\sim 94\%$ of the energy of the remaining fundamental pulse reflected by the beam separator, with the result that the energy of the fundamental pulse at the focal point of the concave mirror is reduced to ~ 100 nJ from 20 mJ at the entrance of the 5 m focal lens for the harmonic generation.

The intensity of the 27th harmonic at the focal point, that we notate as I_0, is estimated to be 1.7×10^{13} W/cm^2, assuming a 3 μm beam waist (w_0) [22], a 10 fs pulse duration (τ), and a 24 nJ pulse energy (E_0). The measured duration of the 27th harmonic pulse will be presented later. The intensity of the remaining fundamental is estimated to be $\sim 2 \times 10^{12}$ W/cm^2, which is approximately 10^{-2} of the intensity at which the helium ion can appear for the visible laser pulse [23]. In fact, we did not observe any helium ions when we applied only the fundamental pulses by switching off the supply of argon gas as a nonlinear medium for the harmonic generation.

The signals of ions detected with the MCP are sent to a digital oscilloscope with a bandwidth of 500 MHz and a sampling rate of 5 Gs/s, and then they are typically averaged over 10^3 shots for each measurement. We note that the signal of the doubly charged helium ion is sufficiently strong to appear in the averaged detection without the signal counting method conventionally used in this type of experiment.

Helium gas, as a target of the ion signals, is supplied via a glass tube with an inner diameter of 800 μm, which is connected to a pulsed gas valve operated synchronously with the laser pulse. We use the isotope helium 3 instead of helium 4 because our setup for the TOF experiment cannot resolve the signal of the singly charged ion of a hydrogen molecule from that of the doubly charged ion of a helium 4 atom. The estimated density of the atoms of helium 3 is $\sim 3 \times 10^{13}$ cm^{-3}.

A typical mass spectrum for the doubly charged ions of helium 3 is shown in Fig. 1. We can clearly see the presence of a peak at the mass number of 1.5 corresponding to the half of the mass for the helium 3 atom. The singly charged ions of the hydrogen atom at 1.0 and those of the hydrogen molecule at 2.0 mainly originate from water molecules that remain in the TOF chamber at the background pressure of 8×10^{-8} torr.

The physical model of the production of a doubly charged helium ion with the intense 27th harmonic pulse should include the three prime candidates for the nonlinear process of ionization. The first of the three candidates is sequential ionization via a singly charged ion to a doubly charged one through two-photon absorption. The second candidate requires three or four photons via the excited states or the ground state of an electron on a singly charged ion which is produced by the two-photon absorption of a neutral atom. The third candidate is simply TPDI from a neutral atom to a doubly charged ion without passing

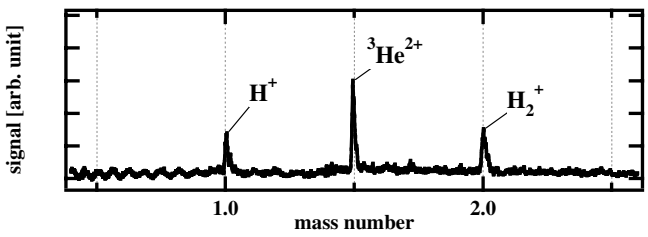

FIG. 1. Typical mass spectrum of doubly charged ions of helium 3 generated with the 27th harmonic pulses.

through any of the states for a singly charged ion. We note that this nonlinear process cannot be induced until the photon energy exceeds half of the energy required for double ionization, namely, 39.5 eV, which is much higher than the photon energies applied in the nonlinear experiments of harmonics reported to date [1,2,9,12,13].

Possible determinations of generalized cross sections of these nonlinear processes [24,25] and determinations of energy spectra of ejected electrons [26–29] have been theoretically examined by several authors. Thus, the experimental determination of these cross sections has great significance. However, instead we show a qualitative result for the dependence of the ion yield on the intensity of the 27th harmonic pulse, in order to clarify the most dominant process on the basis of the assumption that the ion yield is proportional to several ($2 \sim 3$) powers of the intensity.

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 probability of the ionization at the focal point, notated as $P_0^{(+)} = I_0 \tau \sigma^{(+)}/(h\nu)$, can be estimated to be much lower than unity (7×10^{-2} [30]) even if the intensity is the maximum, where $\sigma^{(+)}$, ν, and h are the cross section of one-photon ionization, the optical frequency of the 27th harmonic, and Planck's constant, respectively.

The result of this experiment is shown in Fig. 2. The yields of the doubly charged ions in relation to those of the corresponding singly charged ions are well located on the fitted line of $N^{(2+)} = A(N^{(+)})^{\alpha}$ in the logarithmic scale for both axes in this figure, where $N^{(2+)}$ is the yield of the doubly charged ion, $N^{(+)}$ is the yield of the singly charged ion, and A and α are the constants determined by fitting the line to the experimental data, respectively. We conclude that the exponent α is 2.0, and hence the two-photon absorption with double ionization is found to be the most dominant process in this experiment.

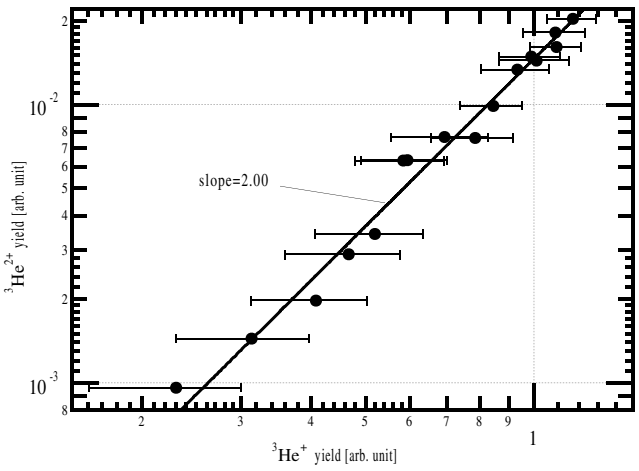

FIG. 2. Yields of the doubly charged ion of helium 3 versus those of the singly charged ion. Both are varied parametrically with the intensity of the 27th harmonic pulse. We fit the line to the plots with weighting values, which are reciprocals of the standard deviations indicated by error bars.

We can explain the reason why the TPDI should be dominantly observed in our experiment without exact calculations. Because the generalized cross sections for the TPDI and for the two-photon ionization from a singly charged ion should have the same order of magnitude without resonant enhancement [31], the primary difference between the yields for these processes originates from the numbers of neutral atoms and singly charged ions. We remember that $P_0^{(+)}$ is estimated to be 7×10^{-2}, and thus most of the doubly charged ions are produced via the TPDI process.

We next report on the application of the TPDI for the measurement of the pulse duration of the 27th harmonic pulse by means of an autocorrelation technique. Two replicas of the measured pulse are generally needed for the autocorrelation measurement. These are obtained, in our experiment, by spatially dividing the harmonic beam with two harmonic beam separators of siliconcarbide which are aligned with each other with a separation of approximately 100 μm. These separators are put in the place of the harmonic separator used in the previous experiment. The center of the harmonic beam enters the boundary of the separators, and then it is reflected separately. This spatial division of the harmonic beam is confirmed with a MCP and a CCD camera attached to the spectrometer behind the TOF chamber. We observe the semicircle profiles of the divided harmonic beams.

One of the harmonic separators is fixed on a translation stage with a piezoactuator that can move the stage within a length of 100 μm, hence the part of the harmonics reflected with this harmonic separator can be delayed or advanced to another part reflected with another harmonic separator within a range of ± 120 fs, which is calculated from the incident angle common to both harmonic separators of 69 degrees. Other configurations of the experimental setup are the same as those used in the previous experiment, or rather we simply replace the harmonic separator.

The yield of doubly charged helium ions to each delay is shown in Fig. 3. We can see a typical autocorrelation trace near zero delay in this figure while the subpeaks, which are generated from those of the fundamental pulse, appear at a delay of ± 40 fs. As was calculated by Tzallas *et al.* [2], 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 3, which agrees well with the ratio in the trace in Fig. 3. We note that this experimental result of the autocorrelation is supported by the theoretical investigation reported by Nakajima and Nikolopoulos [24].

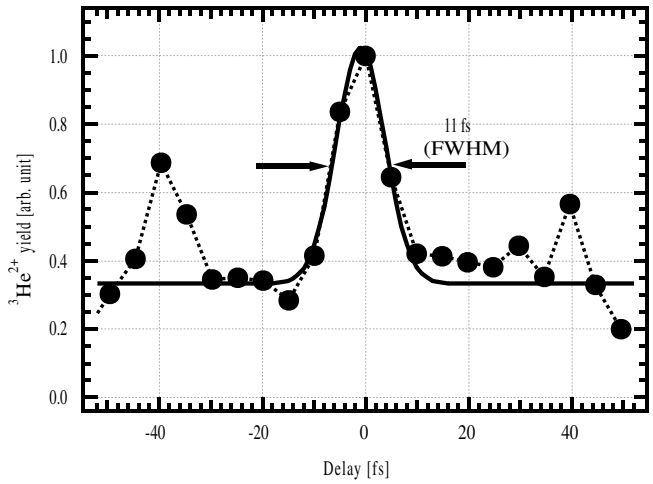

FIG. 3. Autocorrelation trace (solid circles with dotted line) of the 27th harmonic pulse obtained by utilizing the two-photon absorption for the yield of a doubly charged ion of helium 3. The full width at half maximum (FWHM) of the autocorrelation is determined by fitting a Gaussian shape (solid curve) to the trace.

A curve fitted to the central region of the trace assuming a Gaussian temporal profile, indicated as a solid curve in Fig. 3, results in a pulse duration of 8 fs corresponding to $1/\sqrt{2}$ of the full width at half maximum of the fitted curve. 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 and the phase matching conditions, and thus a quantum mechanical calculation of induced dipole moments is required [32] with an effect of the propagation obeying the Maxwell equations [33,34], which is beyond the scope of this Letter. Practical use of the power law of the intensity 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 quintic of the intensity of the fundamental, which is a reasonably good approximation to the result obtained under the quantum theory [4,32], 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 agrees well with the measured one.

In summary, we observed the yield of doubly charged helium ions produced with the 27th harmonic pulse of a femtosecond Ti:sapphire laser pulse. The dependence of the ion yield on the intensity of the 27th harmonic pulse ensures that the TPDI is dominant for the production of the doubly charged ions in the intensity range of 10^{13} W/cm^2. The use of the TPDI enables us to measure the pulse duration of the 27th harmonic pulse, and we found it to be 8 fs. The energy of 42 eV is the maximum energy, to the best of our knowledge, of the photons in the femtosecond pulse whose duration is measured by an autocorrelation technique.

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