Two-Photon Ionization of He through a Superposition of Higher Harmonics

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We present experimental results and theoretical analysis of two-photon ionization of He by a superposition of the 7th to the 13th harmonic of a Ti:sapphire laser. Solving the time-dependent Schrödinger equation for He in a coherent polychromatic field, the He^+ yield is calculated. From this yield the number of He^+ ions produced has been estimated and found in reasonable agreement with its measured value. The present results establish the feasibility of a second-order autocorrelation measurement of superposition of harmonics, and thus they represent the precursor towards the direct temporal characterization of attosecond pulse trains.

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The observation of a nonlinear process, e.g., multiphoton ionization or innershell transition, induced by short-pulse coherent extreme ultraviolet (XUV) radiation, such as the high-order harmonics of fs-laser radiation from rare gases, has been a challenging problem for a long time now. This is not only because of the interesting new physics inherent to the process, but also because it opens up the possibility of applying well-established approaches in fs-laser pulse metrology to pulses in the XUV wavelength region. Thus, second- or higher-order autocorrelation techniques can be appropriately modified for XUV radiation and utilized for the temporal characterization of individual higher-order harmonics or of a harmonic superposition. Since recently, there is experimental evidence that, in agreement with earlier theoretical suggestions [1], superposition of harmonics may form attosecond pulse trains [2,3] or even isolated attosecond pulses [4]. Such pulses provide a unique tool for real time measurements of ultrafast phenomena, mainly in processes involving ultrafast electron dynamics. Consequently, they are of great interest to a variety of scientific areas including atomic, molecular, solid state, and plasma physics, as well as to material science. The main prerequisite for the use of these pulses in ultrafast spectroscopy and nonlinear optics is their detailed and precise temporal characterization. Currently, most of the attosecond pulse characterization approaches are based on cross-correlation methods [3–5] complicating the quantitative pulse analysis, Instead, higher-order autocorrelation methods are more direct and provide rigorous and unequivocal information leading to complete pulse characterization. However, there are two main drawbacks linked with such measurements: (a) the lack of highly dispersionless devices in the XUV spectral region and (b) the low intensity combined with the low photon flux of the harmonics relative to the one needed according to the nonlinear XUV absorption cross sections. Collinear propagation geometries have been recently used as arrangements with negligible dispersion in cross-correlation measurements [3,4]. Furthermore, we have recently numerically assessed and experimentally tested the performance of a dispersionless Michelson interferometer based on a transmission grating beam splitter [6,7]. As for the observation of a nonlinear interaction of harmonics, only few experimental tries are reported in the literature because of the low multi-XUV-photon absorption rates. These include near resonant [7,8] and nonresonant [9–11] two-harmonic-photon ionization (3rd, 5th, or 9th harmonic), the highest harmonic (shortest wavelength) used thus far being the 9th harmonic of the Ti:sapphire fundamental (800 nm).

In this Letter, we report the observation of two-photon ionization of He induced by a broadband XUV spectrum consisting of the 7th harmonic ($\sim$113 nm) to the 13th harmonic ($\sim$61 nm) of a Ti:sapphire laser. This is a second-order process induced by the shortest wavelength and broadest bandwidth ever reported thus far. The observed rate is compared with that expected from the nonperturbatively *ab initio* calculated ionization yield, obtained by numerically solving the time-dependent Schrödinger equation (TDSE) for He in a polychromatic laser field. While a second-order autocorrelation measurement of isolated attosecond pulses seems at the moment to be far from realizable due to the extremely low photon number per pulse, the results of this work indicate that the realization of second-order autocorrelation measurements of attosecond pulse trains can be confidently foreseen for the near future.

The experiment was performed with the 10 Hz Ti:sapphire laser system at Max Planck Institut für Quantenoptik, ATLAS, delivering 130 fs long pulses, at

790 nm. A schematic of the experimental setup used is shown in Fig. 1. An annular beam with energy of about 20 mJ/pulse is used. The beam is focused with a 1.5 m lens into a xenon gas jet operated at the backing pressure of 5 atm. In this jet harmonics are generated. As it is experimentally determined, the harmonic power in the wavelength region of interest was maximized when the focus of the laser beam was positioned 4 cm after the Xe gas jet. A Kirkpatrick-Baez [12] focusing system is introduced in the propagation axis of the XUV radiation. This comprises two gold-coated spherical mirrors of 5 m radius of curvature. The angle of incidence of the XUV light was $i \sim 5° \pm 2°$. An aperture is placed after the second spherical mirror in order to completely block the fundamental of the laser. Directly after the aperture, the harmonics are filtered through a 0.16 μm thick indium filter. At the XUV focus, a second pulsed He gas jet is placed operating at a backing pressure of 5 atm. This second jet was equipped with a collimator tip 15 mm long with 1 mm inner diameter. The harmonic beam focus is located ~2 mm below the collimator tip. A 30 cm long ion time-of-flight energy analyzer is used for the detection of the He ions. The harmonics are monitored simultaneously with the ionization signal by means of an XUV monochromator coupled to the exit of the ionization chamber. The laser intensity at the first jet is estimated to be $\sim 4 \times 10^{13}$ W/cm^2. Although the intensity is not very high, the length of the emitting Xe medium ensures high conversion efficiency, which has been found under similar experimental conditions to be $\sim 10^{-5}$ [13]. This intensity gives rise to plateau harmonics each having an energy of ~200 nJ/pulse, which for the 9th harmonic translates to $\sim 8.8 \times 10^{10}$ photons/pulse. After the indium filter, the remaining harmonics are only the 7th, 9th, 11th, and 13th with a relative intensity ratio of 0.4:1:0.4:0.08, respectively. This intensity ratio was deduced after correction for the spectral response of the coupling mirror and of the XUV monochromator.

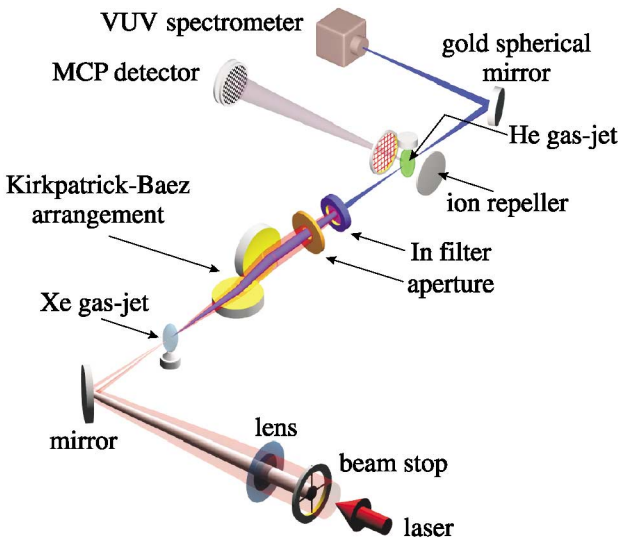

FIG. 1 (color online). The experimental setup.

With the above-described setup and condition, He^+ ions had been detected. Several tests have been performed to ensure that the He is ionized through a two-photon absorption process. Switching off the first jet, no He^+ ions were observed. The He^+ ion signal was further measured as a function of the total XUV intensity, which was varied by throttling the flow of the Xe gas. The results of this measurement in a log-log scale are shown in Fig. 2(a). Each point is the average over 100 sweeps for both the ion and the XUV signal. The slope of a fitted straight line is 2.3 ± 0.3. The number of ions observed in this experiment was estimated to be ~1/pulse for the lower up to ~17/pulse for the higher intensities. Except for the He^+ peak, two further contaminant peaks were observed in the spectra at longer flight times. They can be assigned to the CH^+ and H_2O^+ ions. The signal of H_2O^+ is measured together with the He^+ signal as a function of the initially incident laser energy. The results of the later measurement are depicted in Fig. 2(b). The slope of the fitted straight lines is found to be 4.9 ± 0.1 for the He^+ and 2.7 ± 0.3 for H_2O^+, which is compatible with the higher nonlinearity of the ionization process for He due to its higher ionization potential (ionization potentials: He: 24.6 eV; H_2O: 12.6 eV).

An estimation of the ionization rate at the present experimental conditions has been further performed. For this estimation, we numerically solve the TDSE of helium in the polychromatic laser field, using an *ab initio*, **B**-splines base, configuration interaction two-electron method [14,15]. We will briefly highlight the main features of the technique. The TDSE for an atomic system in the polychromatic laser field is written as

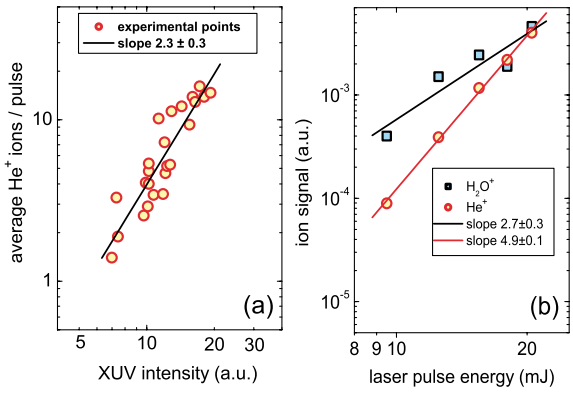

FIG. 2 (color online). (a) Number of He^+ ions produced per pulse as a function of the total XUV light intensity consisting of the harmonics passing through the indium filter in a log-log scale. The slope of the fitted straight line is 2.3 ± 0.3. (b) He^+ (circles) and H_2O^+ (squares) ion signal as a function of the laser pulse energy. The fitted straight lines exhibit different slopes, due to the different ionization thresholds.

$$i\partial_t\psi(t) = [H_0 + D(t)]\psi(t), \quad (1)$$

with H_0 the field-free helium Hamiltonian and D the interaction between the system and the laser field. In the length gauge and within the dipole approximation $D(t) = -\vec{E}(t)\cdot\vec{r}$, where $\vec{r} = \vec{r}_1 + \vec{r}_2$ is the position vector of the active electrons of the system and $\vec{E}$ is the electric field. In our calculations, we assume harmonic fields linearly polarized along the z axis with a $\cos^2(\pi t/\tau_N)$ normalized pulse envelope of total duration τ_N for the Nth harmonic. The time-dependent wave function is expanded onto a stationary eigenbasis,

$$\psi(t) = \sum_{nL} C_n^L(t)\Phi_{E_n}^L + \sum_L \int dE C_E^L(t)\Phi_E^L, \quad (2)$$

constructed within a box.

The basic idea and the formal details of the construction of the two-electron states (with total angular momentum L) can be found elsewhere [16–18]. We obtain the two-electron states $\Phi_{N(E)}^{SL}$ by diagonalizing the Hamiltonian in the basis spanned by two-electron basis functions $\Psi_{n_1 l_1, n_2 l_2}^{SL}(\vec{r}_1, \vec{r}_2)$ constructed as an antisymmetrized product of one-electron hydrogen orbitals. From (1) and (2), we obtain a system of ordinary differential equations (ODE) for the time-dependent coefficients [19] $C_{E_n}^L(t) \rightarrow C_n^L(t)$,

$$i\frac{d}{dt}C_n^L(t) = E_n C_n^L(t) + \sum_{L',n'} \langle \Phi_n^L | D(t) | \Phi_{n'}^{L'} \rangle C_{n'}^{L'}(t). \quad (3)$$

Integration of the above system of differential equations, subject to the proper initial conditions, provides the coefficients at the end of the pulse, $t \rightarrow \infty$.

For the calculation, we have assumed a fundamental field at 790 nm and the polychromatic field consisting either of one of the four or the superposition of the $N = 7$, 9, 11, and 113 harmonics with the experimental intensity ratios. In addition, we have set their phases to zero. The total duration at FWHM of each of the harmonics has been assumed to follow the rule $\tau_N = 130/\sqrt{N}$ fs since measured harmonic durations do not deviate significantly from their perturbative value [20].

The results of the theoretical simulation are depicted in Fig. 3. This figure shows the yield of two-photon ionization by each of the individual harmonics (9th, 11th, 13th, and 15th) as a function of its intensity (points) and by the superposition of the harmonics used in the experiment as a function of the total XUV intensity (line). Within the assumption of negligible phase difference between harmonics, it is clear that the superposition of the harmonics 7th, 9th, 11th, and 13th results to an about one order of magnitude more efficient two-photon ionization yield of He gas as compared with the two-photon-ion yield caused by each individual (9th, 11th, and 13th) harmonic at the same intensity and about a factor of 3 higher yield than the sum of the ionization yield caused by each harmonic,

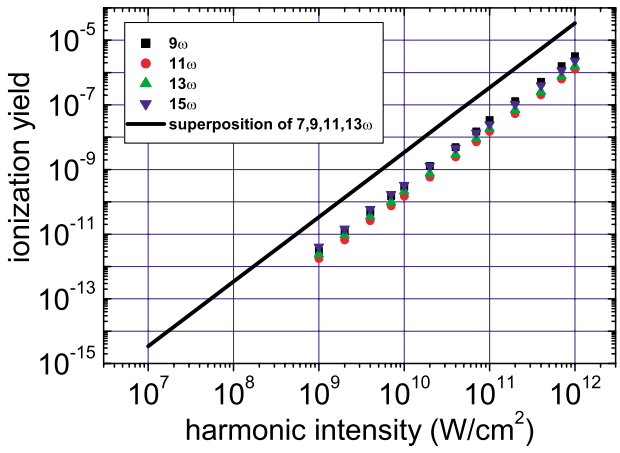

FIG. 3 (color online). The calculated yield of He^+ ions as a function of the intensity of the harmonic superposition 7th, 9th, 11th, and 13th with relative ratio 0.4:1:0.4:0.08, respectively, and no phase difference (solid line).

i.e., the ionization yield caused by the incoherent sum of all the harmonics. The calculation shows also a negligible difference for the two-photon ionization yield produced by each individual harmonic from the 9th to the 15th one. These are two notable results that open favorable perspectives for a second-order autocorrelation measurement utilizing a superposition of harmonics. The flat ionization yield found for the harmonic wavelengths considered merits special attention. Our calculations show that the response of a nonlinear medium to a given set of harmonics is very specific to the atomic system used [18]. The two-photon cross section for the wavelength region under investigation is flat except for resonances too far detuned from the harmonic frequencies to make any difference. Indeed, the specific case we have considered here shows that there is no spectral "filtering" of the higher harmonics by the ionization process itself and that, furthermore, the harmonic superposition leads to an enhanced ionization rate. This is very important for future second-order autocorrelation measurements, for which a flat response detector and enhanced ionization signal is required.

Additionally, in order to obtain an estimate of the harmonic intensity at the interaction region, we have performed a ray tracing analysis for the optical setup used in the experiment. The ray tracing calculations give for the laser beam at the Xe gas jet a spot diameter of $d_L \sim 0.7$ mm. For the diameter d_N, the divergence Θ_N and the pulse duration T_N of each harmonic we assume (perturbation theory) $d_N = d_L/\sqrt{N}$, $\Theta_N = \Theta_L/\sqrt{N}$, $\tau_N = \tau_L/\sqrt{N}$, where L indicates the corresponding quantity of the fundamental.

With 9th harmonic energy of $E_9 \sim 200$ nJ [13] and taking into account the reflectivity of the gold mirrors $R_M = 54\%$, the aperture transmission $T_A = 66\%$, the indium filter transmission $T_F = 0.3$, and the spot size

$S = 6.0 \times 10^{-3}$ cm^2, the 9th harmonic peak intensity at the second jet is calculated from

$$I_9 \approx R_M T_A T_F \times \frac{E_9}{\tau_9 S}, \quad (5)$$

and is found to be 8×10^7 W/cm^2. Consequently, the corresponding total XUV intensity in the superposition is $\sim 1.5 \times 10^8$ W/cm^2.

The He pressure at the interaction volume is estimated to be ~ 3 Torr [21]. Thus, the atomic density at room temperature is $N_a \approx 10^{17}$ atoms/cm^3. The longitudinal extent of the interaction region is obtained from the divergence angle of the atomic beam, which is $\alpha = 28°$ for this type of gas jet with flow collimator [21]. Accordingly, the interaction length is ~ 1.0 mm. The number of ions produced per laser pulse is obtained from

$$N_{\text{ions}} = Y N_a V F_D, \quad (6)$$

where Y is the calculated ion yield produced by the mixing of the harmonics (see Fig. 3), $V = 5 \times 10^{-4}$ cm^3 the interaction volume, and $F_D \approx 50\%$ the overall efficiency of the time-of-flight spectrometer. The thus estimated number of He ions detected per pulse reads then $N_{\text{ions}} \approx 20$ for $i = 5°$. Since there is an uncertainty in the determination of the angle of incidence in the Kirkpatrick-Baez arrangement, this lying between 3° and 7°, we have repeated the calculation for $i = 3°$ and $i = 7°$ as well. For the three angles, the resulting average number of He ions for the condition of our experiment is then $\langle N_{\text{ions}} \rangle \approx 30 \pm 18$. Although this estimation could only be carried out partially based on perturbation theory, harmonic generation being a nonperturbative effect, this number is in reasonable agreement with the experimental value of ~ 17 ions per pulse.

In conclusion, we have demonstrated two-photon ionization of He gas using the particularly broad band XUV radiation of the 7th to the 13th harmonic superposition. The two-photon yield has been calculated and found essentially constant for the individual harmonics, and enhanced for the superposition of the harmonics. These results define the conditions for and open up the prospect of conducting second-order autocorrelation measurements of coherent superposition of higher harmonics aiming at the unambiguous quantitative temporal characterization of attosecond pulse trains.

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