

N.A. PAPADOGIANNIS¹
L.A.A. NIKOLOPOULOS¹
D. CHARALAMBIDIS^{1,2}
G.D. TSAKIRIS^{3,✉}
P. TZALLAS³
K. WITTE³

On the feasibility of performing non-linear autocorrelation with attosecond pulse trains

¹ Foundation for Research and Technology - Hellas, Institute of Electronic Structure and Laser, Laser and Applications Division, P.O. Box 1527, GR-71110 Heraklion (Crete), Greece
² Department of Physics, University of Crete, P.O. Box 2208, GR-71003 Voutes-Heraklion, Greece
³ Max-Planck-Institut für Quantenoptik, 85748 Garching, Germany

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ABSTRACT We present experimental results in which a second-order effect, namely two-photon ionization of atomic He induced by a superposition of harmonics, is observed. The harmonics are generated in a Xe gas-jet using a 790-nm 10-Hz femtosecond Ti:sapphire laser and are subsequently focused into a He gas-jet with a Kirkpatrick–Baez arrangement. The superposition is formed by using a thin In filter and it comprises the 7th to 13th harmonics. Solving the time-dependent Schrödinger equation for He in a polychromatic laser field, the He⁺ ion yield is calculated as a function of the total XUV intensity. Using the calculated yield and taking into account the focusing and transmission properties of the arrangement, the number of He⁺ ions produced per laser pulse is estimated and is found to be in reasonable agreement with its measured value. The total number of ions produced non-resonantly follows a nearly quadratic dependence on the harmonic intensity, thus establishing the feasibility of a second-order auto-correlation measurement of the superposition of harmonics, i.e., of a direct temporal characterization of attosecond pulse trains.

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1 Introduction

Recently, experimental evidence has become available that a superposition of harmonics may form attosecond pulse trains [1, 2] or even isolated attosecond pulses [3], in agreement with earlier theoretical conjectures [4]. Motivated by these developments, applications in atomic spectroscopy have been suggested [5] and the first results involving single attosecond pulses have been reported [6, 7]. The main prerequisite for the use of these pulses in ultrafast spectroscopy and non-linear optics is their detailed and precise temporal characterization. Currently, most of the attosecond pulse characterization approaches are based on indirect cross-correlation methods [2, 3, 8], complicating the quantitative pulse analysis. In contrast, higher order auto-correlation methods are more direct and provide rigorous and unequivocal information leading to complete pulse characterization. In connection with these experiments, the observation of a non-linear process, e.g. multi-photon ionization induced by short-pulse

coherent XUV radiation, such as the high-order harmonics of fs-laser radiation, 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 paves the way to utilization of well-established approaches in fs-laser pulse metrology to short pulses in the XUV wavelength region. Thus, second- or higher-order auto-correlation techniques can be appropriately modified for XUV radiation and applied for the temporal characterization of individual higher order harmonics or of a harmonic superposition. However, there are two main obstacles associated 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 that needed for observable signals according to the non-linear XUV absorption cross-sections. Collinear propagation geometries have been recently used as arrangements with negligible dispersion in cross-correlation measurements [2, 3]. Furthermore, we have recently numerically assessed and experimentally tested the performance of a dispersionless Michelson interferometer based on a transmission-grating beam-splitter [9, 10]. As for the observation of a non-linear interaction involving harmonics, only few experimental attempts have been reported in the literature because of the low multi-XUV-photon absorption rates. These include a near-resonant two 3rd-harmonic-photon ionization of various gases at 267 [10] and 83 nm [11], a two- and three-photon non-resonant ionization of He with the 9th [12] and 5th harmonics [13], respectively, of a Ti:sapphire laser, and more recently, two and three photon non-resonant ionization of rare gases [14] by the 5th harmonic of a Ti:sapphire laser. Higher-order auto-correlation measurements of single harmonics are reported as well in [10] (interferometric and intensity auto-correlation), and in [12] and [13] (intensity auto-correlation).

In this work we report the observation of two-photon ionization of He induced by a broadband XUV spectrum consisting of the 7th harmonic (~ 113 nm) up to the 13th harmonic (~ 61 nm) of a Ti:sapphire laser. The harmonic intensity dependence of the total number of ions produced gives unequivocal evidence that the measured signal is due to a two-photon ionization process in He. The observed rate is compared with that expected from the *ab initio*-calculated ionization yield, obtained by numerically solving the time-dependent Schrödinger equation (TDSE) for He in a polychromatic laser

✉ Fax: +49-89/3290-5200, E-mail: tsakiris@mpq.mpg.de

field. While a second-order auto-correlation 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 performing second-order auto-correlation measurements of attosecond pulse trains appears to be within our reach and it can be confidently foreseen for the near future.

2 Experimental setup

The experiment was performed with the first module of the 10 Hz Ti:sapphire laser system at the Max-Planck-Institut für Quantenoptik, ATLAS, which delivers 130-fs long pulses, with an energy of up to 250 mJ and a spectral distribution centered at 790 nm. A schematic of the experimental set-up used is shown in Fig. 1. The input laser beam was clipped down to 2.5 cm in diameter. A beam stop of 1-cm diameter was introduced into the beam cross-section, forming an annular-shaped beam with an energy of about 20 mJ/pulse. The beam was focused with a 1.5-m lens into a piezoelectrically pulsed Xenon gas-jet (Lasertechnics LPV) operated at a backing pressure of 5 atm. The orifice of the jet had a diameter of 1 mm. In this jet harmonics were generated. As was 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, which is in agreement with previous observations and discussions [15, 16]. It is worth noting that theoretical calculations predict poor phase locking of the harmonics when the laser is focused after the gas jet [17]. However, here our main concern is to demonstrate two-photon ionization, for which the maximum possible harmonic power is required. A Kirkpatrick–Baez [18] focusing system was introduced on the propagation axis of the XUV radiation. This comprised two 10-cm diameter gold-coated spherical mirrors of 5-m radius of curvature, one fixed in the x - y plane and the other in the x - z plane. The angle of incidence of the XUV light at the first and second mirrors was $\sim 5^\circ$. At this nearly graz-

ing angle of incidence, the combined reflectivity of the two mirrors within the spectral region of interest was $\simeq 50\%$. An aperture with a diameter of 0.8 mm was placed 9.5 cm after the center of the second spherical mirror, completely blocking the fundamental of the laser in the annular section, while allowing the harmonics in the center to pass through. Directly after the aperture the harmonics were filtered through a 0.16- μm thick indium filter exhibiting $\sim 30\%$ transmission at 88 nm (9th harmonic). The Kirkpatrick–Baez arrangement focused the XUV light 44.5 cm after the center of the second mirror in both directions transverse to the propagation axis. A second pulsed gas-jet of the same type as the first one was placed at the XUV focus, but was filled instead with He at a backing pressure of 5 atm. This second jet had an orifice 1 mm in diameter and was equipped with a collimator tip 15-mm long with 1-mm inner diameter. The harmonic beam focus was located 1.5–2 mm below the collimator tip and was centered with respect to the gas-jet cone, where the He atomic density was the highest, thus maximizing the height and sharpness of the He⁺ peak in the TOF spectra. A 30-cm long ion time-of-flight energy analyzer having its axis perpendicular to the XUV propagation direction and to the He jet beam was employed for the detection of the He ions (see Fig. 1). The harmonics were monitored simultaneously with the ionization signal by means of an XUV monochromator coupled to the exit of the ionization chamber via a gold mirror with a 60-cm radius of curvature (see Fig. 1). The laser intensity at the first jet was estimated to be $2\text{--}4 \times 10^{13} \text{ W/cm}^2$. Although the laser intensity was not very high, the length

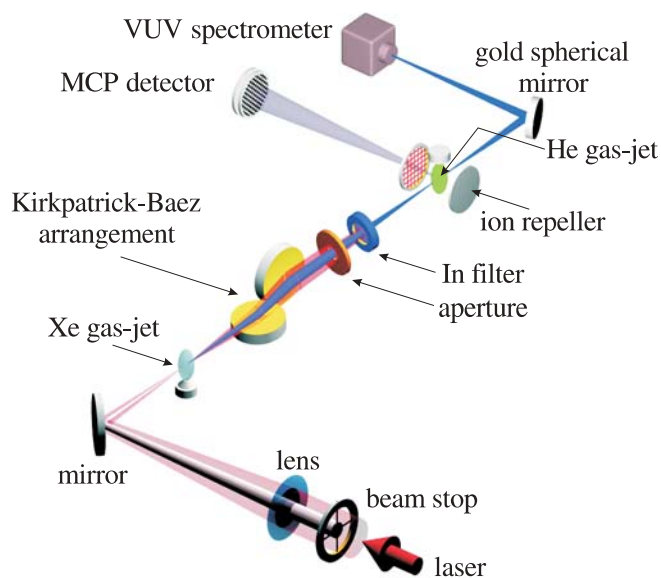


FIGURE 1 The experimental setup

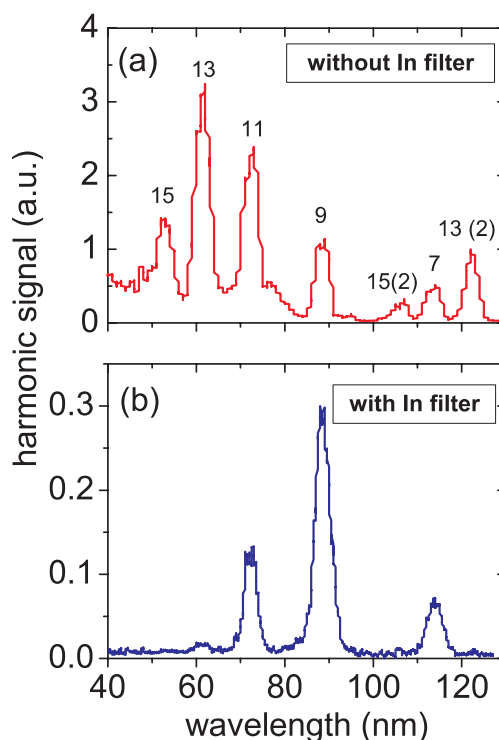


FIGURE 2 Harmonic spectrum produced by the Xe gas-jet **a** before and **b** after the 0.16- μm thick In filter. For each harmonic peak, the ratio obtained from the two graphs gives merely the transmission of the In filter at the particular wavelength. The relative amplitudes in both graphs are not corrected for the spectral response of the monochromator and detector

of the emitting Xe medium ensured a high conversion efficiency, which has been measured under similar experimental conditions to be $\sim 10^{-5}$ [16]. This intensity gives rise to plateau harmonics with energies of 200 nJ/pulse, which translates to $\sim 8.8 \times 10^{10}$ photons/pulse. Figure 2 shows the harmonic spectrum recorded by the XUV monochromator without (a) and with (b) the indium filter. After the indium filter, the remaining harmonics were only the 7th, 9th, 11th, and 13th with relative intensities of 0.4 : 1 : 0.4 : 0.08, respectively. This intensity ratio was deduced after taking into consideration the spectral response of the coupling mirror, that of the XUV monochromator, and the quantum yield of the detector.

3 Experimental results

With the setup and conditions described above, He^+ ions were detected. Several tests were performed to ensure that the He was ionized through a two-photon absorption process. Switching off the Xe gas-jet and hence the harmonic generation, no He^+ ions were observed. This presents unequivocal proof that the ionization was caused by the XUV photons in the harmonic superposition. The He^+ ion signal was further measured (a) as a function of the total XUV intensity, which was deduced from the area of the photo-peak present at $t = 0$ in the time-of-flight signal, and (b) as a function of the intensity of individual harmonics obtained from the harmonic peaks in the XUV spectrum (see Fig. 2). The XUV intensity was varied by changing the voltage on the piezoelectric crystal of the Xe gas-jet, thus by throttling the flow of the Xe gas.

The results of these measurements on a log–log scale are shown in Figs. 3 and 4, respectively. Each point is the average

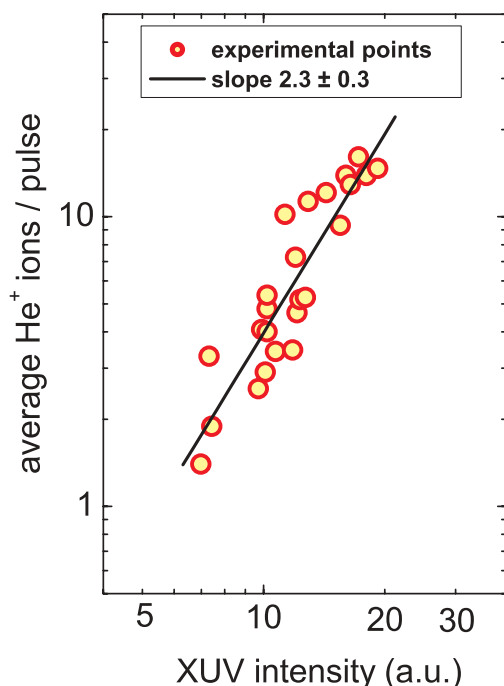


FIGURE 3 Number of He^+ ions per pulse as a function of the total intensity of the XUV light consisting of harmonics passing through the In filter

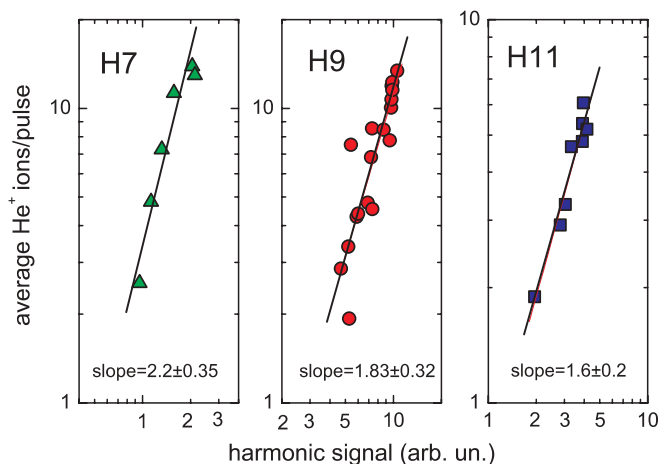


FIGURE 4 Number of He^+ ions per pulse as a function of the intensity of the 7th (triangles), 9th (circles), and 11th harmonics (squares)

over 100 pulses for both the ion and XUV signals. The slope of a fitted straight line in Fig. 3 is 2.3 ± 0.3 . As can be seen from Fig. 4, the variation of the ion number as a function of the intensity of the 7th, 9th, and 11th harmonics, was also quadratic within the experimental error. Since the number of He^+ ions measured is due to the total harmonic intensity, which in turn is proportional to the intensity of each individual harmonic, the results in Fig. 4 represent an independent confirmation of the quadratic dependence of the ion yield on the harmonic intensity. 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 of heavier elements were observed in the spectra at longer flight times. The signal of one of them was measured together with the He^+ signal as a function of the initially incident laser energy. It was found that while the signal scales as $\propto I_L^{4.9 \pm 0.1}$ for He^+ , the corresponding scaling was $I_L^{2.7 \pm 0.3}$ for the heavier element. This is compatible with the higher non-linearity of the ionization process for He due to its high ionization potential (24.6 eV).

4 Theoretical analysis

The combination of He as ionization medium and In as filter material for the selection of a group of harmonics is a judicious choice presenting several advantages. This becomes clear if one considers the possible ionization channels due to the available photon energies in relation to the ionization threshold of 24.6 eV for He. These channels are schematically shown in Fig. 5. On the high frequency side, only harmonics higher than the 15th (17th, 19th, ...) can give rise to single-photon ionization. For the laser intensity used, the cut-off harmonic for Xe is the 11th [19]. Therefore, the 17th harmonic is produced at a greatly reduced efficiency and, in addition, the In filter further attenuates it by 2–3 orders of magnitude. As can be seen in Fig. 2, in which the transmitted spectrum through the 0.16- μm In filter is shown, there was no measurable signal corresponding to the 17th harmonic at 46.5 nm. Although it is difficult to a priori ascertain whether the 17th harmonic was sufficiently suppressed as not to cause single-photon ionization, the experimental results presented

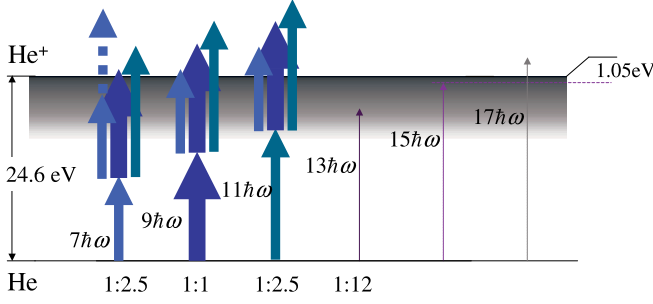


FIGURE 5 Schematic diagram showing the possible ionization channels for the harmonics in the superposition. *Dashed lines* indicate absorption channels with negligible contribution under the present experimental conditions

in Sect. 3 provide clear evidence that the influence of the 17th harmonic was negligible. On the low frequency side, the 7th harmonic was transmitted, but all lower harmonics were practically filtered out. Moreover, all harmonics below the 7th can induce in the non-mixing combination three-photon ionization only. As is discussed later on in this section, the corresponding yield in this case is many orders of magnitude less than that of the two-photon ionization process. With the exemption of the 7th harmonic, the remaining harmonics in the superposition can induce only two-photon ionization, either by two photons of the same energy or by any combination of the photon energies in the superposition.

It follows that an estimation of the ionization rate under the present experimental conditions has to take into account all of the possible channels. For this estimation, we numerically solved the TDSE of helium in a polychromatic laser field, using an ab-initio, B-splines-based, configuration interaction (CI) two-electron method [20, 21]. We will briefly highlight the main features of the technique. All equations that follow are given in atomic units. The TDSE for an atomic system in a polychromatic laser field is written as

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

with H_0 being the field-free helium Hamiltonian and D the time-dependent interaction between the system and the laser field. In the length gauge and within the dipole approximation, the interaction operator is $D(t) = -\mathbf{E}(t) \cdot \mathbf{r}$, where $\mathbf{r} = \mathbf{r}_1 + \mathbf{r}_2$ is the position vector of the active electrons of the system and \mathbf{E} the electric field. In our calculations, we assumed harmonic fields linearly polarized along the z -axis with

$$\mathbf{E}(t) = \hat{z} \sum_N E_N f_N(t) \sin(N\omega t + \varphi_N) \quad (N = 7 - 15), \quad (2)$$

where $f_N(t) = \cos^2(\pi t/\tau_N)$ is the normalized pulse envelope of total duration τ_N for a field of angular frequency ω_N , and E_N is the maximum field amplitude for the N -th harmonic.

In our two-electron calculations, we expanded the time-dependent wave function onto a stationary eigenbasis constructed within a box, an approach that has been used in calculations in many atomic systems [20, 22, 23]. Given that the ground state of helium is an 1S_0 state and considering that for linearly polarized light and dipole-allowed transitions, $\Delta S = 0$, $\Delta M_L = 0$, only the singlet states need to be included

in the expansion of the time-dependent wavefunction $\psi(t)$, we have

$$\psi(t) = \sum_{nL} C_n^L(t) \Phi_{E_n}^L + \sum_L \int dE C_E^L(t) \Phi_E^L. \quad (3)$$

The basic idea and the formal details of the construction of the two-electron states (with total angular momentum L) can be found elsewhere [24, 25]. We obtained the two-electron states Φ_E^{SL} by diagonalizing the Hamiltonian in the basis spanned by two-electron basis functions $\Psi_{n_1l_1, n_2l_2}^{SL}(\mathbf{r}_1, \mathbf{r}_2)$ constructed as antisymmetrized products of one-electron hydrogen orbitals. The radial part of the one-electron orbitals was expanded on a set of 600 B-splines of order $k = 9$ inside a finite interval $[0, R]$ with R up to 800 a.u. The two-electron basis functions Φ_E^{SL} were constructed in terms of about 2000 configurations (n_1l_1, n_2l_2) for the symmetry $L = 0$ and about 1500 for each of the other symmetries. Inserting the expansion in (3) into the TDSE (1) we obtained a system of ordinary differential equations (ODE) for the time-dependent coefficients $C_{E_n}^L(t) \rightarrow C_n^L(t)$,

$$i\frac{d}{dt}C_n^L(t) = E_n C_n^L(t) + \sum_{L'n'} \langle \Phi_n^L | D(t) | \Phi_{n'}^{L'} \rangle C_{n'}^{L'}(t). \quad (4)$$

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 assumed a fundamental laser field at 790 nm and a polychromatic field consisting either of one of the four $N = 9, 11, 13$, and 15 or the superposition of the four harmonics $N = 7, 9, 11$, and 13. In addition, we set their phases to zero (phase-locked harmonics). The peak intensity ($I_N \propto |E_N|^2$) of each of the harmonics followed the experimental ratios $I_7 : I_9 : I_{11} : I_{13} \rightarrow 0.44 : 1 : 0.45 : 0.085$. The total duration at FWHM of each of the harmonics was assumed to follow the rule $\tau_N = 130/\sqrt{N}$ fs. Although this is a perturbation theory result, it is found that it does not deviate considerably from the values for the harmonic duration obtained experimentally [26]. The convergence of the results was checked against the box radius (up to 800 a.u.), the number of B-splines (up to 1000), the highest total angular momentum (up to $L = 5$), as well as in the total number of two-electron states (up to 1000) included in the basis.

Figures 6 and 7 summarize the results of the theoretical simulation. Figure 6 shows the yield of two-photon ionization by each of the individual 9th, 11th, 13th, and 15th harmonics as a function of its intensity (points) and by the superposition of the harmonics used in the experiment (7th, 9th, 11th, and 13th) 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 7th, 9th, 11th, and 13th harmonics results in an approximately one order of magnitude more efficient two-photon ionization yield of He gas compared with the two-photon-ion yield caused by each individual (9th, 11th, or 13th) harmonic at the same intensity and about a factor of three higher yield than the sum of the ionization yields caused by each harmonic, i.e. the ionization yield caused by the incoherent sum of all harmonics. A related study pertaining to the ionization of hydrogen by

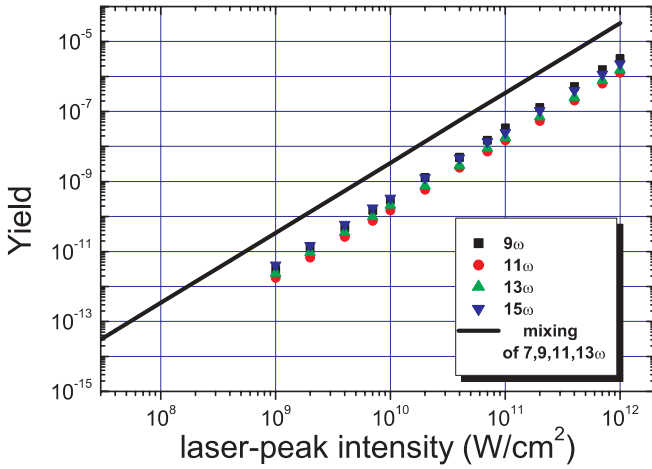


FIGURE 6 The calculated yield of He^+ ions as a function of the intensity of different harmonics (points) and as a function of the total intensity of the superposition of the 7th, 9th, 11th, and 13th harmonics with relative ratios 0.44 : 1 : 0.45 : 0.085, respectively, and no phase difference (solid line)

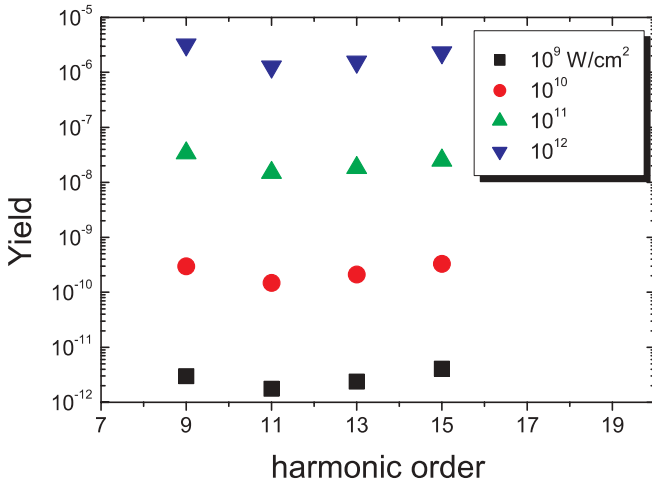


FIGURE 7 Variation of the two-photon ion yield of the indicated harmonics for different intensities

a group of harmonics shows similar effects [27]. As can be seen in Fig. 7, the calculation shows also a negligible variation in the two-photon ionization yield produced by each individual harmonic from the 9th to the 15th. These are two notable results that open favorable perspectives for a second-order auto-correlation measurement utilizing a superposition of harmonics. The flat ionization yield found for the harmonic wavelengths considered (see Fig. 7) merits special attention. Our calculations and those reported in [23] have shown that the response of a non-linear medium to a given set of harmonics is very specific to the atomic system used. Thus, the predicted rapid decrease for shorter wavelengths of the two-photon ionization cross-section, $\sigma^{(2)}(\omega) \propto \lambda^6$ for the case of absence of intermediate near resonances [28, 29], appears not to be generally applicable. Indeed, in the more detailed calculation of [23] it is seen that although He exhibits resonances in the spectral range we have considered here, the harmonics comprising the superposition do not fall within these resonances. This is a unique property of He associated with its large ionization potential (24.6 eV), which causes the first in-

termediate resonance ($|1s2p\rangle$) to be at ~ 20 eV. This fortuitous coincidence results in a flat response without spectral “filtering” or “enhancement” of the higher harmonics by the ionization process itself. We also calculated (but do not show here) the three-photon ionization yield by the 7th harmonic, which was found to be at least 4 orders of magnitude lower for the range of intensities of Fig. 6. The higher two-photon ionization yield obtained by the coherent superposition of the harmonics compared with that produced by any of the individual harmonics may be understood in terms of the additional ionization channels opened by the mixing of two harmonics or by the temporal profile of their coherent superposition.

5 Ionization estimates

In order to obtain an estimate of the number of ions produced per pulse by the harmonic superposition, one needs the harmonic intensity at the interaction region. This was obtained by means of a ray-tracing analysis of the optical setup used in the experiment. The ray tracing calculations gave for the laser beam at the Xe gas-jet a spot diameter of $d_L \simeq 0.7$ mm. For the diameter d_N , the divergence Θ_N , and the pulse duration τ_N , of the N -th harmonic, we assumed (perturbation theory) $d_N = d_L/\sqrt{N}$, $\Theta_N = \Theta_L/\sqrt{N}$, and $\tau_N = \tau_L/\sqrt{N}$ where ‘L’ indicates the corresponding quantity of the fundamental. Table 1 contains the results of the ray-tracing calculations and the estimated number of ions produced per pulse for three different angles of incidence at the mirrors of the Kirkpatrick–Baez arrangement ($i = 3^\circ, 5^\circ, \text{ or } 7^\circ$). The reason for considering a variation in the angle of incidence is that, in practice, it is difficult to accurately determine its value, but the final result depends sensitively on it. The ray-tracing analysis from the point of harmonic generation (Xe gas-jet) to the interaction region (He gas-jet) was performed for the 9th harmonic, which is also the most dominant. The total harmonic intensity was then obtained using $I_H \approx \alpha I_9$, where the factor $\alpha = 0.44 + 1.0 + 0.45 + 0.085$ takes into the account the

i	3°	5°	7°
R_M	70%	54%	43%
T_A	92%	66%	55%
R_{9y} (cm)	0.046	0.068	0.072
R_{9z} (cm)	0.045	0.028	0.048
S (cm ²)	6.5×10^{-3}	6×10^{-3}	11×10^{-3}
T_F		0.3	
I_9 (10^7 W/cm ²)	13.7	8.3	3.0
I_H (10^7 W/cm ²)	27.1	16.4	6.0
Y	2.2×10^{-12}	8.1×10^{-13}	1.1×10^{-13}
V (cm ³)	6.0×10^{-4}	5×10^{-4}	1.0×10^{-3}
N_a (cm ⁻³)		1×10^{17}	
F_D		50%	
N_{ions}	66	20	5
$\langle N_{\text{ions}} \rangle$	30 ± 18		

TABLE 1 Results of the ray-tracing analysis of the Kirkpatrick–Baez arrangement and the estimated number of ions per pulse N_{ions} for three angles of incidence i . Here R_M is the combined reflectivity of the two mirrors, T_A the aperture transmission, R_{9y} and R_{9x} the spot radii at focus, S the focal spot area, T_F the In filter transmission at 88 nm, I_9 the 9th harmonic intensity, I_H the total harmonic intensity, Y the calculated ion yield corresponding to I_H (see Fig. 6), V the interaction volume, N_a the atomic density, and F_D the overall efficiency of the time-of flight detector

relative contribution of each harmonic to the superposition. This is also in accordance with the calculations described in Sect. 4.

With the 9th harmonic energy of $E_9 \simeq 200$ nJ per pulse [16], the 9th harmonic peak intensity at the second jet was calculated from

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

The expression (5) takes into account the reflectivity of the gold mirrors R_M , the transmission T_A through the 0.8-mm aperture (see Fig. 1), the spot size S , and the indium filter transmission T_F (see Table 1). Due to the optical properties of the Kirkpatrick–Baez arrangement the spot at focus was elliptical with radii R_{9y} and R_{9z} in the two transverse directions, hence $S = \pi R_{9y} R_{9z}$. For a total XUV intensity of $I_H \approx \alpha I_9$, the yield Y was then obtained from the theoretically calculated ion yield produced by the coherent mixing of the harmonics (line in Fig. 6).

The He pressure at the interaction volume was estimated to be 3 Torr [30]. Thus the atomic density at room temperature was $N_a \approx 1.0 \times 10^{17}$ atoms/cm³. The longitudinal extent of the interaction region was estimated from the divergence angle of the atomic beam, which is $\alpha = 28^\circ$ for this type of gas-jet with flow collimator [30]. Thus the interaction length was ~ 1.0 mm. The ray-tracing calculations show that the length of the focal volume formed by the Kirkpatrick–Baez arrangement was much larger than the atomic beam extent. Consequently the interaction volume V was defined by the width of the atomic beam in the propagation direction and the focal spot in the transverse direction. The number of ions produced per laser pulse was calculated from

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

where $F_D \approx 50\%$ is the overall efficiency of the time of flight spectrometer. From Table 1 it is seen that for an angle of incidence $i = 3^\circ$ the number of ions per pulse N_{ions} was ~ 66 , but only ~ 5 for $i = 7^\circ$. Taking into account the uncertainty in the angle of incidence, the estimated average number of He ions for the conditions of our experiment was 30 ± 18 . This is within a factor of 2–3 in agreement with the experimental value of ~ 17 ions/pulse. Although our experimental conditions were quite different, the total number of ions produced per pulse is almost the same as that observed in a previous experiment using only the 9th harmonic [12]. In the present experiment, besides using a superposition of harmonics (including the 9th), the harmonic intensity employed was relatively low ($I_H \simeq 2 \times 10^8$ W/cm²), but instead the interaction volume ($V \simeq 5 \times 10^{-4}$ cm³) was large. In contrast, in the experiment described in [12], a high harmonic intensity ($I_9 \simeq 1.1 \times 10^{11}$ W/cm²) and a small interaction volume ($V \simeq 6 \times 10^{-8}$ cm³) were used.

6 Conclusions and discussion

In summary, we have demonstrated two-photon ionization of He-gas using the particularly broad-band XUV radiation of the superposition of the 7th to 13th harmonics. This is based primarily on the nearly quadratic dependence

of the average number of He ions produced per pulse on the total harmonic intensity, as well as on the individual harmonic intensities. The corresponding two-photon yield has been calculated and found to be essentially constant for all of the harmonics utilized in the experiment. This is of pivotal importance for an auto-correlation measurement. The estimated average number of ions per pulse was found to be within a factor of ~ 3 in agreement with the measured absolute ion number. This provides additional evidence that the ionization process is predominantly due to two-photon ionization by the harmonics in the superposition. The uncertainty of the experimentally determined average number of He ions in this experiment does not allow any conclusions on the deviation introduced by the use of the perturbation theory under these conditions (high-order harmonic generation being a non-perturbative effect). However, a more accurate measurement of the number of He ions could provide some information on the extent of the deviation from the perturbation theory. Furthermore, given that the ion yield is by a factor of 3 higher if the harmonics in the superposition add coherently, it could give some indication on whether the harmonics are phase locked or not. These results define the conditions for, and open up the prospect of, conducting second-order auto-correlation measurements of coherent superpositions of higher harmonics aiming at the unambiguous quantitative temporal characterization of attosecond pulse trains.

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