PHYSICAL REVIEW A **74**, 051402(R) (2006)

# Two-photon double ionization of rare gases by a superposition of harmonics

E. P. Benis,[1] D. Charalambidis,[1,2] T. N. Kitsopoulos,[1,3] G. D. Tsakiris,[4] and P. Tzallas[1,*]
[1]*Foundation for Research and Technology-Hellas, Institute of Electronic Structure and Laser, P.O. Box 1527, GR-71110 Heraklion (Crete), Greece*
[2]*Department of Physics, University of Crete, P.O. Box 2208, GR71003 Heraklion (Crete), Greece*
[3]*Department of Chemistry, University of Crete, P.O. Box 2208, GR71003 Heraklion (Crete), Greece*
[4]*Max-Planck-Institut für Quantenoptik, D-85748 Garching, Germany*
(Received 7 September 2006; published 15 November 2006)

Two XUV-photon double ionization of Kr and Ar by an intense superposition of higher-order harmonics of a Ti:sapphire laser is demonstrated. The logarithm of the $Kr^{2+}$ and $Ar^{2+}$ signals measured as a function of the logarithm of the XUV intensity depicts a linear dependence with a slope of $1.8\pm0.2$. Under the XUV intensities and photon energies employed, this slope value provides evidence that to the observed double ionization the direct process has a significant contribution. Applications of the above process to attosecond pulse metrology are also discussed.

DOI: 10.1103/PhysRevA.74.051402 PACS number(s): 32.80.Rm, 42.65.Ky, 42.65.Re, 42.50.Hz

Nonlinear processes in the optical regime have been extensively studied in the second half of the last century. An extension of these processes in the XUV and x-ray spectral region was until recently considered as a formidable task due to the lack of sufficiently intense short-wavelength radiation sources. In recent years, higher-order harmonic generation [1] has reached intensities high enough as to induce few-photon (mainly two-photon) ionization processes. Thus, using a selected single harmonic, few-photon single ionization [2–5], two-photon above-threshold ionization [6], and direct double ionization has been demonstrated [7]. Furthermore, the recent intensive developments in free-electron-based XUV sources opened up an alternative path to XUV nonlinear studies [8]. A key development in XUV nonlinear optics was the observation of a two-XUV-photon ionization of He by a superposition of harmonics [9]. This was a decisive step towards the temporal characterization of an attosecond pulse train through second-order autocorrelation (ac) [10–12]. In these types of experiments, the ionization threshold of He sets an upper limit for the harmonic order of the superposition to be characterized. This limitation may be overcome through alternative two-XUV-photon processes such as above-threshold ionization [12], ionic ionization, or atomic direct double ionization (DDI).

With respect to double ionization, single-photon double ionization of atoms has attracted lots of interest because it requires and thus provides information on electron-electron correlations [13]. In optical multiphoton processes double ionization does not necessarily need electron-electron correlations in order to occur. For many years multiphoton double ionization studies resulted only in what is known as sequential double ionization. Here, the atom first ionizes through single-electron ejection and subsequently the produced ion absorbs further photons to be doubly ionized. However, in the mid 1990's the pioneering experiment by Walker *et al.* [14] changed the picture. At low frequencies, direct double ionization of He has been demonstrated. Both electrons of He were ejected "simultaneously," sharing the excess energy once reaching the double continuum. Multiple ionization in strong laser fields has been extensively studied in recent years (for a review see Ref. [15] and references therein), exploiting correlated charged-particle techniques such as time-of-flight electron-ion coincidence measurements [16] and cold target recoil-ion momentum spectroscopy (COLTRIMS) [17].

At high-field frequencies, i.e., in few-photon ionization, the sequential dominates the direct process if the nonlinearity order of both processes is the same. However, work by Kornberg and Lambropoulos [18] has pointed out that in specific schemes of high-frequency ionization the direct process might become more efficient than the sequential one. This is the case when the nonlinearity of the direct process is lower than the overall nonlinearity of the sequential one, and it is the case investigated in our study.

In the present work, we demonstrate two-XUV-photon double ionization of Ar and Kr by superpositions of the 11th, 13th, and 15th harmonics of the 800 nm central wavelength Ti:sapphire laser. Under the given experimental conditions the observed double ionization can be attributed mainly to the direct process. The sequential process, although not to be completely excluded, occurs at much lower rates. Alongside of the central issue of application of such processes in attosecond pulse metrology, experimental two-XUV-photon DDI provides a test ground for numerical approaches to the three-body problem in strong field-atom interactions and might become an important tool for the experimental determination of electron-electron correlation times.

The experiment was performed utilizing a 10 Hz repetition rate Ti:sapphire laser system delivering 50 fs pulses with energy up to 150 mJ/pulse and a carrier wavelength of 800 nm. The experimental setup, shown in Fig. 1, consisted of two parts, i.e., the XUV production chamber and the detection chamber. The two parts were separated by a differential pumped region for the high-vacuum requirements of the detection chamber. An annular laser beam of 2 cm outer diameter and of 15 mJ/pulse energy was introduced into the XUV production chamber, and focused by a 3.5 m focal-length lens onto a pulsed Xe gas jet where the odd number of

*Email address: ptzallas@iesl.forth.gr

 

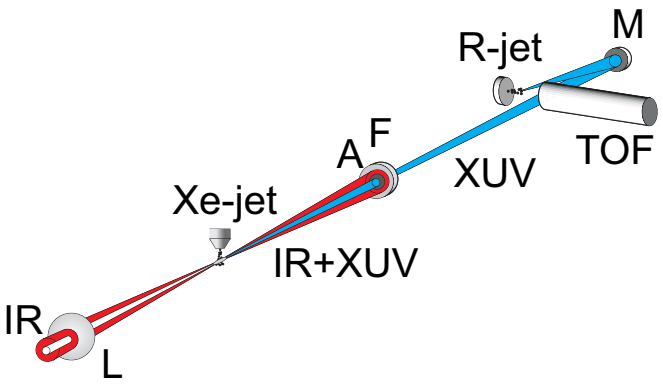


FIG. 1. (Color online) The experimental setup is shown. *L*: 3.5 m focal length lens; *A*: aperture; *F*: Sn filter; *M*: spherical mirror; TOF: time-of-flight spectrometer; R: face plate of the target jet serving as the extraction plate for the TOF spectrometer.

higher-order harmonics (HOH) were generated. The Xe target was favored for its low HOH cutoff as also for its high HOH generation efficiency. As the XUV and infrared (IR) beams were spatially expanded to the detection chamber, a 0.2-$\mu$m-thickness Sn filter supported by a 5 mm aperture allowed for selecting the 9th–15th harmonics and also for blocking the IR radiation. The resulting XUV beam was then reflected by a tungsten spherical mirror of 5 cm focal length, located about 3 m downstream from the Xe gas jet to be focused onto the target pulsed gas jet. The gas jet was electrically isolated and floating at desired voltages such that its face plate served as the repeller plate of the time-of-flight (TOF) spectrometer, as shown in Fig. 1. The ionic products were detected utilizing two spectrometers of similar TOF

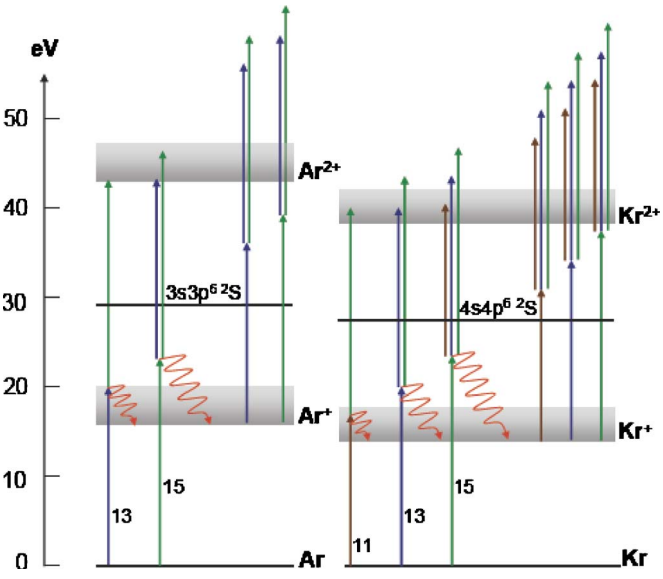


FIG. 2. (Color online) The double ionization energy level schemes for $Ar^{2+}$ and $Kr^{2+}$. The direct double ionization succeeded via two-XUV-photon absorption as also the sequential double ionization succeeded via three-XUV-photon absorption are depicted in the leftmost and rightmost sides of each scheme, respectively. The first excited states, $3s3p^6\ ^2S$ for $Ar^+$, and $4s4p^6\ ^2S$ for $Kr^+$ are also shown.

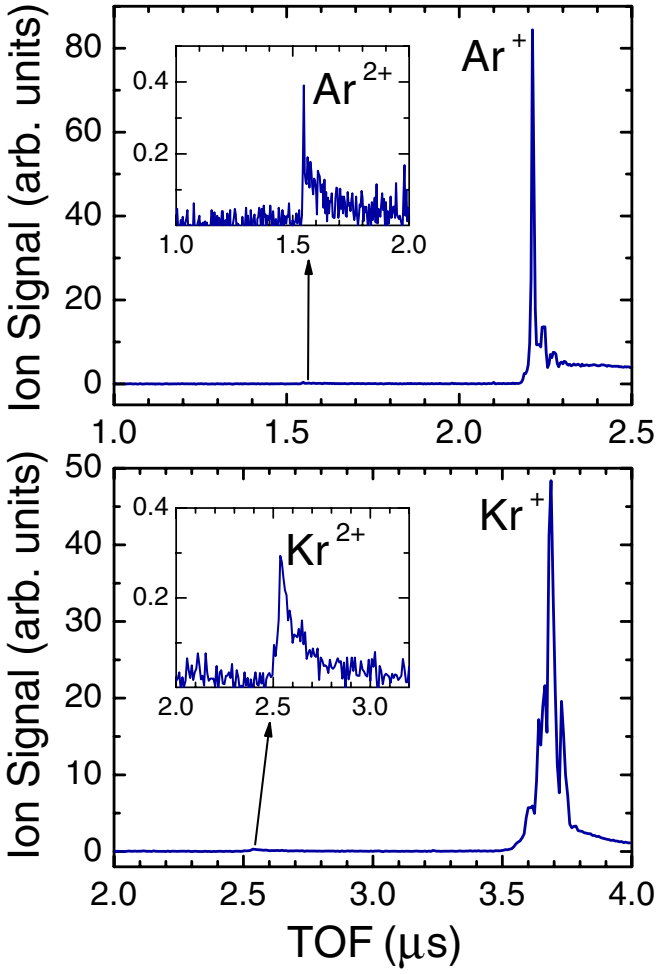


FIG. 3. (Color online) Typical TOF spectra showing the observation of Ar and Kr double ionization. The much smaller signals of $Ar^{2+}$ and $Kr^{2+}$ are shown in the insets for clarity reasons. Note that different extraction voltages have been used for Ar and Kr.

spectroscopic properties. Based on the findings of similar experimental conditions [19] the maximum XUV intensity at the interaction region was estimated to be a few times $10^{12}$ W/cm$^2$.

The ionization schemes involving two-XUV-photon direct double ionization as also three-XUV-photon sequential double ionization (SDI) are shown in Fig. 2. Although the allowed bandwidth of the Sn filter covers the 9th to the 15th harmonics, not all of them participate in the ionization process. Thus, in Ar DDI occurs through three different channels involving the 13th and 15th harmonics (13+15, 15+13, and 15+15), while in Kr it occurs through six channels involving all possible combinations of the 11th to the 15th harmonics (11+15, 13+13, 13+15, 15+11, 15+13, and 15+15). It should be noted that the maximum number of harmonics combinations that participate in the DDI is limited by the energy gap between the first and the second ionization thresholds of the target. This is because any harmonic with higher photon energy than the energy difference between the first and the second ionization thresholds will result in an overall two-photon SDI process that is much more efficient and hence will dominate over the DDI. In this case, the tar-

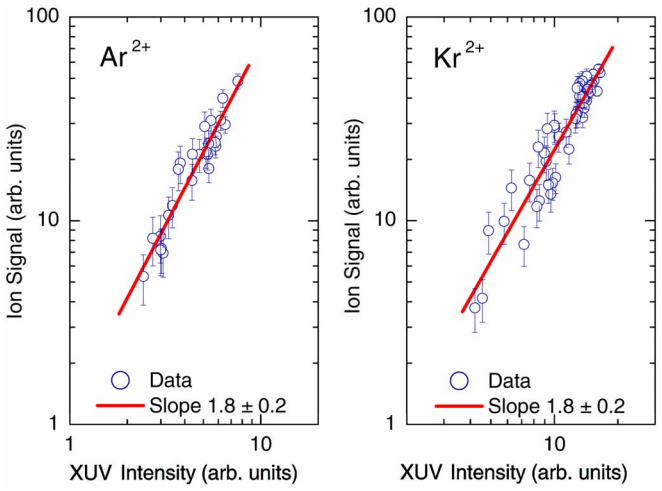


FIG. 4. (Color online) $Ar^{2+}$ and $Kr^{2+}$ ion signals obtained as a function of the XUV intensity. The slope of 1.8±0.2 for both of them ascertains the second-order nonlinearity of the ionization process.

geted observation of the DDI process would be unattainable. Thus, in Ar, DDI is restricted in a bandwidth of harmonics ranging from 11th to 17th, while in Kr it is restricted from 11th to 15th. The presence of more channels in Kr in this work is due to the selected bandwidth by the Sn filter. It should be noted that single-photon absorption from the atomic ground state is above the singly charged ionic ground state but below any singly charged ionic excited state of both atoms. Indeed, single-photon ionization populates only the ground states of the $Kr^+$ and $Ar^+$ ions, respectively, requiring absorption of at least two more photons for the double ionization. Thus, SDI occurs only through the ground ionic states and is a three-photon process for both atoms investigated. Such a scheme favors DDI over SDI since it requires less number of photons, i.e., a lower nonlinearity degree as mentioned earlier. DDI is expected to dominate over SDI for conditions of insignificant saturation of ionic states, i.e., at low XUV intensities, while the picture is inverted when saturation becomes appreciable, i.e., at high XUV intensities [20].

Typical TOF spectra depicting the observation of Ar and Kr double ionization are shown in Fig. 3. The much smaller signals of $Ar^{2+}$ and $Kr^{2+}$ are shown in the insets for clarity reasons. Note that the isotopes of Kr cannot be discerned in the $Kr^{2+}$ signal most probably due to the low-signal statistics. The large background on the right side of the $Ar^+$ peak is due to the single-photon ionization from the incoming beam. This effect has been substantially reduced in the Kr experiment after introducing a small deviation from the normal incidence into the reflected from the spherical mirror XUV beam ($\sim 2°$), and accordingly modifying the TOF spectrometer, so that the incoming beam does not overlap with the gas jet. In order to confirm the nonlinearity of the double ionization process, the XUV power dependence of the $Ar^{2+}$ and $Kr^{2+}$ signals was studied as a function of the total XUV intensity (Fig. 4). The variation of the XUV intensity was succeeded by changing the target pressure seen by the laser pulse. The pressure was varied in a controlled way by varying the delay time between the opening of the jet and the laser pulse. Thus, different delay times result in different gas pressures during the interaction, and therefore to different XUV intensities. The XUV intensity was considered proportional to the $Ar^+$ signal due to the linear dependence of the single-ionization process. The slopes are 1.8±0.2 for both targets indicating the dominance of the two-photon process.

Before we discuss the observation of DDI we need to examine the nonlinearity observation more thoroughly. Let us reiterate here that there is no intensity region below the single-ionization saturation intensity in which the three-photon SDI and the two-photon DDI processes have comparable rates, that would have resulted in a slope larger than 2. A slope of 2 is also expected for SDI at high XUV intensities, that is, at single-ionization saturation conditions. At intensities approaching the saturation value, both processes contribute substantially, thus rendering a determination of a slope in this region rather meaningless [20]. Indeed, according to Ref. [20], the different rates and saturation intensities of the DDI and SDI processes result in a small "knee" between the two regions in which DDI or SDI becomes dominant. However, the statistics of the experiment do not allow for the observation of such a tiny "knee" feature. Based on similar experimental conditions of our previous works [10,19] we estimate that the intensities are in the range of $10^{11}$ W/cm$^2$ to a few times $10^{12}$ W/cm$^2$, allowing for the single-photon single ionization to approach a nonnegligible degree of saturation at the higher intensities. Therefore SDI possibly becomes of some importance. In more detail the DDI rate can be expressed as

$$\frac{dP_{dd}}{dt} = \sigma_{dd}^{(2)} \left( \frac{I_{XUV}}{\hbar\omega} \right)^2, \tag{1}$$

where $\sigma_{dd}^{(2)}$ is the two-photon generalized cross section, $I_{XUV}$ the intensity of the XUV radiation, and $\hbar\omega$ the photon energy. In conditions far from saturation, the three-photon SDI rate may be written as the product of the ionic ground-state population times the ionic two-photon ionization rate, i.e.,

$$\frac{dP_{sd}}{dt} = \sigma \frac{I_{XUV}}{\hbar\omega} \Delta t \sigma_i^{(2)} \left( \frac{I_{XUV}}{\hbar\omega} \right)^2. \tag{2}$$

$\sigma$ is the single-photon ionization cross section of the atom, $\sigma_i^{(2)}$ the two-photon SDI generalized cross section of the ion, and $\Delta t \simeq 30$ fs the duration of the XUV pulse (in this rough estimation we ignore any temporal substructure, like attosecond bursts). Thus, the ratio of SDI to DDI rate yields

$$\frac{dP_{sd}}{dt} \Big/ \frac{dP_{dd}}{dt} \simeq \sigma \frac{\sigma_i^{(2)}}{\sigma_{dd}^{(2)}} \frac{I_{XUV}}{\hbar\omega} \Delta t. \tag{3}$$

In the above formula, $\sigma$ is known quite accurately for rare gases, ranging from 20 to 40 Mb for the involved XUV energies [21]. However, to our knowledge, there are no data available for $\sigma_{dd}^{(2)}$ and $\sigma_i^{(2)}$ for rare gases other than He. Calculations for He under similar ionization conditions [20] indicate that $\sigma_{dd}^{(2)}$ is about an order of magnitude higher than $\sigma_i^{(2)}$, while a value closer to that of $\sigma_i^{(2)}$ cannot be safely excluded. Under the expectation of a similar behavior of the

DDI and SDI in the present experimental conditions, and for intensities between $10^{11}$ W/cm$^2$ and $10^{12}$ W/cm$^2$, the ratio of the SDI to DDI rate is between 0.3 and 3%. Even in the "extreme" case where $\sigma_{dd}^{(2)}=\sigma_i^{(2)}$, the SDI to DDI rate ranges between 3 and 30%. The above rough estimation indicates that the observed double ionization can be assigned primarily to the DDI process except maybe from the higher intensities where SDI is expected to contribute appreciably.

As mentioned earlier, DDI is a candidate process for use in attosecond metrology. The appropriateness of a second-order process for attosecond metrology lies on the requirements of the flatness of the spectral response and of the "instantaneous" temporal response [11]. However, there are no available experimental data or theoretical calculations as in the case of He [10–12]. Experimentally, second-order ac measurements using Ar or Kr could be cross examined against the available He measurements. The above is true for the range of XUV intensities where DDI surpasses SDI. At much higher intensities, where DDI is negligible, SDI is also a second-order process and could be used in attosecond metrology by means of second-order ac as well.

In conclusion, two-XUV-photon double ionization of Kr and Ar by an intense superposition of higher-order harmonics of a Ti:sapphire laser has been observed for XUV intensities spanning one order of magnitude. DDI has been attributed as the main mechanism for the measured double ionization. The competing mechanism of SDI cannot be unambiguously excluded at the higher XUV intensities employed in the experiment. With respect to the exploitation of double ionization in pulse-duration metrology based on second-order ac measurements it is worth noting that both the direct two-photon, as well as the sequential three-photon double ionization for which the single-photon–single-ionization step is saturated, are appropriate schemes.

This work was supported in part by the European Community's Human Potential Programme under Contract Nos. MRTN-CT-2003-505138 (XTRA) and MTKD-CT-2004-517145 X-HOMES and the Ultraviolet Laser Facility (ULF) operating at FORTH-IESL (Contract No. HPRI-CT-2001-00139).

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