Photoionization in combined ultra short XUV and infrared laser pulses

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Abstract. Multi-photon-double ionization of xenon by Ti:Sapphire laser pulses combined with their 25th harmonic has been studied by means of a momentum imaging spectrometer. The determination of the momenta of the emitted photoelectron pair and of its energy and angular correlation gives insight into the various mechanisms leading to double ionization. Although the conditions for non sequential direct multi-photon double ionization are met in the experiment, it is found that two-step sequential processes prevail.

1. Introduction

Photoionization is a fundamental quantum physics process and one of the basic tools giving access to structural and dynamical properties of atoms. Depending on the light frequency and intensity, it can result in the ejection of two valence shell electrons from a variety of mechanisms. (i) Following the absorption of a single high-energy photon: Photo Double Ionization (DI) is governed by electron correlation. It is most intensively studied for that property [1; 2]. (ii) Through multiphoton absorption: DI can occur in a low frequency, intense \((10^{12} W/cm^2)\) laser beam. Multiphoton DI (MPDI) was discovered in the mid-seventies [3] and investigated by several groups [4]. (iii) In an electric field mediated three-step process at even higher light intensity \((10^{14} W/cm^2)\) [5]. This “strong field” DI was the object of tremendous interest during the past decade [6] and the tag “non-sequential DI”, as opposed to a sequence of two independent field ionization processes, is attached to it. Despite a long history, little is known about low-order N-photon DI. Experimental constraints restricted most studies to orders \(N \geq 8\) where a perturbation analysis is already intractable [4]. Although laser advances [high order harmonic generation (HHG) and free electron lasers (FEL)] have stimulated new theoretical [7; 8] and experimental [9; 10] studies, two-photon DI experiments have been limited so far to the investigation of total ionization yields [9; 10]. Only recently the analysis of the sum-momentum of the photoelectron pair became possible using COLTRIMS (Cold Target Recoil Ion Momentum Imaging).
Spectroscopy) [11]. Nevertheless, a complete kinematical analysis of two-photon DI of helium, the basic two-electron system, in particular seems to become feasible in the near future, as intense XUV sources with high enough repetition rate become available [11]. This will undoubtedly be a considerable breakthrough and will provide new insights into the underlying three-body problem. Meanwhile, the superposition of XUV and IR laser pulses which can either be achieved with synchrotron [12], or high order harmonic sources, approaches this goal. Combining the latter technique with a complete momentum analysis of the two emitted photoelectrons is the object of the work presented here [13; 14].

Xenon is chosen for the present MPDI study because of its relatively low ionization potentials, which are well adapted to the possibilities of the high harmonic source we used. The intensity of the harmonic source allows us to study DI pathways which involve the absorption of one XUV photon and of one or several infrared photons. The large Xe mass limits the capabilities of a reaction microscope to derive the individual momenta of the DI photoelectron pair from a measurement of the Xe$^{++}$ ion recoil momentum and of the momentum of one of the photoelectrons [13]. For this system it is thus necessary to measure directly the momenta of both photoelectrons in order to arrive at kinematically complete results [14].

2. The double ionization pathways
The possible MPDI pathways for xenon turn out to be quite complex. One reason for the complexity is the presence of five DI thresholds lying quite close together. This is schematically shown in figure 1. The MPDI pathways are falling into two categories, “non-sequential” and “sequential”. The non-sequential direct DI mechanism (Fig. 1a) results in the simultaneous emission of two electrons with a continuous distribution of their kinetic energy only subject to the constraint:

$$E_{\text{tot}}^\alpha = \hbar \omega_{\text{XUV}} + n\hbar \omega_{\text{IR}} - I_\alpha^{++} = E_1^\alpha + E_2^\alpha$$

where $E_{\text{tot}}^\alpha$ is the total and the $E_i^\alpha$ ($i = 1, 2$) are the individual photoelectron kinetic energies. $I_\alpha^{++}$ is the DI threshold with Xe$^{++}$ left in state $\alpha$ and $n$ is the number of absorbed infrared photons. Only one XUV photon is assumed to be absorbed. Non-sequential indirect MPDI is a stepwise process: a first electron is emitted and a second one simultaneously excited to a
Figure 2. Schematic view of the experimental setup.

Figure 3. Pathways to DI of Xe when the XUV photon energy is higher than the DI threshold. $s$ denotes sequential, $nsd$ non-sequential direct and $nsi$ non-sequential indirect MPDI.

Rydberg state of Xe$^+$ above the DI threshold which finally autoionizes with some delay. The kinetic energy distribution is discrete and one component ($E_2$ in figure 1a) does not depend on the energies of the absorbed photons. On the “sequential” route to DI (Fig. 1b) typically first one short wavelength photon is absorbed ending up with single ionization of Xe and Xe$^+$ left behind in a possibly excited state below the DI threshold. Double ionization is then reached after further absorption of at least one infrared photon. The possible kinetic energies of the photoelectrons are then further restricted by:

$$E_1 = \hbar \omega_{XUV} + n_1 \hbar \omega_{IR} - I^{++}$$

(2)

$$E_2^\alpha = n_2 \hbar \omega_{IR} + I^{++} - I_{\alpha}^{++}$$

(3)

Here $I^{++}$ is the Xe ionization potential with the singly charged ion possibly left in an excited state. The number $n_1$ of IR photons absorbed in the first step is usually zero.

3. Experimental setup

The scheme of the setup used to achieve a complete kinematical analysis of the DI final state (the momenta of both photoelectrons) is shown in figure 2 [14]. The laser facility PLFA (SLIC, France) delivering 10 mJ pulses at 812 nm, with a width of 35 fs (FWHM) at 1 kHz repetition rate, was used for the experiment. The beam was split into a main (8 mJ) IR1, and a dressing (2 mJ) IR2 branch. The IR1 beam was focused into an argon gas cell to generate the XUV harmonics. Passing the beam through an aluminium filter removed low harmonics and the infrared radiation. The XUV light was then reflected and filtered by a mirror pair consisting of a plane and a spherical multilayer mirror designed to reflect the 25th harmonic. The spherical mirror focussed the beam through a drilled plane mirror to a spot size of about 100 $\mu$m within the spectrometer. The reflectivity of the multilayer mirrors was typically 20% for the 25th harmonic. In each case harmonics of higher order were almost completely rejected. Adjacent
harmonics of lower order were observable, but at a lower intensity level. The XUV signal was monitored by means of a removable photomultiplier. The number of harmonic photons on target was estimated to be $5 \times 10^5 - 5 \times 10^6$ photons per pulse.

The IR2 laser pulses were delayed with sub-fs accuracy with respect to the XUV pulses. The beam intensity was adjusted by means of a half-wave plate followed by a polarizer. Focusing was done with a lens with 1500 mm focal length (numerical aperture). An annular part of the IR2 beam was reflected into the spectrometer by the drilled mirror which superimposed the IR2 on the XUV beam. The spatial overlap of the XUV and IR2 beams was achieved by projecting the IR1 and IR2 beams on a CCD camera after removal of the Al filter in the XUV beam path. The overlap of the pulses in time was adjusted by observing the side-band peaks in the photoionization spectrum of helium [15]. IR2 pulse energies typically in the range $50 - 100 \mu J$ were used, giving rise to light intensities in the $10^{12} - 10^{13} W/cm^2$ range within the focal spot in the spectrometer. The diameter of the focal spot was 100 – 150 \(\mu m\).

The momentum imaging spectrometer CIEL2, based on a previous one built for synchrotron experiments [16], was optimized for the present experiment. Electrons are detected by a pixel detector similar to that already described in [17] with important improvements incorporated. The gas jet is emitted from a 20 \(\mu m\) nozzle and shaped by two skimmers, with a resulting beam diameter of 1 \(mm\) at the point where the jet and the light beams intersect (12 \(cm\) downstream from the nozzle). The background gas pressure in the spectrometer is about 2 \(Torr\). Homogeneous magnetic (typically 5 Gauss) and electric fields (4 \(V/cm\)) guide the photoelectrons and ions to their respective detectors. The ion detector consists of a stack of three micro channel plates (MCP, diameter 40 \(mm\)) equipped with a conventional resistive anode for position encoding. For each ion impinging on the detector its time of flight and the position where it hit is determined. The photoelectron pairs from DI events are detected by a stack of two MCP’s (diameter 80 \(mm\)) followed by a pixel anode with 64 detection lines along each (x and y) direction. The readout of this detector is done by a fast multi-hit TDC with 128 inputs. Each TDC channel has a time resolution of 100 ps. It is triggered in the "stop" mode within a time window centred on the time of flight of doubly charged Xe ions. Thus, single ionization is discarded and only triple Xe\(^{++}\)-electron-electron coincident events are recorded, at a rate of typically a few times \(10^{-1} Hz\). The momenta of the two photoelectrons are accurately determined by deconvoluting their trajectories. With the time resolution indicated above, and the fields applied, electrons with kinetic energies in the [0, 6eV] range can be measured, with typical angular and energy resolutions of $\Delta \vartheta \approx 5$ deg and $\Delta E/E \approx 0.15$, respectively.

4. Results and discussion

Here we present the MPDI results for the 25\(th\) harmonic (\(\hbar \omega_{XUV} = 38.17 eV\)) applied together with the IR pulses (\(\hbar \omega_{IR} = 1.53 eV\)). The 25\(th\) harmonic already by itself causes double ionization of Xe (DI threshold: 33.10 eV). At this XUV photon energy all possible MPDI pathways are open as figure 3 shows and all low lying Xe\(^{++}\) states are accessible already after absorption of the XUV photon.

Insight into the prominent MPDI pathways is gained by scanning the delay of the IR with respect to the XUV pulses and measuring the yield of Xe\(^{++}\) ions. The result is shown in figure 4. Negative delay times mean the IR is ahead of the XUV pulse. A step like increase in the signal is observed when the pulses start to overlap in time (\(\tau = 0\)). For the whole delay range with \(\tau > 0\) the Xe\(^{++}\) yield then stays constant on the elevated level. For \(\tau < 0\) the origin of the non-zero Xe\(^{++}\) ion yield is DI after absorption of one XUV photon (see figure 3). This DI pathway contributes also to the Xe\(^{++}\) yield for \(\tau > 0\). The increase of the yield near \(\tau = 0\) is due to starting MPDI. The constancy of the yield on the elevated level when the IR follows the XUV pulse indicates that MPDI follows prominently a step wise pathway with long lived intermediate states (lifetime considerably longer than the maximum delay [13]). The relevant
intermediate states are excited \((5s)^2(5p)^4(\text{nl})^1\) Xe\(^+\) states lying above the Xe\(^+\) \((5s)^1(5p)^6\text{^2S}_{1/2}\) state but below the DI threshold [13; 14]. These states are reached by absorption of one XUV photon (the “s” pathway in figure 3). It promotes one electron to the continuum while a second one is simultaneously excited via electron-electron correlation. Direct MPDI pathways are not identified in this pump-probe experiment. They would only contribute to MPDI while the IR and XUV pulses overlap in time, that is for a delay interval of less than \(\approx \pm 50\) fs around \(\tau = 0\) in addition to the step wise routes. For \(\hbar \omega < I^{++}\) (\(I^{++}\) the DI threshold) this should be observable as a local enhancement in the Xe\(^{++}\) ion yield. However, such an enhancement may not exist in the present situation since direct absorption of one XUV and an IR photon would, similar to single ionization, probably not change the total DI probability but only redistribute electrons in the continuum (see for example [15]). Differential photoelectron detection schemes should be able to shed more light on the possible presence of direct routes in MPDI of Xe for \(\hbar \omega > I^{++}\).

Deeper insight into the PDI mechanisms is gained by the 2D plot of the kinetic energy correlation \(f(E_1, E_2)\) of the two photoelectrons in figure 5, and by the distribution

\[
f_{\text{tot}}(E_{\text{tot}}) = \int_0^{E_{\text{tot}}} dE_1 \ f(E_1, E_{\text{tot}} - E_1)
\]

of the total energy in figure 6. Lines at \(E_{\text{tot}} = 1.7, 3.5, 7.1, 10.1\) eV have been drawn in both figures. They separate the contributions of the \(^1\text{S}\) state at \(E_{\text{tot}} = 0.6\) eV from the \(^1\text{D}\) state at \(E_{\text{tot}} = 2.98\) eV and from the \(^3\text{P}_{0,1,2}\) states at 3.89, 4.09 and 5.15 eV to the spectra, respectively. \(f_{\text{tot}}(E_{\text{tot}})\) shows that total energies higher than 7.1 eV become accessible to the photoelectron pair. Beyond this energy, no photoelectron pairs from DI by absorption of one XUV photon alone can be found. Solely multi-photon DI routes contribute to the photoelectron yield in the \([7.1, 10]\) eV total energy range, such as those shown in figure 3. To simplify matters, we only included representative pathways in figure 3 with one infrared photon absorbed on the non-sequential routes to DI, and two infrared photons absorbed on the sequential ones. However, more infrared photons may be involved in both cases. The total energy regime where MPDI is isolated from other DI pathways corresponds to the region labelled “4” in the electron kinetic energy correlation plot (figure 5).

Here we want to focus on this regime only. A detailed discussion of the mixed regimes
Figure 6. Total energy distribution $E_1 + E_2$ for the photoelectron pair from MPDI by IR together with the 25th harmonic radiation. The black arrows indicate the maximum total pair energy one expects after DI by the 25th harmonic alone when Xe$^{++}$ is left in the denoted state.

Figure 7. Relative emission angle $\theta_{12}$ distribution of the photoelectron pair for events from region “4” of figure 5.

can be found in [14]. Interestingly, the region labelled “4” in figure 5 (with $E_{tot}$ in the interval [7.1, 10] eV), appears to be populated only close to the $E_1$ and $E_2$ axes, respectively. This means that strongly asymmetric energy sharing is favoured. It is worth studying the angular correlation of the two emitted electrons. The statistics of the present experiment does not yet allow a full differential cross section to be extracted from the data by fixing the energy and direction of emission of one of the emitted electrons and plotting the emission diagram of the second one in various planes. However, for each PDI event the angle $\theta_{12}$ enclosed by the momentum vectors of the two photoelectrons can be calculated, and it is instructive to look at the histogram of this angle over the whole ensemble of events for region “4” in figure 5. The result is shown in figure 7, where $\theta_{12}$ ranges from 0.5 to 3 radians. The histogram has been weighted by the factor $1/\sin\theta_{12}$, to eliminate the solid angle effect well known in momentum imaging experiments. Note that the statistical error bars increase towards $\theta_{12} = 3$ rad., where the solid angle becomes small. The angular correlation histogram is rather flat with no specific $\theta_{12}$ favoured. This fact, together with the strongly asymmetric kinetic energy sharing of the photoelectron pair (figure 5) in regime “4” points at sequential MPDI to prevail. Two possible processes can account for the findings. The first one is non-sequential indirect “nsi” involving Xe$^{++}$ states which autoionize yielding Xe$^{+}$ (5p)$^4$ 3P$_2$. This scenario would result in a low kinetic energy electron from the second step, not correlated with the high energy one from the first step. The second scenario is a sequential “s” process via Xe$^{+}$ states lying below the double ionization threshold. Spectroscopic data [18] show that there exist many of these states with binding energies around 29 eV together with the Xe$^+$ (5p)$^4$ 1D 5d $^2S_{1/2}$ state at 28.88 eV which is involved in sequential PDI induced by the 19th harmonic together with the absorption of three infrared photons in the second step (see [14]). The same route is possible with the 25th harmonic, but now with a much higher energy photoelectron in the first step. Additionally, also in the second scenario, Xe$^{+}$ states at higher binding energy (above 30 eV) may be responsible for sequential MPDI, with only two infrared photons absorbed in the second step (this case is illustrated in figure 3). Only an improvement of the statistics and energy resolution will allow one to disentangle all these routes and to determine their respective contributions to MPDI in region “4” of the kinetic energy correlation plot (figure 5).
5. Summary
The present work demonstrates that complete measurements on PDI, where the momentum vectors of both outgoing photoelectrons are determined, can be achieved with high harmonic light sources, running at a repetition rate of 1 kHz. The signal to noise ratio in the coincidence mode, estimated to be around 20, can be further improved in forthcoming experiments by decreasing the background pressure below $2 \times 10^{-10}$ Torr. With infrared and harmonic laser pulses combined, a representative MPDI mechanism has been analysed for Xe (a more complete analysis is given in [14]). Since two-step MPDI appears to be dominant, the present study suggests repeating this experiment on lighter atoms, either with XUV plus infrared or visible photons, or with two XUV photons. With these lighter atoms MPDI is expected to be simpler to analyse. For example, it is well known that non sequential direct DI induced by absorption of two equal energy XUV photons can be isolated for helium. The rapid progress in the development of harmonic and X-FEL sources is likely to make these experiments possible in the near future.

References
[1] Briggs J S and Schmidt V 2000 J. Phys. B: At. Mol. Opt. Phys. 33 R1
[2] Avaldi L and Huetz A 2005 J. Phys. B: At. Mol. Opt. Phys. 38 5861
[3] Suran V V and Zapesochnyi I P 1975 Sov. Techn. Phys. Lett. (Engl. Transl.) 1 420
[4] Lambropoulos P, Maragakis P and Zhang J 1998 Phys. Rep. 305 203
[5] Corkum P B 1993 Phys. Rev. Lett. 71 1994
[6] Becker A and Faisal F H M 2005 J. Phys. B: At. Mol. Opt. Phys. 38 R1
[7] Kornberg A M and Lambropoulos P 1999 J. Phys. B: At. Mol. Opt. Phys. 32 L603
[8] Colgan J and Pindzola M S 2002 Phys. Rev. Lett. 88 173002
[9] Nabekawa Y, Hasegawa H, Takahashi E J and Midorikawa K 2005 Phys. Rev. Lett. 94 043001
[10] Benis E P, Charalambidis D, Kitsopoulos T N, Tsakiris G D and Tzallas P 2006 Phys. Rev. A 74 051402
[11] Moshammer R, Jiang Y H, Foucar L, Rudenko A, Ergler T, Schröter C D, Ldemann S, Zrost K, Fischer D, Titze J, Jahnke T, Schöffler M, Weber T, Dörner R, Zouros T J M, Dorn A, Ferger T, Kühnel K U, Disterer S, Treusch R, Radcliffe P, Plönjes E and Ullrich J 2007 Phys. Rev. Lett. 98 203001
[12] Aloise S, O’Keefe P, Cubaynes D, Meyer M and Grum-Grzhimalo A N 2005 Phys. Rev. Lett. 94 223002
[13] Böttcher M, Rottke H, Zhavoronkov N, Sandner W, Agostini P, Gisselbrecht M and Huetz A 2007 Phys. Rev. A 75 033408
[14] Guyénotand O, Gisselbrecht M, Huetz A, Agostini P, Carré B, Breger P, Gobert O, Garzella D, Hergott J F, Tcherbakoff O, Merdji H, Bougeard M, Rottke H, Böttcher M, Ansari Z, Antoine P and DiMauro L F 2008 J. Phys. B: At. Mol. Opt. Phys. 41 065601
[15] Guyénotand O, Gisselbrecht M, Huetz A, Agostini P, Tăueb R, Maquet A, Carré B, Breger P, Gobert O, Garzella D, Hergott J F, Tcherbakoff O, Merdji H, Bougeard M, Rottke H, Böttcher M, Ansari Z and Antoine P 2008 J. Phys. B: At. Mol. Opt. Phys. 41 051002
[16] Gisselbrecht M, Huetz A, Lavollée M, Reddish T J and Secombe D P 2005 Rev. Sci. Instrum. 76 013105
[17] Lavollée M 1999 Rev. Sci. Instrum. 70 2968
[18] Kikas A, Osborne S J, Ausmees A, Svensson S, Sairanen O P and Aksela S 1996 J. Electron Spectrosc. Relat. Phenom. 77 24