## Direct and sequential two-photon double ionization at FLASH

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32.80.Rm, 41.60.Cr, 42.65.-k

## Abstract:

Recoil-ion momentum distributions for two-photon ( $\hbar\omega_0 \approx 44eV$ ) double ionization of He and Ne have been recorded with a reaction microscope at FLASH at an intensity of ~1·10<sup>14</sup> W/cm<sup>2</sup> exploring the dynamics the two fundamental two-photon – two-electron reaction pathways, namely sequential and direct (or non-sequential) absorption of the photons. We find strong differences in the recoil-ion momentum patterns for the two mechanisms pointing to the significantly different two-electron emission dynamics and, thus, provide serious constraints for theoretical models.

Since Einstein's revolutionary explanation of the photoelectric effect in 1905 [1], the break-up of bound systems as a result of their interaction with single light-quanta, the photons, has remained in the very focus of interest in experimental and theoretical physics as well as in chemistry and biology as one of the most fundamental reactions occurring in nature.

Whenever there is more than one electron actively involved in the photo-absorption process, however, one faces serious problems in calculations as well as in measurements, even if only a single photon was absorbed at a time. Thus, the simplest situation where two electrons emerge from the helium atom has numerically been solved only within the last decade where fully differential experimental cross sections have become available (for a review see [2]). The four-particle problem, double ionization of hydrogen (deuterium) molecules, has resisted any comprehensive numerical solution until most recently (see [3] and references therein) where fully differential data as e.g. obtained in [4] were successfully described on an absolute scale. Keeping the simple He target but increasing the number of photons instead, as e.g. in strong-field double ionization at optical frequencies needing more than 50 quanta, still represents a serious challenge for computations (see e.g. [5] - [7] and references therein). Likewise, kinematically complete experiments have only been reported within the last two months [8,9].

In this letter we report on the first differential measurement, recoil-ion momentum distributions, for the most basic non-linear two-electron light-matter interaction, where two VUV photons (44 eV each) "simultaneously" remove two electrons from He. The results are compared to double ionization of neon, where a sequential, step-wise absorption pathway with intermediate relaxation to a bound state of the Ne<sup>+</sup> ion is energetically allowed. Vastly different momentum distributions are observed for both reactions and compared with theoretical predictions. Since the measured recoil-ion momentum spectra reflect the sum-

momentum distributions of the emitted electrons and, thus, provide first information about the relative emission angles and the energy sharing between both electrons for different nonlinear processes, the data provide stringent test-grounds for theoretical models. The experiments became feasible by exploiting a unique combination of modern multi-particle momentum imaging techniques, "reaction microscopes" [10], and a novel light source, the *F*ree electron *LAS*er in *H*amburg (FLASH), delivering ~25 fs VUV light pulses at unprecedented intensities (here ~1.10<sup>14</sup> W/cm<sup>2</sup>), orders of magnitude more intense than thirdgeneration synchrotrons or state-of-the-art high harmonic pulses from optical lasers (HHG, [11,12]).

Due to its fundamental character two-photon double ionization (TPDI) of He has received enormous theoretical attention (for an incomplete collection see e.g. [13-23]). Surprisingly, even for the moderate intensities investigated, where second-order perturbative approaches are expected to be valid, the calculated total cross-sections for this reaction ( $\sigma_{TPDI}$ ) vary over more than one order of magnitude [15]. Two measurements of  $\sigma_{TPDI}$  have been recently performed for photon energies of 41.8 [11,12] and 42.8 eV [24] employing HHG and FLASH radiation, respectively. At least the latter however, does not yet represent a stringent test of theory since neither the pulse profile nor the pulse length are determined experimentally and, thus, the estimated intensity usually exhibits large error bars.

The photon energy range between 40-54 eV is especially attractive since here the  $2^{nd}$  ionization potential, i.e. of He<sup>+</sup> (1s), is larger than the energy of a single photon and, thus, the so-called 'sequential' mechanism, where the electrons are emitted independently with intermediate relaxation into a bound state of the singly-charged ion, requires at least three photons, whereas the two-photon sum energy is still large enough to promote both electrons into the continuum, which than requires a direct ('non-sequential' (NS)) process. Both of the earlier experiments [11,12,24] revealed the dominance of the two-photon NS channel for

TPDI at light intensities up to  $10^{14}$  W/cm<sup>2</sup> reflected in the quadratic intensity dependence of the doubly-charged ion yield. However, as it was found previously for the single-photon and the strong-field regimes, measurements of the total cross-sections do not allow one to draw a definite conclusion concerning the physical mechanisms underlying the few-electron ionization dynamics, and, thus, differential data are required.

Unfortunately, He, being the "simplest" atom for the theoretical description is, at the same time, the most challenging one for experimental investigations. This is due to its huge two-electron ionization potential resulting in extremely low double-ionization rates and setting severe vacuum requirements for coincidence studies or recoil-ion momentum spectroscopy [18]. To the best of our knowledge, the only differential data for any transitions in He has been presented in [25]. There, a feature occurring around 24 eV to 26 eV in the photoelectron spectrum of He irradiated by intense FLASH radiation of 38.5 eV was interpreted to result from subsequent processes involving as a first step resonant two-photon absorption into the doubly excited state of even parity.

Our present measurements were performed using an experimental setup similar to the one described in detail before [26]. Briefly, the reaction microscope spectrometer [10] was installed in the focus of the FLASH beam line BL2. Linearly polarized ~25 fs FEL radiation was focused onto a 30  $\mu$ m spot within the collimated supersonic gas jet in the centre of the ultra-high vacuum chamber. In order to enable the experiment on TPDI of He, the base pressure had to be reduced below 8·10<sup>12</sup> mbar. Created ions and electrons were guided to position-sensitive channel plate detectors by weak electric (1 V/cm) and magnetic (30 Gauss) fields. From the measured times-of-flight and positions on the detectors the full momentum vectors of emitted ions and electrons were calculated. Whereas for Ne we thus were able to record fully differential data determining the complete momenta of all emitted particles, two electrons and the doubly charged ion (subject of a forthcoming paper), this was not feasible

for He. Here, (i) electrons from the rest gas dominated the yield on the detector even at the very low background pressure achieved and (ii) the total number of He<sup>2+</sup> events recorded during the available beam time was still too small to extract higher differential data due to the exceedingly small  $\sigma_{TPDI}$ . The laser polarization was parallel to the direction of the gas jet propagation and the pulse energy was monitored shot by shot using a Faraday cup. For the averaged value of ~ 20 µJ per pulse the estimated peak intensity corresponded to ~1.10<sup>14</sup> W/cm<sup>2</sup> keeping in mind the caveats mentioned before. Special care was taken in order to avoid the contributions of higher harmonics of the FEL radiations, which were suppressed on a level of < 10<sup>-5</sup> by using specially designed filters.

Fig. 1a depicts the measured two-dimensional momentum distribution of singly-charged He ions (shown in the plane defined by the jet propagation direction and the spectrometer axis). The data are integrated over the third momentum component (parallel to the FEL beam propagation). Since the momentum carried by the photons is negligibly small, this spectrum represents the mirror image of the corresponding photoelectron momentum distribution. The data exhibit the well-known, characteristic dipole structure along the polarization direction, which is clearly resolved. No events due to the two-photon above-threshold single ionization have been observed.

The measured He<sup>2+</sup> momentum distribution is presented in Fig. 1b. In contrast to the single-ionization spectrum, as well as to double-ionization induced by a single photon with similar energy above the threshold [27], but in qualitative agreement with our earlier results for Ne [26], it displays a clear maximum at the origin, with most of the events lying well within the inner circle marking the maximum He<sup>2+</sup> momentum for the case where one electron would have taken all the energy. Since for double ionization the momentum of the doubly-charged ion is balanced by the sum-momentum of both emitted electrons, the dominance of the events with very low ion momenta indicates that both electrons are

preferentially emitted back-to-back into opposite hemispheres with similar energies, thus, compensating the momenta of each other. This is in striking contrast with the dynamics found in single-photon double ionization, where exact back-to-back emission along the polarization direction is forbidden by the dipole selection rules. Thus, since this limitation is lifted for the case of TPDI, the difference between the spectra for these two processes might readily be understood qualitatively. Here it should be noted that according to the results of [11,12,24], the contribution of the three-photon sequential process is expected to be negligible under our experimental condition.

In order to come to a more quantitative understanding, the He<sup>2+</sup> momentum distribution has been calculated for  $\hbar \omega_0 = 44 eV$  and is shown in Fig. 2a. The transition amplitude for the two-electron ionization caused by the absorption of two photons from the laser field is given by (atomic units are used):

$$a_{fi}^{(2)} = \frac{\pi}{2i} \delta(\varepsilon_f - \varepsilon_i - 2\omega_0) \cdot \sum_m \frac{\left\langle \varphi_f \middle| \hat{W}_0 \middle| \varphi_m \right\rangle \left\langle \varphi_m \middle| \hat{W}_0 \middle| \varphi_i \right\rangle}{\varepsilon_m - \varepsilon_0 - \omega_0 + i\alpha}, \quad \text{with} \quad \hat{W}_0 = (\vec{r}_1 + \vec{r}_2) \cdot \vec{F}_0.$$
(1)

Here,  $\varphi_i$  and  $\varphi_f$  are the initial and final states of the electrons, respectively,  $\vec{F}_0$  is the amplitude of the laser field strength,  $\omega_0$  is the field frequency and  $\varepsilon_f$ ,  $\varepsilon_i$ ,  $\varepsilon_m$  are the final, initial and intermediate energies of the two electron, respectively. In the above expression the sum runs over all the possible intermediate states  $|\varphi_m\rangle$  of the electrons including also the continuum.

Provided the photon frequency is less than the second ionization potential of helium, the transition amplitude (1) can be estimated using the closure approximation. Within the latter all the intermediate states are supposed to have the same energy and the summation over the intermediate states is easily performed. In our calculation, results of which are shown in Fig. 2a, the initial state of helium was approximated by a four-parametric two-electron wave function which includes both radial and angular electron-electron correlations. The final state,

which describes two electrons moving in the continuum and interacting with each other and the He nucleus, was approximated by the so-called 3C-wave function. In addition, we perform more elaborated calculation employing a highly correlated Hylleraas ground state and the convergent close-coupling final state (see [16] for details), which yielded essentially identical emission pattern.

Whereas the electron-electron correlation in the initial state influences the shape of the calculated emission pattern very little, the inclusion of this interaction in the final continuum state turns out to be of crucial importance. It is this interaction which makes the pattern almost spherically symmetric despite the fact that this symmetry is broken by the linearly polarized laser field. The two electrons moving in the continuum with relatively low energies strongly repel each other and, even though still preferring to be emitted along the polarization axis, have to "avoid" to move in the same direction. This in turn makes the momentum pattern of the recoil ions essentially spherically symmetric. In the absence of the electronelectron interaction in the final state both electrons would tend to be emitted along the polarization axis, a feature that would manifest itself as well in more elongated recoil-ion momentum distributions. The calculated spectrum is in good agreement with the experimental data (Fig. 1b), but in disagreement with some earlier calculations that predicted a dominant contribution of both electrons being emitted into the same direction. The spectrum in Fig. 2a exhibits a slight anisotropy, which is more pronounced for large recoil momenta. This feature, however, seems not to be observed in the experimental distribution. Yet, the limited statistical significance of the data along with the finite resolution both prohibit interpreting this discrepancy as a deviation between theory and experiment.

In the case when the incident photon has enough energy to cause the electron removal from the ground state of  $\text{He}^+(1s)$ , the physics of the two-photon – two-electron ionization is quite different. Now the two electrons can be emitted not only together but also

independently, "sequentially" one after the other as discussed before. Since the length of the laser pulse even though very short by any technical means is much larger than the typical atomic time, the overall time delay between the emission of the first and second electron is also large. This makes the independent emission channel to dominate the process and enables consider the latter sequence of two independent one to as a events:  $He^{-\frac{\hbar\omega}{\omega}} He^{+} \xrightarrow{\hbar\omega} He^{2+}$ . As illustrated in Fig. 2b, the calculated emission pattern for the recoil ions considerably changes as compared to NS ionization. Instead of the spot at zero momentum, the spectrum exhibits two pairs of dipole-like half-rings along the polarization direction. Here, both electrons are most likely emitted independently along the polarization direction, one from the neutral He and one from the ground state of He<sup>+</sup>, each forming a dipole-like structure in its momentum distribution. Outer and inner rings on both sides than reflect the situation where both electrons either go into the same or opposite hemispheres, respectively.

Since the FLASH wavelength for the current experiment was fixed to 28 nm (44 eV), sequential TPDI of He was out of reach. Therefore, in order to prove the different dynamic mechanisms, experiments have been performed for Ne, where at 44 eV photon energy one does enter the sequential regime for TPDI: 1<sup>st</sup> and 2<sup>nd</sup> ionization potentials for Ne are 21.5 and 40.9 eV, respectively (see also [24]). Fig. 3a and 3b show the measured momentum distributions for singly and doubly charged Ne ions. In both cases one clearly observes a ring-like shape of the distribution, though considerably different compared to the case of He single ionization (Fig. 1a). This is mainly due to a different symmetry of the Ne ground state [28], though it should be noted that the observed pattern might be also slightly modified by the experimental momentum resolution along the polarization (and jet propagation) direction, which, being defined by the temperature of the atomic target, is somewhat worse for Ne than for He ( $\Delta p \sim 0.7$  a.u. and 0.4 a.u.(FWHM), respectively).

Most importantly, however, the momentum distribution of Ne<sup>2+</sup> ions exhibits significant qualitative changes as compared to both, the He<sup>2+</sup> data at 44 eV (Fig. 1b) and the Ne<sup>2+</sup> results at 38.8 eV (Fig. 4 of [26]), thus, providing first clear evidence for another ionization mechanism, namely sequential ionization being at work. The spectrum in Fig. 3b does not show two clear rings as in the theoretical prediction for He (Fig. 2b). This is most likely due to the different emission patterns of both electrons with a  $\beta$ -parameter of the second between 0 and 0.5, i.e. close to isotropic emission [28]. It should be noted that the electron angular distributions in double ionization of Ne are considerably less studied than those for the case of He, and, for instance, in single-photon double ionization no clear dipole-like structure in the Ne<sup>2+</sup> momentum distributions could be observed at about 30 eV above the double ionization threshold [29], in striking contrast to the corresponding He data [27]. Thus, in order to draw a definite conclusion concerning the reason for these differences, one needs to extend the calculation to the Ne target. Since the anisotropy parameters for the sequential TPDI have been recently published [28], the calculation of the recoil-ion momentum distribution should become feasible in the nearest future.

In summary we have measured recoil-ion momentum distributions for the most fundamental non-linear two-photon – two-electron interaction, where the absorption of two photons results in double ionization of helium. Two basically different mechanisms, sequential and non-sequential ionization were clearly disentangled, which do show distinctly different recoil-ion momentum patterns as a result of the different two-electron emission characteristics. Good agreement with the results of a model calculation is observed for direct ionization, partly in agreement with earlier predictions and the distributions for the sequential pathway are found to be qualitatively reproduced. In the future we envision recording fully differential cross sections for both processes thus providing ultimate experimental benchmark data for advancing theory. The authors are greatly indebted to the scientific and technical team at FLASH, in

particular, the machine operators and run coordinators, for the optimal beamtime conditions.

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Figure 1. Density plot of the measured recoil-ion momentum distributions for single (a) and double (b) ionization of He by 44 eV FLASH photons. The arrow indicates the direction of the FLASH polarization. Inner and outer circles mark the maximum He<sup>2+</sup> momentum for the cases where one electron would have taken all the excess energy, and for the equal energy sharing with the emission of both electrons in the same direction, respectively.

Figure 2. Density plot of the calculated recoil-ion momentum distributions for two-photon double ionization of He at 44 eV (a) and 60 eV (b) photon energies.

Figure 3. The same as Fig. 1 for Ne.



Figure 1



