# Recoil-Ion Momentum Distributions for Two-Photon Double Ionization of He and Ne by 44 eV Free-Electron Laser Radiation

A. Rudenko,[1,2] L. Foucar,[3] M. Kurka,[2] Th. Ergler,[2] K. U. Kühnel,[2] Y. H. Jiang,[2] A. Voitkiv,[2] B. Najjari,[2] A. Kheifets,[4] S. Lüdemann,[2] T. Havermeier,[3] M. Smolarski,[3] S. Schössler,[3] K. Cole,[3] M. Schöffler,[3] R. Dörner,[3] S. Düsterer,[5] W. Li,[5] B. Keitel,[5] R. Treusch,[5] M. Gensch,[5] C. D. Schröter,[2] R. Moshammer,[1,2] and J. Ullrich[1,2]

[1]*Max-Planck Advanced Study Group at CFEL, Notkestrasse 85, 22607 Hamburg, Germany*
[2]*Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany*
[3]*Institut für Kernphysik, Universität Frankfurt, 60486 Frankfurt, Germany*
[4]*Research School of Physical Sciences, The Australian University, Canberra, ACT 0200, Australia*
[5]*DESY, Notkestrasse 85, 22607 Hamburg, Germany*

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Recoil-ion momentum distributions for two-photon double ionization of He and Ne ($\hbar\omega = 44$ eV) have been recorded with a reaction microscope at FLASH (the free-electron laser at Hamburg) at an intensity of $\sim 1 \times 10^{14}$ W/cm$^2$ exploring the dynamics of the two fundamental two-photon–two-electron reaction pathways, namely, sequential and direct (or nonsequential) 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.

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Since Einstein's revolutionary explanation of the photoelectric effect in 1905, the breakup 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 photoabsorption process, however, one faces serious problems in calculations as well as in measurements, even if only a single photon is absorbed at a time. Thus, the simplest situation where two electrons emerge from the He atom has numerically been solved only within the last decade when fully differential experimental cross sections have become available (see [1] for a review).

Keeping the simple He target but increasing the number of photons, 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., [2]). Likewise, kinematically complete experiments for this regime have been reported only within the last two months [3,4]. Also for the process of double ejection by Compton scattering, the comparison of experiment and theory does not go beyond the level of total cross sections [5].

In this Letter we report the first differential measurement, recoil-ion momentum distributions, for the most basic nonlinear two-electron light-matter interaction, where two vacuum ultraviolet photons (44 eV each) "simultaneously" remove two electrons from He. The results are compared to the double ionization of Ne, where a sequential, stepwise absorption pathway with intermediate relaxation to a bound state of the Ne$^+$ 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 yield 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 multiparticle momentum imaging technique, "reaction microscope" [6], and a novel light source, the free-electron laser in Hamburg (FLASH), delivering ~25 fs vacuum ultraviolet light pulses at unprecedentedly high intensities.

Because of its fundamental character and the considerable importance of such processes for the application of free-electron laser radiation in other fields of science, two-photon double-ionization (TPDI) of He has received enormous theoretical attention (for an incomplete recent selection, see [7–15]). 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 ($\sigma_{\mathrm{TPDI}}$) vary over more than 1 order of magnitude [9]. Two measurements of $\sigma_{\mathrm{TPDI}}$ have been recently performed for photon energies of 41.8 [16,17] and 42.8 eV [18], employing high harmonics and FLASH radiation, respectively. This photon energy range (40–54 eV) is especially attractive since here the second ionization potential, i.e., of He$^+$ (1$s$), 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 then requires a direct ["nonsequential" (NS)] process. Both of the experiments [16–18] revealed the dominance of the two-photon NS channel for TPDI at light intensities up to $10^{14}$ W/cm$^2$ reflected in the quadratic intensity dependence of the doubly charged ion yield. However, as was found previously for the single photon [1] and the strong-field regimes [2–4], 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, 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 spectroscopy [4]. To the best of our knowledge, the only differential data for any two-photon transitions in He has been presented in [19]. There, a feature occurring around 24 to 26 eV in the photoelectron spectrum of He irradiated by 38.5 eV FLASH radiation 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 [20]. The reaction microscope spectrometer [6] was installed in the focus of the FLASH beam line BL2. Linearly polarized radiation was focused onto a 30 $\mu$m spot within the collimated supersonic gas jet in the center of the ultrahigh vacuum chamber with a base pressure below $8 \times 10^{-12}$ mbar. Created ions were guided to a position-sensitive channel plate detector by a weak (1 V/cm) electric field. From the measured times of flight and positions on the detectors, the full momentum vectors of the emitted ions were reconstructed. The laser polarization was parallel to the direction of the gas jet propagation. The pulse energy was monitored shot by shot using a Faraday cup. For the averaged value of $\sim$20 $\mu$J per pulse, the estimated peak intensity corresponded to $\sim 1 \times 10^{14}$ W/cm$^2$. High harmonic contributions of the free-electron laser fundamental radiation of less than 1% [21] were suppressed by two Al filters with a total thickness of 0.3 $\mu$m. They provide a transmission reduction for the second and third harmonics of more than a factor of 1000 larger compared to the one for the first order [22], ensuring that the harmonic fraction did not exceed $10^{-5}$ with respect to the fundamental.

Figure 1(a) 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. 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.

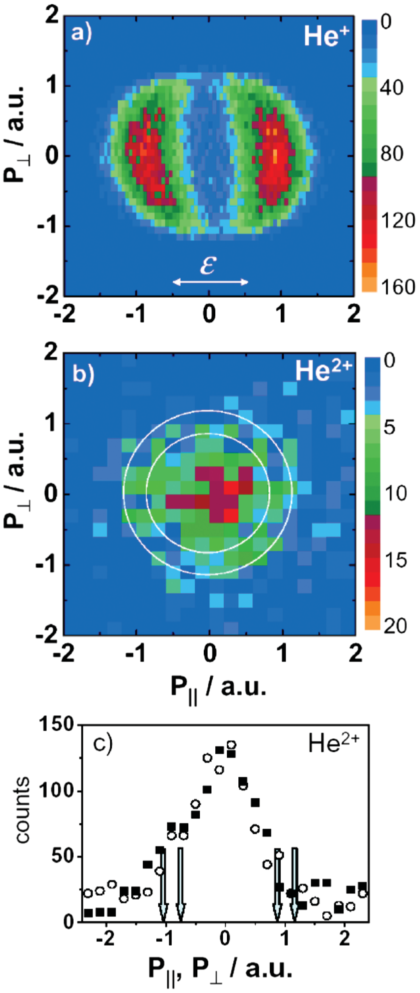

FIG. 1 (color). Density plot of the measured recoil-ion momentum distributions for (a) single and (b) double ionization of He by 44 eV FLASH photons. The arrow in (a) indicates the direction of the FLASH polarization. Inner and outer circles in (b) mark the maximum He$^{2+}$ momentum for the cases where 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. (c) Projections of the 2D distribution of panel (b) onto the axis parallel (solid squares) and perpendicular (open circles) to the polarization directions. Arrows indicate the positions of the circles shown in panel (b).

The measured He$^{2+}$ momentum distribution is presented in Figs. 1(b) and 1(c). In contrast to the single-ionization spectrum, as well as to double ionization induced by a single photon with similar energy above the threshold [23] but in qualitative agreement with our earlier results for Ne [20], it displays a clear maximum at the origin, with most of the events lying well within the inner circle marking the maximum He$^{2+}$ 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 of 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 qualitative difference between the spectra for these two processes might be readily understood.

In order to come to a more quantitative understanding, the $\mathrm{He}^{2+}$ momentum distribution has been calculated for $\hbar\omega_0 = 44$ eV and is shown in Fig. 2(a). The transition amplitude for the two-electron ionization caused by the absorption of two photons is given by (in atomic units)

$$a_{fi}^{(2)} = \frac{\pi}{2i}\delta(\varepsilon_f - \varepsilon_i - 2\omega_0) \times \lim_{\alpha\to+0}\sum_m \frac{\langle\varphi_f|\hat{W}_0|\varphi_m\rangle\langle\varphi_m|\hat{W}_0|\varphi_i\rangle}{\varepsilon_m - \varepsilon_0 - \omega_0 + i\alpha}, \quad (1)$$

with $\hat{W}_0 = (\vec{r}_1 + \vec{r}_2)\cdot\vec{F}_0$.

Here, $\phi_i$ and $\phi_f$ 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 $\varepsilon_f$, $\varepsilon_i$, and $\varepsilon_m$ are the final, initial, and intermediate energies of the two electrons, respectively. In the above expression, the sum runs over all the intermediate states $|\varphi_m\rangle$ of the electrons, also including 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 in (1) is easily performed. In our calculation, results of which are shown in Fig. 2(a), the initial state of He was approximated by a four-parametric two-electron wave function that 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 three-body double continuum Coulomb wave function (3C). In addition, we performed a more elaborated calculation employing a highly correlated Hylleraas ground state and the convergent close-coupling final state (see [10] 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 have to "avoid" moving in the same direction. In the absence of the electron-electron interaction in the final state, both electrons would tend to be emitted along the polarization axis, a feature that would manifest itself in more elongated recoil-ion momentum distributions. The calculated spectrum is in good agreement with the experimental data [Fig. 1(b)], 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. 2(a) exhibits a slight anisotropy, which seems not to be observed in the experimental distribution [see Fig. 1(c)].

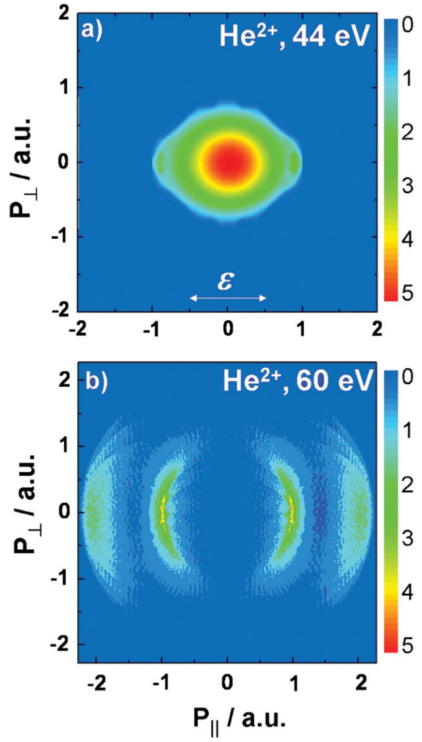

FIG. 2 (color). Theoretical density plot of the recoil-ion momentum distributions for two-photon double ionization of He at (a) 44 eV and (b) 60 eV photon energies.

In the case when the incident photon has enough energy to cause the electron removal from the ground state of $\mathrm{He}^+(1s)$, 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" as discussed before. Since the length of the laser pulse (even though it is 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 dominate the process and enables one to consider the latter as a sequence of two independent events: $\mathrm{He} \xrightarrow{\hbar\omega} \mathrm{He}^+ \xrightarrow{\hbar\omega} \mathrm{He}^{2+}$. As illustrated in Fig. 2(b), the calculated emission pattern for the recoil ions changes considerably 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 emitted independently along the polarization direction, one from the neutral He and one from the ground state of $\mathrm{He}^+$, each forming a dipolelike structure in its

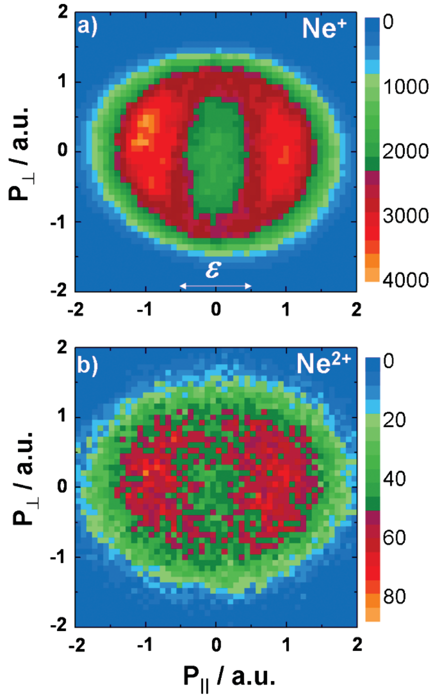

FIG. 3 (color). The same as Fig. 1 but for Ne.

momentum distribution. Outer and inner rings on both sides then 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 at 28 nm (44 eV), sequential TPDI of He was out of reach. Therefore, in order to prove the different dynamic mechanism, experiments have been performed for Ne, where at 44 eV photon energy one does enter the sequential regime for TPDI: first and second ionization potentials for Ne are 21.5 and 40.9 eV, respectively (see also [18]). Figures 3(a) and 3(b) show the measured momentum distributions for singly and doubly charged Ne ions. In both cases one clearly observes the ringlike shape of the distribution, though considerably different compared to the case of He single ionization [Fig. 1(a)], mainly due to the fact that in Ne a different outermost orbital is ionized [24]. Most importantly, however, the momentum distribution of $Ne^{2+}$ ions exhibits significant qualitative changes as compared to both the $He^{2+}$ data at 44 eV [Fig. 1(b)] and the $Ne^{2+}$ results at 38.8 eV (Fig. 4 of [20]), thus providing first clear evidence for another ionization mechanism, namely, sequential ionization being at work. The spectrum in Fig. 3(b) does not show two clear rings as in the theoretical prediction for He [Fig. 2(b)]. 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 [22]. 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 dipolelike structure in the $Ne^{2+}$ momentum distributions could be observed at about 30 eV above the double-ionization threshold [25], in striking contrast to the corresponding He data [23]. Since the anisotropy parameters for the sequential TPDI have been recently published [24], the calculation of the recoil-ion momentum distribution for Ne should soon become feasible.

In summary, we have measured recoil-ion momentum distributions for the most fundamental nonlinear two-photon–two-electron interaction, where the absorption of two photons results in double ionization of He. Two basically different mechanisms, sequential and nonsequential ionization, were clearly disentangled, which 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, 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 the ultimate experimental benchmark data for advancing theory.

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