Direct and Sequential Two-Photon Double Ionization of He and Ne by Intense FLASH Radiation

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Light-induced double and multiple ionization of atoms represent prototype reactions for exploring the dynamics of correlated many-body quantum systems, a subject of increasing interest in many fields of physics, chemistry and possibly in biology. The simplest example is double ionization of He by a single-photon, a realization of the fundamental dynamic three-particle quantum problem. Nowadays, after decades of intense research, experimentally and theoretically, sophisticated quantum mechanical ab-initio calculations are in excellent agreement with all available experimental data [1]. Hence, single-photon double ionization of He is considered to be well understood. This situation changes if more than only one photon is involved. In the extreme case of multi-photon multiple ionization, where typically more than 50 photons at optical frequencies are needed to release both electrons from He, a comprehensive theoretical description is still outside the range of vision [2].

With intense VUV laser light at FLASH it is now possible to experimentally access for the very first time the completely unknown intermediate regime of few-photon multiple ionization, i.e. the interaction of two or three photons with two or three electrons. Beside the relevance of such non-linear light-matter interactions for all those FEL applications that rely on large photon fluxes, from imaging of single bio-molecules to fs time-resolved surface diagnostics, the investigation of few-photon multiple ionization is of decisive importance to advance theory. Mainly due to its perturbative nature along with the fact that only a few photons are involved, an impressively large number of quantum calculations appeared during the last few years on two-photon double ionization (TPDI) of He (see [3] and references therein). Especially intriguing is the so-called 'direct' two-photon induced two-electron emission at wavelengths where the 2nd ionization potential lies above the energy of a single photon, whereas the two-photon sum energy is large enough to overcome the total binding potential of both electrons. The occurrence of direct TPDI of He has been demonstrated using high-harmonics radiation [4] and FLASH light [5], but differential data, needed to benchmark theory, were not available. Such measurements are now possible using FLASH in combination with multi-particle imaging spectrometers ('Reaction Microscopes' [6]). Very recently we have published first differential data on direct TPDI of Ne at 38 eV [7].

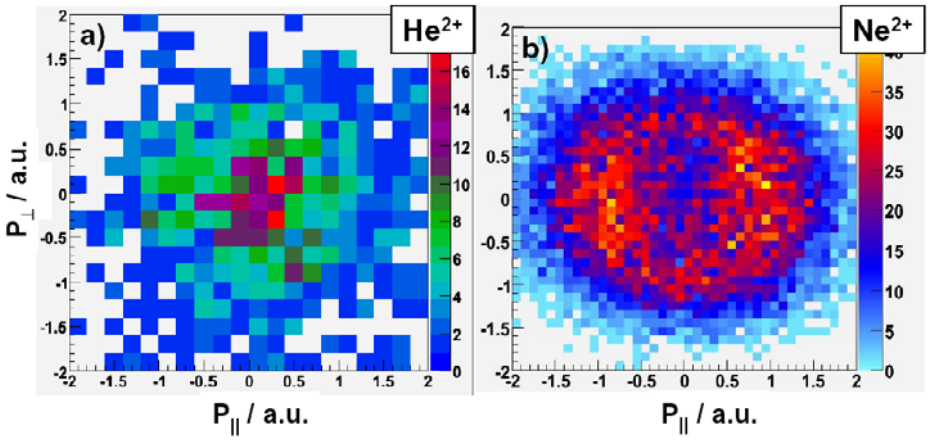

Figure 1. Momentum distributions of doubly charged He (a) and Ne (b) ions measured at 28 nm (44 eV).

Here we report on two-photon double ionization of He and Ne at 44 eV photon energy, results of our last FLASH campaign (beam line BL2). The FEL beam was focused onto a collimated supersonic gas jet in the centre of our ultra-high vacuum ($8 \cdot 10^{-12}$ mbar) chamber. Created ions and electrons were guided to two

position-sensitive channel plate detectors by weak electric and magnetic fields. From the measured times-of-flight and positions on the detectors their full momentum vectors were calculated. The estimated maximum intensity was ~ 10^{14} W/cm^2.
Fig. 1 shows the two-dimensional momentum distributions of He^{2+} (a) and Ne^{2+} (b) ions created by 28 nm (44 eV) radiation. The light polarization is parallel to the horizontal $p_{\parallel}$-axis. At this wavelength two-photon double ionization of Ne can proceed sequentially (1st and 2nd ionization potentials are 21,6 and 40.9 eV, respectively), whereas for He (24.6 and 54.4 eV) only the direct process is energetically possible. The spectra exhibit clear differences between the two cases: whereas for Ne, where double ionization occurs sequentially, the distribution manifests a dipole-like structure with two maxima along the polarization direction, the He spectrum shows a maximum at zero momentum indicating that the two electrons are emitted in a correlated manner, most likely back to back into opposite hemispheres. This is in good agreement with most theoretical predictions for direct TPDI of He at this wavelength [8]. Interestingly, the He^{2+} distribution at 44 eV closely resembles the one measured earlier for Ne at lower photon energies [7], where the sequential two-photon process is energetically forbidden, thus, indicating similar ionization mechanisms being at work.

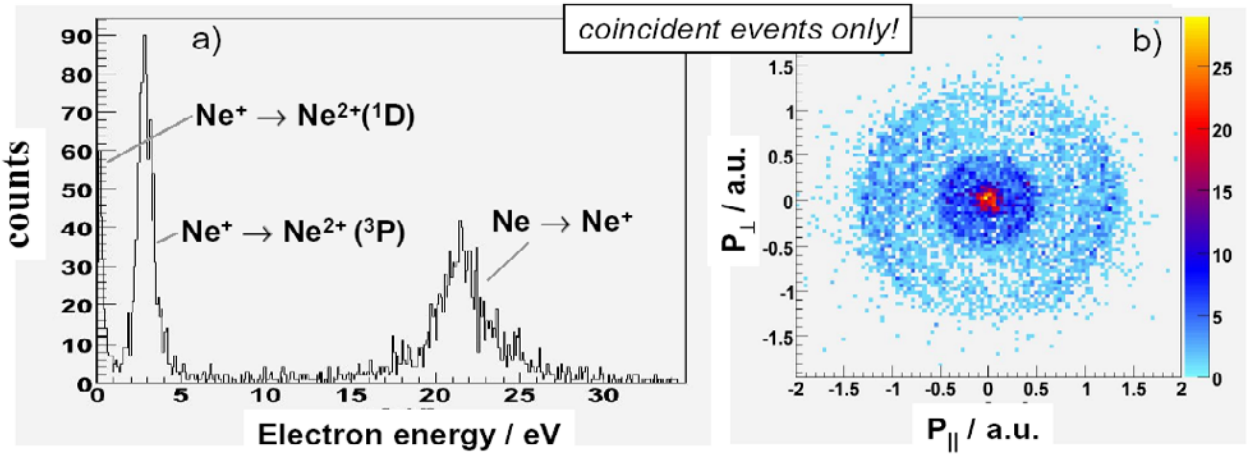

Figure 2. Total energy spectrum (a) and two-dimensional momentum distribution (b) of the electrons measured in coincidence with Ne^{2+} ions. The photon energy is 44 eV, the intensity is around 10^{14} W/cm^2.

During the same beam time we succeeded for the very first time to collect fully-differential data on sequential TPDI of Ne at 44 eV by measuring both electrons in coincidence with the Ne^{2+} ion. Fig. 2 displays the total energy spectrum (a) and the two-dimensional momentum distribution (b) of electrons emitted in Ne double ionization. The high-energy peak in Fig. 2a reflects those electrons which are ejected in the first ionization step (Ne^0 -> Ne^{1+}), constituting the outer ring in the 2D-momentum plot (Fig. 2b). Electrons emitted in the second step (Ne^{1+} -> Ne^{2+}) appear, because of the larger ionization potential of the ion, with smaller kinetic energies. Depending on the electronic configuration of the remaining Ne^{2+} ion, the second electron either carries about 3 eV (the inner ring in Fig. 2b), or it appears in the continuum with nearly zero energy (spot centered at zero momentum in Fig. 2b). Due to this strongly asymmetric electron energy sharing for sequential TPDI the Ne^{2+} ion momentum distribution (see Fig. 1b), which mirrors the sum of both electron momenta, is mainly defined by the fast electron leading to a dipole or ring-like structure. Presently, data evaluation is still in progress.

References

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