Non-Sequential Few-Photon Multiple Ionization of Ne and Ar Atoms by Intense FLASH Radiation

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The dynamics of many-body systems is of central importance in many fields of physics, from solid state to nuclear physics. A particularly clean way to address this problem is the investigation of few-electron ejection from atoms induced by interaction with light. The simplest example of such process is single photon-induced multiple ionization, in particular, double ionization of He. Nowadays, with quantum mechanical *ab initio* calculations being in excellent agreement with all available experimental data [1], this prototype reaction after decades of research is generally considered to be well understood. In contrast, the other extreme case, multi-photon multiple ionization, where about 50 photons of optical frequencies are needed to release both electrons from He, has until the present day resisted any comprehensive theoretical description due to tremendous complications of solving the higly non-linear problem in the non-perturbative regime [2].

Few-photon multiple ionization, i.e. the interaction of two or three photons with two or three electrons bridges the gap between the single and multi-photon regime and, thus, is of decisive importance to advance non-linear theories. Mainly due to its perturbative nature along with the fact that only few photons are involved, full quantum calculations, at least for two-photon double ionization (TPDI) of He are on the horizon [3]. Especially intriguing is the so-called 'direct' or 'non-sequential' (NS) few-photon-induced few-electron emission in the wavelength range where one of the ionization steps can not be made with a single photon, whereas the sum of the photon energies is enough to overcome the total binding potential of all emitted electrons. Very recently NS TPDI of He with 42 eV high-harmonics radiation has been demonstrated [4]. The FLASH FEL, delivering VUV-photons at unprecedented intensities, in combination with the most advanced multi-particle detection systems ('Reaction microscopes' [5]) provides an experimental arrangement with a perspective to measure fully-differential cross-sections of such reactions.

Here we report on the experimental study of the few-photon multiple ionization of Ne and Ar atoms with a dedicated 'Reaction microscope'. Measurements were performed at FLASH beam line BL 2

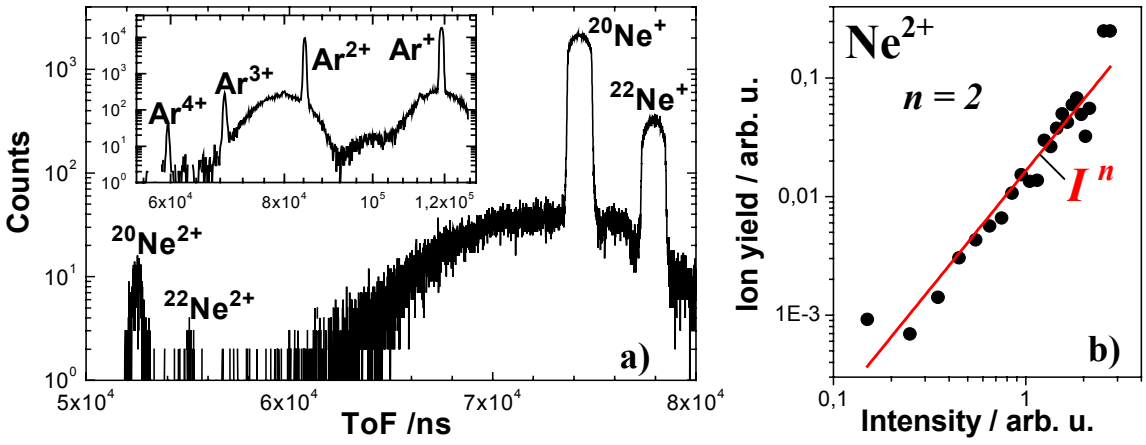

Figure 1. (a) Time-of-flight spectrum of Ne ions created by 32 nm light. Inset: the same for Ar at 44 nm. (b) Intensity dependence of the Ne^{2+} ion yield.

with the wavelengths of 44 nm (28.2 eV) and 32 nm (38.7 eV) and the repetition rate of 2 and 5 Hz, respectively. The FEL beam was focused onto a collimated supersonic gas jet in the middle of ultra-high vacuum ($5x10^{-11}$ mbar) chamber. Created ions and electrons were guided to two position-sensitive channel plate detectors by weak electric and magnetic fields, and from the measured time-of-flight and positions on the detector the full momentum vectors were calculated. After passing through the reaction volume the FEL beam was damped in the Faraday cup, the signal of which was used for relative intensity calibration. The estimated maximum intensity was ~ 10^{13} W/cm^2.

Fig. 1 shows the time-of-flight spectra of Ne and Ar ions created by 32 and 44 nm radiation, respectively. The Ne spectrum is dominated by sharp peaks of singly charged ions from the target ($^{20}Ne^+$ and $^{22}Ne^+$) with small contributions from the residual gas (broad humps). In addition, a small but significant yield of Ne^{2+} ions is observed. A minimum energy of 62.5 eV is needed to doubly ionize a Ne atom, which means that at least 2 photons have to be absorbed at the present photon energy of 38.7 eV. If, however, during the laser pulse the Ne atom gets first singly ionized (binding energy 21.6 eV) by absorption of one photon and then a second electron is removed from the Ne^{1+} ground state (40.9 eV) by absorption of two additional photons, in total 3 photons are needed. According to lowest order perturbation theory, which is expected be valid for non-linear few-photon transitions at medium intensities (< 10^{13} W/cm^2), the ionization yield should increase with intensity as $Y = \sigma_n \cdot I^n$, where σ_n is the generalized n-photon cross-section, I the intensity, and n the number of involved photons [6]. The observed I^2 dependence (Fig. 1b) shows that Ne double ionization is dominantly a 2-photon transition, clearly favouring the NS mechanism mentioned above.

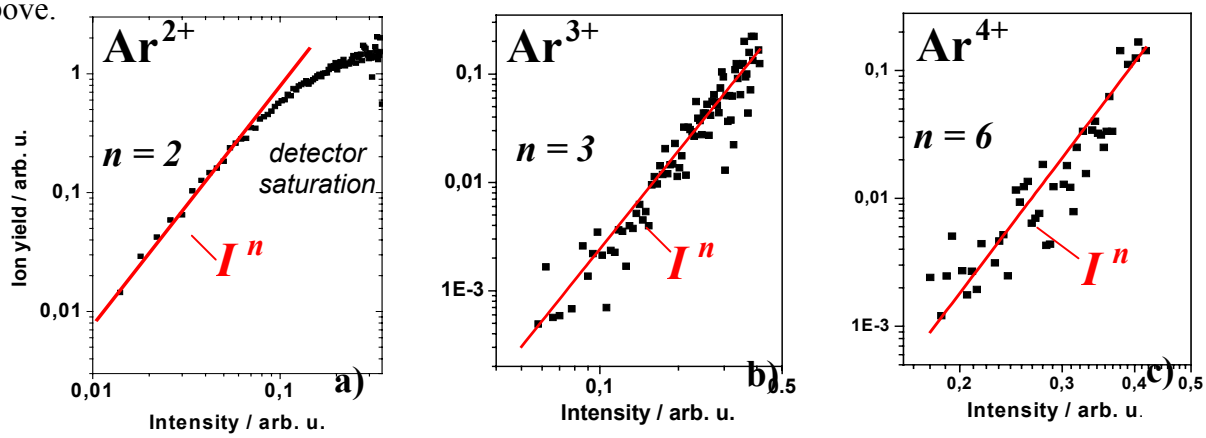

Figure 2. Intensity dependence of the Ar^{2+} (a), Ar^{3+} (b) and Ar^{4+} ion yields at 44 nm

The intensity-dependent ionization yields for various charge states of Ar at 44 nm are shown in Fig. 2. According to the total ionization potentials one needs 2, 3 and 6 photons for double, triple or four-fold ionization, respectively. However, for the sequential production of these charge states 2, 4 and 7 photons are needed. In contradiction to this, Ar^{3+} and Ar^{4+} yields excibit I^3 and I^6 intensity dependence, respectively, again indicating the dominance of NS mechanism. The deviation of the Ar^{2+} yield from the quadratic dependence is due to the detector saturation at high intensities.

Thus, the experimental data are in qualitative agreement with the expectations for an instantaneous NS absorption of exactly as many photons as needed to just overcome the sum ionization potential of the corresponding charge state, which for Ne^{2+} and Ar^{3-4+} definitely means NS ionization. This conclusion is also supported by the first measurements of ion momentum distributions.

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