

Multiple Ionization of Atoms by 25 and 7 fs Laser Pulses

A. Rudenko, B. Feuerstein, K. Zrost, V.L.B. de Jesus, C.D. Schröter,
R. Moshhammer and J. Ullrich

Max-Planck-Institut für Kernphysik, Saupfercheckweg 1,
69117 Heidelberg, Germany
e-mail: Artem.Rudenko@mpi-hd.mpg.de

Abstract. Double, triple and fourfold ionization of rare gas atoms has been studied using a “reaction microscope”. We show that ionization dynamics drastically depends on atomic species and present the first data obtained with few-cycle pulses.

Since the first experimental observation of doubly charged ions produced by intense laser field [1], many-electron dynamics in laser-induced ionization has been the subject of numerous experimental and theoretical studies. It was soon found out that the yields of second and higher charge states produced by intense linearly polarized laser pulses exceed significantly those expected for independent successive removal of two or more electrons. The origin of this enhancement (which was called “non-sequential” ionization) remained controversial until 2000 when first measurements of recoil ion momentum distributions [2,3] along with the results of earlier experiments using circularly polarized light [4] provided convincing evidences in favour of the so-called “recollision” model. However, the details of the double ionization dynamics are still far from being completely understood. In particular, one of the most intriguing questions is whether and how atomic structure can influence ionization dynamics [5].

For the case when more than two electrons are ionized, there is much less experimental results available. Intensity dependence of ion yield was measured for different rare gases (see, e.g. [6]). Ion momentum distribution obtained in [3] for Ne^{3+} at 1.3 PW/cm^2 remains, to the best of our knowledge, the only differential data published up to now. Here we report on the systematic experimental study of multiple (up to fourfold) ionization of Ar and Ne by intense laser field and present the first results obtained with the laser pulses as short as 7 fs. The experiments were performed using a new “reaction microscope” [7] designed to meet the specific requirement of the experiments with high-intensity lasers. We used linearly polarized radiation of a Kerr-lens mode locked Ti:sapphire laser at 795 nm wavelength with 25 fs pulse width (FWHM). To generate few-cycle pulses they are spectrally broadened in a gas-filled hollow fiber and then compressed to 6-7 fs (FWHM) by chirped mirrors and a prism compressor.

Fig. 1 shows momentum distributions of Ne^{2-4+} ions along the laser polarization direction. All spectra exhibit a clear double peak structure which

is a signature of the recollision process. In the momentum distributions of Ne^{3+} and Ne^{4+} ions this structure is even more pronounced than for double ionization, with almost no ions produced with zero momentum. Momentum distribution of Ne^{2+} ions created by 7 fs laser pulse (solid line in Fig.1a) closely follows the results obtained with 25 fs pulses, indicating that momentum distribution is mainly defined by the recollision within one or two cycles after the removal of the first electron. The widths of the spectra are in good agreement with kinematical constraints following from the classical consideration [8]. In general, confirming the results of [3] we can state that ion momentum distributions of Ne^{n+} ions agree well with the model assuming direct impact ionization by rescattered electron via (e,ne) process.

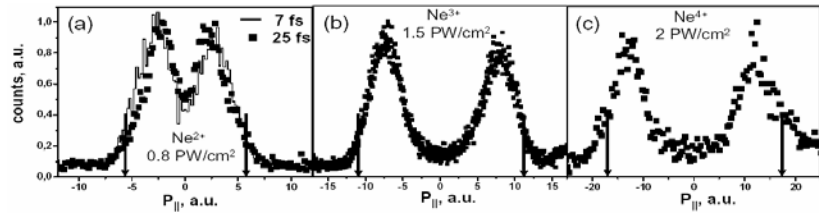


Fig. 1. Longitudinal momentum distributions of Ne^{n+} ions. Arrows indicate ion momenta of $\pm 2n\sqrt{U_p}$

However, considering the momentum distributions of Ar^{2-4+} ions (Fig. 2), we can conclude that for Ar other mechanisms play an important role. In longitudinal momentum distributions of Ar^{2-3+} ions obtained with 25 fs pulses (squares in Fig.2a,b) minimum at zero momentum is pronounced much less than in the case of Ne. For Ar^{4+} ions created by 25 fs laser pulse we do not observe double peak structure at any intensity! Such ‘filling of the valley’ can be attributed (at least for the case of double ionization) to the contribution of electron-impact excitation during recollision followed by field ionization of the excited ion in one of the subsequent cycles of the oscillating laser field.

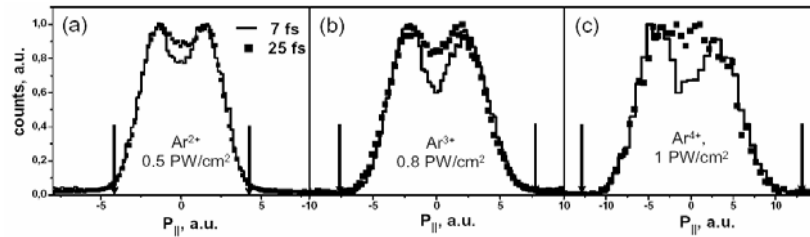


Fig. 2. Longitudinal momentum distributions of Ar^{n+} ions. Arrows indicate ion momenta of $\pm 2n\sqrt{U_p}$

Here the ionization step occurs close to the maximum of the field, leading to relatively small ion momenta. Analysis of the ratios of collision-induced ionization/excitation cross sections shows that for Ne direct electron impact ionization clearly dominates, whereas for Ar (as well as for He) the mechanism involving excitation plays a decisive role [5].

The contribution of the latter mechanism should be suppressed for a shorter laser pulses. This is in a good agreement with the data we have obtained with 7 fs pulses. Comparing the results obtained for Ar with two different pulse lengths, we see that the fraction of ions with zero momentum is considerably reduced for a 7 fs pulse. In this case the minimum at zero momenta is more pronounced for double and triple ionization, and can be also observed for Ar^{4+} ions. The same depletion of the region around zero momentum can be also caused by the suppression of the sequential ionization from the ground state. However, the contribution of purely sequential production of Ar^{2+} , Ar^{3+} and Ar^{4+} at corresponding intensities is still a small fraction of the total yield [9]. Nevertheless, for the production of triply and fourfold charged ions there is a possibility to reach the final state via a combination of sequential and nonsequential processes, and this dynamics might be changed for a few-cycle pulses. Contribution of such combined processes can also explain the relative narrowing of Ar^{3+} and Ar^{4+} momentum distributions (compare the position of the arrows at Fig.2 and 3) [8].

In conclusion, we have studied multiple ionization of Ar and Ne by 25 and 7 fs laser pulses. Whereas for all charge states of Ne we have observed ion momentum distributions consistent with the kinematics of direct (e,ne) process induced by the electron recolliding with its parent ion, for Ar both the shape and the width of the spectra indicate more complex process with different mechanisms being of importance. For double ionization recollision-induced excitation can explain the differences observed. This explanation is strongly supported by the results obtained with 7 fs laser pulses. For higher charge states more elaborated description, which considers the possibility of multiple excitations and different combinations of sequential and nonsequential processes, is required.

References

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