

Ultrafast quantum control of ionization dynamics

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Abstract. The unprecedented combination of transient absorption and ion mass spectroscopy with attosecond resolution is used to study and control the complex multidimensional excitation and decay cascade of an ultrafast Auger process in krypton.

1 Introduction

Attosecond spectroscopy has revolutionized our understanding of ultrafast electron dynamics in atoms, molecules and solids. Typically, these experiments use attosecond pulses or pulse trains and explore dynamical processes in a pump-probe scheme. Either the photoelectrons [1] generated during the experiment or XUV absorption [2] or changes of the ionic state [3] are usually detected. While photoelectron studies generally suffer from electron backgrounds, the detection of the correspondingly produced ions is background-free. However, both methods involve the release of photoelectrons and thus are only suitable for the study of ionizing events. Transient absorption spectroscopy is not limited to ionizing events and it is the method of choice for bound-bound transitions. However, it typically lacks the dynamic range to simultaneously detect absorption features of different co existing ionic charge states.

Here, we overcome these limitations by the first combination of transient absorption and ion spectroscopy, both with attosecond temporal resolution. With this, we acquire a full picture of the excitation of rare gas atoms and of the subsequent ultrafast Auger decay mechanism including ionization dynamics. On top of that, we control the temporal

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evolution and the absolute yield of the ionization dynamics by accessing different level groups in the cascade with different near infrared (NIR) intensities.

2 Experiment and Results

As depicted in figure 1 a, an attosecond XUV pulse centered at 90 eV (for its spectrum see, figure 1b) highly excites krypton atoms. A few cycle NIR pulse with adjustable time delay with respect to the XUV pulse interacts with the sample. The excitation and decay dynamics are investigated in dependence of that time delay by means of an XUV grating spectrometer and a reflectron type ion spectrometer, the latter one recording different ion charged states (see figure 1c).

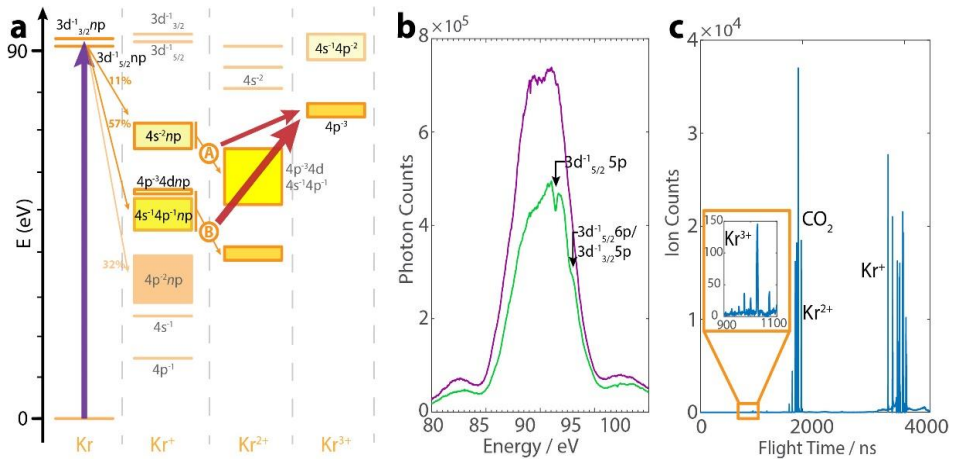


Fig. 1. **a** Krypton energy scheme with XUV excitation (violet arrow), potential decay channels (orange arrows) and further ionization by an NIR few cycle laser pulse (red arrows). The different channels are addressed by different NIR laser intensities (indicated by a thin and thick red arrow). **b** Incident XUV spectrum centered at 90 eV (violet) and a typical krypton transmission spectrum (green). **c** Measured ion spectrum yielding singly, doubly and triply charged krypton ions. For the measurements shown in b and c, the time delay between XUV and NIR pulse was set to $\Delta t = -150$ fs (the NIR pulse advancing the XUV pulse by 150 fs).

For the detailed study of ultrafast excitation and decay dynamics in krypton, the spectrally resolved change in absorbance (optical density $OD = -\log_{10}[I(\Delta t)/I_0]$, with I_0 as incident XUV spectrum) and the isotope resolved Kr ion yield are recorded as a function of the XUV-NIR time delay, and for different NIR intensities.

Adjusting the laser electric field amplitude allows us to selectively address different intermediate states of the autoionization cascades that follow the XUV excitation resulting in different apparent lifetimes. With that, we obtain control of the temporal evolution and the absolute yield of the ionization dynamics by accessing different level groups in the cascade with different NIR intensities. The accompanying absorption measurement enables the determination of the instrument response function, the lifetimes of resonantly excited states involved in the process and proves that the overall conditions of the experiment (resonant excitation and ionization) remain unchanged for a large range of NIR intensities.

Our experimental observations (see figure 2) are confirmed by our multiconfiguration Dirac-Fock calculations and by taking the average of all the individual lifetimes, weighted by their relative decay probabilities. With this, the effective lifetimes of different potential group cascades are revealed and the involved level groups can be identified for the first time [4].

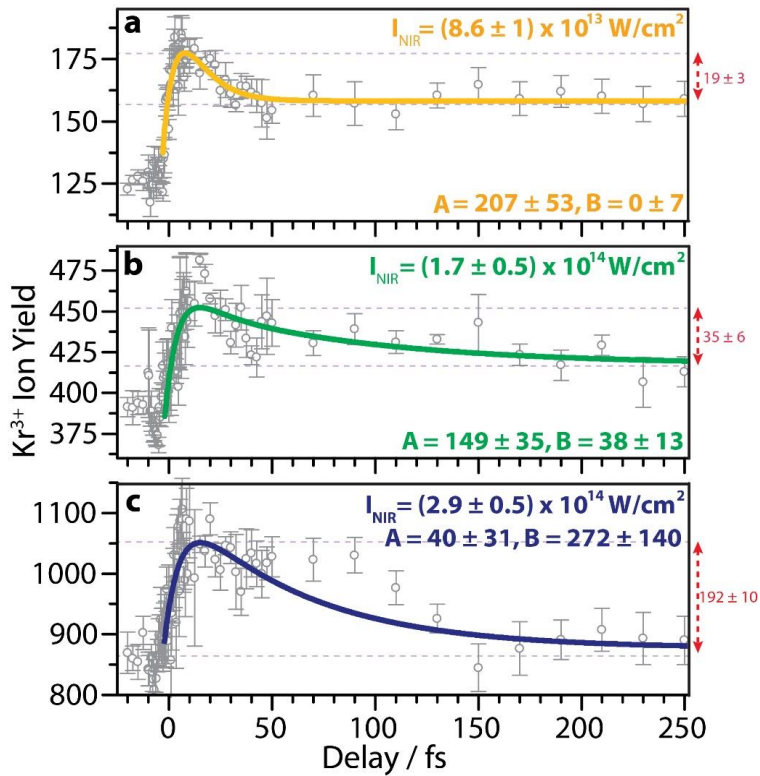


Fig. 2. Time dependent Kr^{3+} ion yield for three different NIR intensities. The krypton ionization dynamics are controlled by variation of the NIR intensity I_{NIR} addressing different decay channels (indicated by the bold letters A and B, please compare with figure 1a). This results in different apparent lifetimes.

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