Controlling the Double Photoionization Dynamics of Li Atoms by Optical Pumping

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Synopsis Double photoionization (DPI) of lithium atoms in a magneto-optical trap by 85 eV and 91 eV photons from the Hamburg Free Electron Laser (FLASH) is investigated using cold target recoil ion momentum spectroscopy. For ejection of a $1s$ and the optically excited $2p$ electron we demonstrate that the DPI cross section significantly depends on the alignment of the $2p$ valence orbital. Thus, the dynamical electron-electron correlation can be manipulated giving insight into the DPI mechanisms close to threshold.

With the goal to perform kinematically complete double and triple ionization studies on lithium we have set up a new apparatus implementing a magneto-optical trap for Li atoms into a Reaction Microscope (MOTREMI) capable of multi-particle imaging. Thus, in combination with ultra-intense free electron lasers, fully differential studies of photon-induced triple ionization of Li with tiny cross section of several barns and even below will become possible in future.

This pilot work on DPI of Li was performed at FLASH, which delivered VUV photon pulses with duration about 30 fs and up to 10^{12} photons per pulse. Our setup allowed for the preparation of the target initial electronic state by optical pumping and, for the first time, this was demonstrated to be used to control the DPI dynamics. With a linearly polarized laser beam on the transition $\mathrm{Li}(2s\,^2S_{1/2}) \rightarrow \mathrm{Li}(2p\,^2P_{3/2})$ a fraction of about 45% of the atoms could be excited and aligned along the laser polarization. Besides single ionization, the subsequent FLASH pulse initiated double ionization involving one of the two $1s$ electrons and the valence electron.

Interestingly, the DPI cross section sensitively depends on the spatial alignment of the $2p$ excited initial state. This could be understood intuitively as illustrated in Fig.1: DPI should be more likely if the $2p$ orbital is parallel to the $1s$ dipole emission pattern, since the electron-electron interaction is stronger compared to the perpendicular alignment. Preliminary theoretical calculations confirm this assertion. Thus, just by modifying the geometry of the system without changing its internal energy, a two-electron process is strongly influenced via dynamical electron correlation.

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Even more compelling is the observation, that this alignment sensitivity decreases as the photon energy increases. It demonstrates that this effect is not enforced by symmetry, but rather is a subtle dynamical correlation which seems to be more effective towards the double ionization threshold.

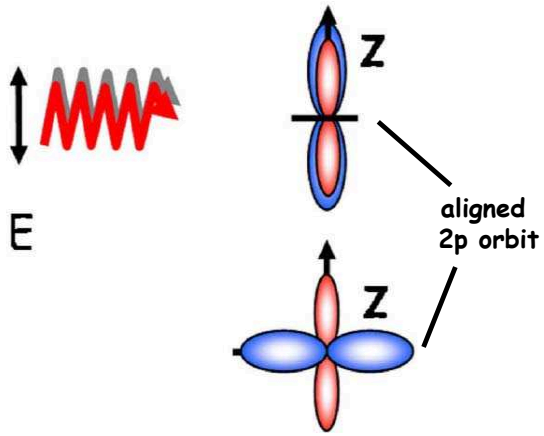

Fig. 1. Schematic of the DPI process, showing that the lithium target is initially prepared with different spatial alignments of the laser excited $nl = 2p$ orbital (blue lobes) relative to the indicated FEL polarization (E), and the dipole emission pattern of the ionized $1s$ electron (red lobes).

In future, combining the upcoming Stanford Linac Coherent Light Source (LCLS) with MOTREMI, will enable to trace the non-linear correlated many-electron quantum dynamics, e.g. in sodium atoms. This novel super-brilliant x-ray source providing photon energies in the keV regime will allow to investigate many phenomena as multiple photon absorption, electron delocalization, and ultra-fast electronic rearrangement of the electronic shell.