Controlling two-electron threshold dynamics in double photoionization of lithium by means of initial state preparation

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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 a small cross section of several barns and even below will become possible in future.
In April 2008, a pilot experiment on double photoionization (DPI) of lithium close to threshold (by 85 eV and 91 eV photons) was performed at FLASH, which delivered VUV photon pulses with a duration of about 30 fs and up to 10^{12} photons per pulse. Our setup allowed for the preparation of the target initial electronic state $\{L\, M_L\, S\, \pi\}$ by optical pumping and, for the first time, this was demonstrated to be used to control the DPI dynamics [1]. With a linearly polarized laser beam on resonance with the transition $\mathrm{Li}(2s\,^2S_{1/2}) \rightarrow \mathrm{Li}(2p\,^2P_{3/2})$ up to 45% of the atoms could be excited, leading to a symmetry change of the initial state. Moreover, alignment along the laser polarization could be achieved, varying the magnetic sublevel population with respect to the quantization axis defined by the VUV electric field vector. In this study the recoil ion momenta from single ionization and double ionization involving one $1s$ electron and the valence electron were observed.
Fig. 1 shows the DPI cross sections in dependence of the recoil ion momentum for the differently prepared initial states at the photon energy of 85 eV. The significant increase of the integral cross section in going from the $2s$ to the $2p$ initial state is consistent with the 1.8 eV increased excess energy due to the excited initial state. Interestingly the cross section depends also on the geometry of the excited state as sketched in Fig. 1. For alignment of the $2p$ orbital (blue lobes) parallel to the dipolar emission pattern of the $1s$ electron (red lobes) double ionization is more likely in particular for small recoil ion momentum below 0.6 a.u. compared to perpendicular alignment. Since the ion compensates the sum momentum of the electrons, a small recoil ion momentum corresponds to their symmetric back-to-back emission. Thus, a possible explanation which is supported by preliminary theoretical calculations is related to the threshold dynamics of double ionization which requires the back-to-back emission of both electrons in order to minimize their Coulomb repulsion. This ideal emission configuration can be reached easily for parallel alignment leading to an increased cross section at small recoil ion momentum. On the other hand for the perpendicular case not only the spatial distribution but also the respective momentum wave function is perpendicular to the VUV polarization axis suppressing back-to-back electron emission with small recoil ion momentum. This picture is supported by two observations. Firstly, no alignment dependence was observed for simultaneous ionization and excitation. While this reaction is also mediated by electron correlation no long range continuum interaction is involved. Secondly, the alignment dependence for DPI was observed to decrease significantly for increasing photon energy to 91 eV, i.e. for growing energy distance from threshold. This demonstrates that this effect is not enforced by symmetry, but rather is a subtle dynamical correlation which is more effective towards the double ionization threshold.

In conclusion a new means to modify the dynamic electron correlation leading to double photoionization is demonstrated by laser preparing the target atoms in an excited state and with different spatial alignment. While in the present experiment the cross section could be modified by a factor of 1.34 for 6 eV excess energy a much higher contrast can be expected closer to threshold. Thus, by means of a purely geometrical modification of the target initial state without changing its internal energy the dynamical electron correlation can be controlled such that PDI is either enabled or essentially suppressed.
In future the increased photon flux of the free electron laser FLASH as compared to 3rd generation synchrotrons in combination with the MOT reaction microscope will make kinematically complete studies of triple photoionization of lithium possible. Thus, one of the simplest four-body Coulomb break-up reactions will become accessible despite its small cross section in the order of a few $10-24$ cm^2. Highly desirable for such studies is an increased repetition rate of the FLASH light pulses in the order of 10 kHz.

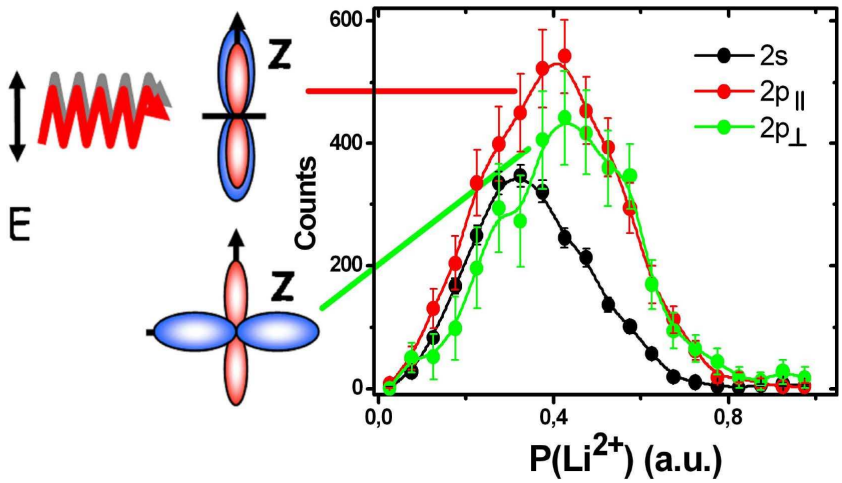

Figure 1: Left: Scheme of the DPI process, showing that the lithium target is initially prepared with different spatial alignments of the laser excited nl = $2p$ orbital (blue lobes) relative to the indicated FEL polarization (E), and the dipole emission pattern of the ionized $1s$ electron (red lobes). Right: DPI cross section as function of the Li^{2+} recoil momentum for parallel $Li(2p\,\|)$ (red) and perpendicular $Li(2p\,\perp)$ (green) alignments, compared to the ground state $Li(2s)$ (black) as a reference.

References

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