Controlling Double Photoionization of Lithium Atoms and an Ultracold Plasma

G. Zhu, M. Schuricke, R. Hubele, G. Veeravalli, J. Steinmann, J. Albrecht, A. Dorn, I. Ben-Itzhak[1], T.J.M. Zouros[2], J. Colgan[3], M. Pindzola[4], and J. Ullrich

Max-Planck-Institut für Kernphysik, D-69029 Heidelberg, Germany

[1]J.R.Macdonald Laboratory, Physics Department, Kansas State University, KS 66506 Manhattan, USA

[2]Dept. of Physics, Univ. of Crete, P.O. Box 2008, 71003 Heraklion, Crete, Greece

[3]Los Alamos National Laboratory, NM 87545 Los Alamos, USA

[4]Department of Physics, Auburn University, AL 36849 Auburn, USA

Two projects were pursued combining three forefront technologies: a magneto-optical trap (MOT) for lithium generating an ultra-cold and, by means of optical pumping, a state-prepared target; a reaction microscope (ReMi), enabling the momentum resolved detection of all reaction fragments with high-resolution and the free-electron laser in Hamburg (FLASH), providing an unprecedented brilliant photon beam.

The first study aims at a better understanding of fundamental many-electron reactions and at developing possible schemes to control their dynamics [1]. Especially intriguing and of fundamental interest is the complete break-up of simple atoms at threshold. Here, the total kinetic energy available in the final state approaches zero resulting in a subtle balance and, ultimately, in complete correlation between all particles involved dispensing with any attempt at an independent particle or self-consistent field approximation.

We have investigated the photo double ionization (PDI) and the simultaneous ionization-excitation for lithium prepared in different initial states Li(1s^2 2*l*) (l = s, p). The excess energy of the linearly polarized VUV-light was between 4 and 12 eV above the DPI-threshold (85 eV and 91 eV photons) for ejection of the valence and one K-shell electron. Close to threshold the total as well as differential DPI cross sections were observed to critically depend on the excitation level and the symmetry of the initial state. As an example in Fig. 1 Li^{2+}(1s) momentum distributions are shown. Since the absorbed photon momentum can be neglected the ion balances the sum momentum of the ejected electrons. For PDI from the initial Li(2s ^{2}S^e) groundstate a minimum for small ion momentum is observed (*a*). This is due to the odd parity of the two-electron continuum state suppressing their back-to-back emission with equal energies which is the preferred configuration close to threshold (the Wannier-configuration). Accordingly, the initial Li(2p ^{2}P^o) state with reversed parity results in a maximum at zero ion momentum since here the Wannier-configuration is allowed (*b*). Interestingly, for the initial excited state the PDI dynamics strongly depends on the alignment of the 2p-orbital with respect to the VUV-light polarization and, thus, from the population of the magnetic substates ($m_p = 0, \pm 1$) [1]. The alignment sensitivity decreases for increasing excess energy and is completely absent for ionization-excitation. Our experimental observations can be consistently understood in terms of the long range electron correlation among the continuum electrons which gives rise to their preferential back-to-back

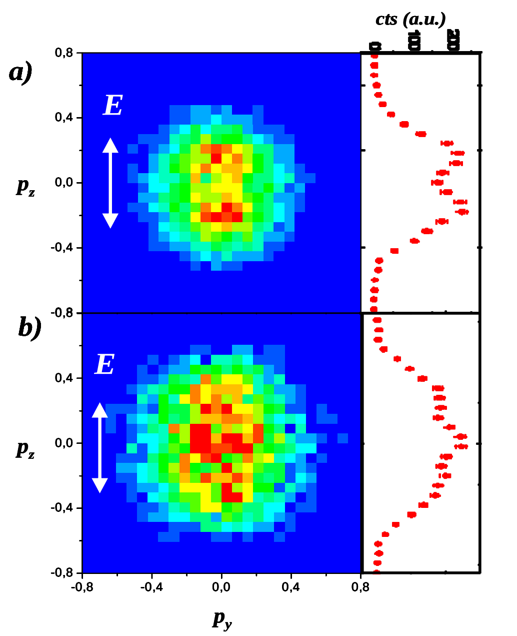

Figure 1: Two-dimensional recoil-ion momentum distributions for PDI by 85 eV FEL light of a) Li(2s ^{2}S) and b) laser exited Li(2p ^{2}P). The VUV- and in (b) also the optical pumping laser polarization are aligned parallel along the marked axis. Right: projections of the data onto the z-axis.

emission. This alignment effect, which was observed here for the first time, allows controlling the PDI dynamics through a purely geometrical modification of the target initial state without changing its internal energy. Time-dependent close-coupling calculations are able to reproduce the experimental total cross sections with deviations which are below 15 % in most cases. In the next step we will exploit the ultra-high intensities available at FLASH for studies of *VUV non-linear few-electron quantum dynamics, e.g.* two- and three-photon absorption leading to double- and triple-ionization of lithium.

In a second project we aim at exploring the dynamics of an ultracold neutral plasma by photoionizing a cloud of laser-cooled lithium-atoms near the ionization threshold such that the emitted electrons are slow and trapped by the positive space charge. Fundamental interest in such systems stems from the possibility of creating strongly coupled plasmas where the Coulomb interaction energy E_C between the charged particles exceeds the thermal energy E_T (Coulomb coupling parameter $G = E_C/E_T > 1$). Equilibrium in these systems may involve the establishment of spatial correlations between the particles and theoretically so called Coulomb crystallization has been predicted but not observed experimentally so far [2].

In the first step using intense FLASH radiation a cloud of 10^7 cold Li atoms is inner-shell ionized at threshold: $Li(1s^2\ 2s\ ^2S) + \gamma(64.5\ eV) \rightarrow Li^+(1s2s\ ^3S) + e^-(E \approx 0\ eV)$. Thus, a plasma of metastable lithium ions (decay-time τ = 40 s) is generated in the first step. Since each ion carries 59 eV excitation energy the stored “inner” energy of the ion cloud can easily exceed 5 GeV. This system constitutes a new plasma regime which can be considered as a threefold inverted plasma since initially its internal excitation energy exceeds the mutual ion’s Coulomb energy which in turn is larger than the plasma’s thermal energy while in conventional plasmas this order is reversed. At present the time evolution dynamics of such a system is completely unknown. Therefore one has to determine the lifetime of the neutral plasma which might be limited if the internal energy is efficiently converted into kinetic energy, e.g. via superelastic collisions with the surrounding slow electrons. Then, shortly after production of the plasma a burst of fast electrons would escape the plasma until its positive potential exceeds the electron energy resulting in strong plasma heating.

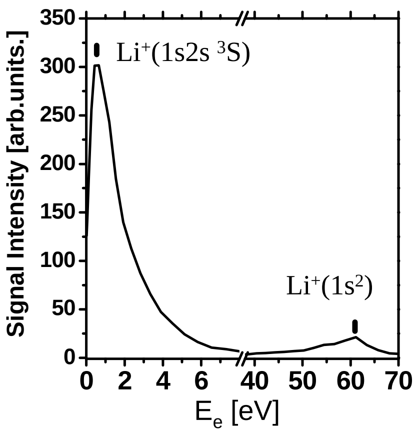

Figure 2: Photoelectron spectrum deduced from the recorded recoil ion momentum after K-shell ionisation of Li at threshold ($E_\gamma \approx 65$ eV).

The first test experiments at FLASH at a photon energy of 65 eV provided photoelectron spectra as shown in Fig. 2. Besides 2s-ionization resulting in the peak around 60 eV a strong line at 0.6 eV originating from 1s-ionization is visible. Due to low target density and relatively low FLASH pulse energy of 10-20 μJ only about 200 ion-electron pairs were produced in one shot. Therefore, no significant plasma effects such as space charge induced line shifts or new lines due to super-elastic collisions were observed. In future the improved FLASH parameters as pulse energies beyond 100 μJ, a high number of micro-bunches in order to accumulate plasma density in combination with a factor of 10 higher target density will provide at least 10^5 electron-ion pairs in the trap giving rise to strong plasma phenomena. In order to build up the plasma in as short time as possible the minimum nominal micro-bunch separation of 200 ns would be highly desirable.

References

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