FLASH photofragmentation of fast molecular and cluster ions

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With the intense short-wavelength laser pulses of FLASH, photofragment imaging experiments on molecular ions using dilute fast moving ion targets in the crossed beams geometry have proven feasible in the VUV and soft-X-ray spectral range. With the TIFF facility (Trapped Ion Fragmentation at FLASH), an ion-beam infrastructure has been created for this purpose at the FLASH monochromator beam line (PG2). A mono-energetic fast ion beam is mass-separated and transported in an electrostatic beam line that includes a fast-beam ion trap for intermediate storage of the ions on the time scale of milliseconds to seconds, and fast ion pulsing facilities (rise and fall times down to $\sim$100 ns). Following the crossed ion and photon beam interaction region, three-dimensional event-by-event reaction imaging can be performed using space and time resolving particle detectors for all charged as well as neutral products. Thus, chemical dynamics on the high-lying potential surfaces reached by energetic photons can now be probed for a wide range of species and fragmentation channels inaccessible until now, extending from elementary ionic molecules up to larger molecules and cluster systems. For larger systems, numerous neutral fragment channels are expected even after multiple ionization by X-ray photons.

The excited potential curves leading to VUV photodissociation of the elementary ion HeH^+ were probed in a first experiment [1, 2] using 38.7 eV excitation energy (32 nm photons) and a beam of 4.2 keV HeH^+ ions. The momenta and directions of fast He products after the photoexcitation from the $^1\Sigma^+$ ground state of HeH^+ to Σ and Π potential curves below the double ionization threshold were imaged, showing that Π final levels (70% to 90%) and $\sim$50% of highly excited atoms He($1snl$) with $n \geq 3$ dominate the photodissociation cross section and need to be included in future theoretical studies.

Ongoing studies are focused on polyatomic molecular ions with particular emphasis on H_3O^+. At energies sufficient for L-shell ionization of H_3O^+, yielding in general excited H_3O^{++}, a significant

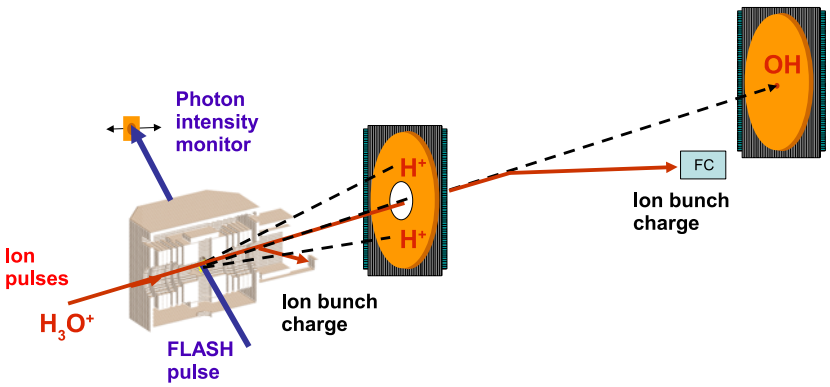

Figure 1: Scheme of the TIFF experiment showing the new arrangement of two detectors and illustrating their application for full detection of light and heavy dissociation fragments. Fragments are shown for the example of dissociative ionization of H_3O^+ by FLASH into the channel OH + 2 H^+.

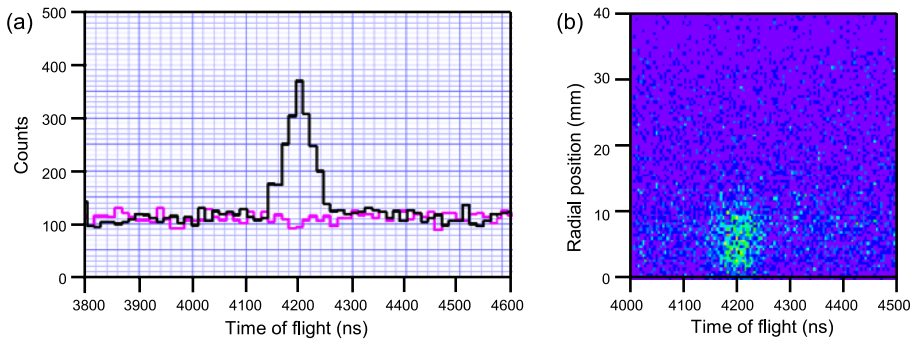

Figure 2: Low-energy neutral fragments (<1 eV kinetic energy release) from the interaction of H_3O^+ ions with photons of 92 eV (13.5 nm). (a) Time-of-flight distribution (black: laser on, purple: laser off) after a measurement time of 1.4 hrs (220 000 pulses) with about 5 μJ per pulse. (b) Spatial and temporal distribution of events, showing a transverse kinetic energy release below 1 eV. (Preliminary)

fraction of heavy neutral products (O or OH) has been detected, momentum imaging revealing a small energy release below $\sim$1 eV. Work towards identifying coincidences with lighter photofragments is in progress. A much smaller yield of neutral products is found using a target ion beam of H_2O^+. By the universality of neutral fragment detection at TIFF, the work complements recent findings on neutral products generated by K-shell ionization of water [3].
Comprehensive technical developments were implemented to enlarge the capabilities of the TIFF setup. In particular, the detection of light fragments leaving the interaction region at large angles was made possible by installing a new 80-mm diam. channelplate detector with a hexanode delay-line imaging anode with a central 20-mm bore at $\sim$30 cm distance from the interaction region. The interaction region was modified in order to minimize the FLASH photon-generated background on this open detector to a level that allows its rejection by time-of-flight discrimination. Moreover, the intensity of molecular ions derived from water was strongly increased by new procedures in the hollow-cathode ion source yielding, e.g., $\sim$50 nA of H_3O^+ and $\sim$20 nA of $H^+(H_2O)_3$. Hardware development is underway for electron detection at the interaction region, aiming at the investigation of fragmentation in polyatomic molecules and clusters with identification of the initial FLASH-induced photoionization process.

References

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