New insight into excited molecular matter: FLASH photodissociation of HeH^+

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The fragmentation of small quantum systems following single photon absorption in the VUV to soft X-ray regime is of central interest for understanding the effect of naturally occuring radiation fields. The absorption of an energetic photon excites or ionizes the electronic system, and the internal energy redistribution then leads to dissociation fragments that determine the further radiation effects. Moreover, these fragments reflect the dynamical processes on the quantum mechanical potential surfaces of the excited small quantum systems, that can be explored in a so far inaccessible regime using the intense light pulses of FLASH. With a crossed fast ion beam, the fragmentation channels of these processes, including all neutral products, become accessible on an event-by-event basis for systems with a wide range of complexity, comprising atomic ions, small and large molecules, and clusters.

An ion beam infrastructure has been created for this purpose at the monochromator beam line (PG2) at FLASH. A mono-energetic fast (keV) 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 a milli-second to second time scale, and fast ($\geq$100 ns) ion pulsing facilities (Trapped Ion Fragmentation at FLASH, TIFF). When the PG2 beamline started to operate with high transmission and a well-controlled photon beam path, the photofragment geometry (Fig. 1) and absolute yield were measured at 38.7 eV excitation energy (32 nm) for HeH^+ on the channel H^+ + He(1snl).

HeH^+ is the simplest heteronuclear two-electron system and a benchmark case as the ionic analogue of H_2; its excited states, however, have so far been addressed only theoretically [1] and indirectly through plasma collision studies involving He. For the isotopomer HeT^+, formed in the beta decay of T_2, the excited state properties are of large interest as they determine the size of molecular

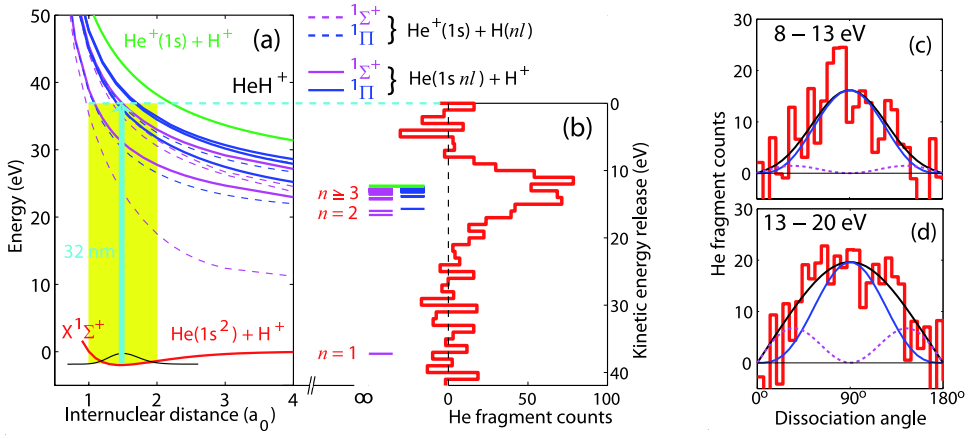

Figure 1: FLASH photodissociation of HeH^+ at 32 nm. (a) Predicted potential energy curves of symmetries $^1\Sigma$ (magenta) and $^1\Pi$ (blue) at the photoexcitation energy of $\sim$39 eV. (b) Kinetic energy release distribution derived from He fragment imaging in comparison with the expected He(1snl) final levels below the He^+(1s) limit (green). Symmetries and adiabatic final-state correlations of the potentials as indicated. (c) and (d): Energy release ranges analysed regarding the fragment angle distribution relative to the FLASH polarisation with fitted sum (full black line) of Σ-Π (blue; full) and Σ-Σ (magenta; dashed) transition symmetries.

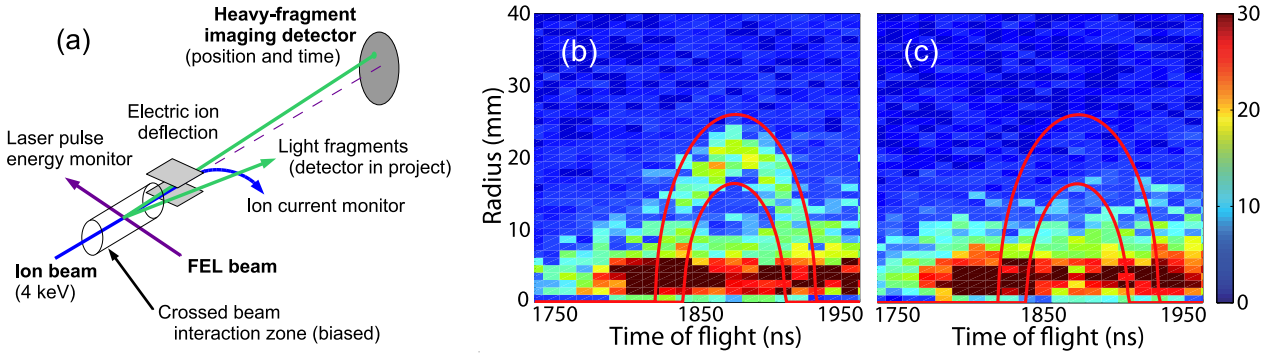

Figure 2: (a) Target region and fast beam momentum system at the TIFF facility. (b) Momentum distribution of He($1snl$) fragments from photodissociation of HeH^+ at 38.7 eV, as extracted from radial positions and arrival times on the imaging detector. Red: Lines for energy releases of 8 and 20 eV, respectively. Net measuring time is roughly 13 h with typical photon pulse energy of 10 μJ. (c) Background spectrum (laser off) with ion-beam related events of small transverse momenta.

contributions [3] in the beta electron energy spectrum, addressed with utmost precision in electron neutrino mass measurements. Moreover, HeH^+ plays a role in astrophysical models [2] of e.g. the early universe and planetary nebula, where in particular photodissociation in the local radiation field represents its major destruction mechanism. As for practically all astrophysically relevant ionic molecular species, photodissociation cross sections and product branching ratios used in modeling these environments for energetic photons are experimentally unexplored and based on estimates mostly.

With a high-transmission photon beam realized behind the monochromator in autumn 2006, photon pulses with ∼5–15 μJ/pulse average power at 32 nm were available, which yielded signal count rates of the order of 10^{-4} to 10^{-3} per pulse for the photodissociation of HeH^+ ions using the fast beam momentum imaging system of Fig. 2(a). In the momentum distribution [Fig. 2(b)] of the neutral He fragments a structure corresponding to an energy release of ∼10–20 eV is clearly identified as a characteristic structure (marked with red lines) in the measured position-time correlation displayed in Fig. 1(b). From measuring fragment counts with and without FLASH, the ion and photon pulse intensities, and the spatial beam overlap, the absolute size of the total photodissociation cross section for HeH^+ at 32 nm on the observed fragment channel was derived to as 1.5×10^{-18} cm^2 with a present systematic error estimate of ±50%. For the energy release spectrum of these data [Fig. 1(b)] the instrumental width is presently estimated to ∼2 eV from the position and time resolution of the imaging system and the so far unmonochromatized photon energy spread. A further broadening toward higher energies arises from vibrationally excited levels in the ion beam. This will be suppressed in future measurements by ion trapping. Even at the present stage it is evident that high Rydberg curves, not yet included in the recent theoretical study [1] nor in astrophysical models [2], yield significant photodissociation into the He + H^+ channel at 32 nm. Moreover, the angular distributions in Fig. 1(c) and (d) show a clear dominance of fragmentation transverse to the photon polarization, which should mainly proceed via the $^1\Pi$ curves, while theoretical studies have so far emphasized parallel transistions via the $^1\Sigma$ states. — Improvements of the fragment energy resolution and an additional detector system for light fragments (such as H and H^+ from HeH^+) are underway; ion trapping at TIFF is operational and schemes for photoelectron detection are in progress.

References

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