

Probing the transition from molecular to atomic photo-absorption with XUV FEL radiation

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The chemical environment of an atom significantly changes its behaviour in photo-absorption. This might be most obvious for wavelengths that correspond to excitation and ionization of valence electrons, but it holds at higher photon energies, where inner shells are predominantly addressed.

We investigated the photo-ionization of $4d$ electrons from iodine in small molecules. The emerging fragments (ions and electrons) have been detected with a reaction microscope to record their energy and angular distributions. We used intense XUV pulses around 90 eV from the free-electron laser in Hamburg (FLASH) to enable multi-photon processes.

IR-pump–XUV-probe: In a first experiment an infrared (IR) laser dissociated diatomic iodine molecules thus creating individual iodine atoms before they were exposed to the XUV radiation. The results can be compared to those for non-dissociated molecules (XUV arrives before IR): For high sum charges $Q_1 + Q_2 > 6$ or asymmetric charge sharing $|Q_1 - Q_2| > 1$, larger yields were observed for dissociated molecules. In the opposite case, low sum charges with symmetric charge sharing exhibit higher count rates for non-dissociated molecules. In conclusion, dissociating the molecule with the IR laser prior to XUV irradiation increases the creation of higher charge states.

XUV-pump–XUV-probe: By adjusting the time delay between the dissociating pulse and the ionization pulse we study XUV photo-absorption of I_2 as a function of the internuclear separation, and, hence, along the transition from molecular to atomic photo-ionization. We show the internuclear-distance dependent formation of ion pairs with asymmetric charge sharing. Additionally, we have extended the investigations to methyl iodide (CH_3I), where electrons have been recorded in addition to the ionic fragments, probing the transition from molecular to atomic Auger electron emission.