

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. In pump–probe experiments performed at the free-electron laser in Hamburg (FLASH) we studied the absorption of multiple 90 eV photons by iodine atoms in I₂ molecules. The pump pulse dissociates the molecule, leading to an increase in internuclear separation and hence the isolation of the iodine atom, while the probe pulse photo-ionizes iodine along this molecule-to-atom transition. The emerging fragments (ions and electrons) have been detected with a reaction microscope to record their energy and angular distributions.

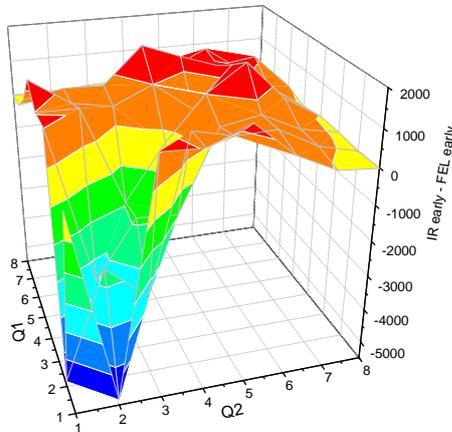


Figure 1: Results of the IR-pump–XUV-probe experiment on I₂: Shown is the count rate for the ion pair (I^{Q₁+}, I^{Q₂+}) when the IR precedes the XUV pulse minus the rate for the case where the XUV hits the molecule before the IR pulse.

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). In fig. 1 the count rate difference is plotted as a function of the charge states Q_1 and Q_2 of the two detected ions. A positive value corresponds to a larger yield for dissociated molecules, which is the case for high sum charges $Q_1 + Q_2 > 6$ or asymmetric charge sharing $|Q_1 - Q_2| > 1$. On the other hand, low sum charges with symmetric charge sharing display a negative value, meaning a

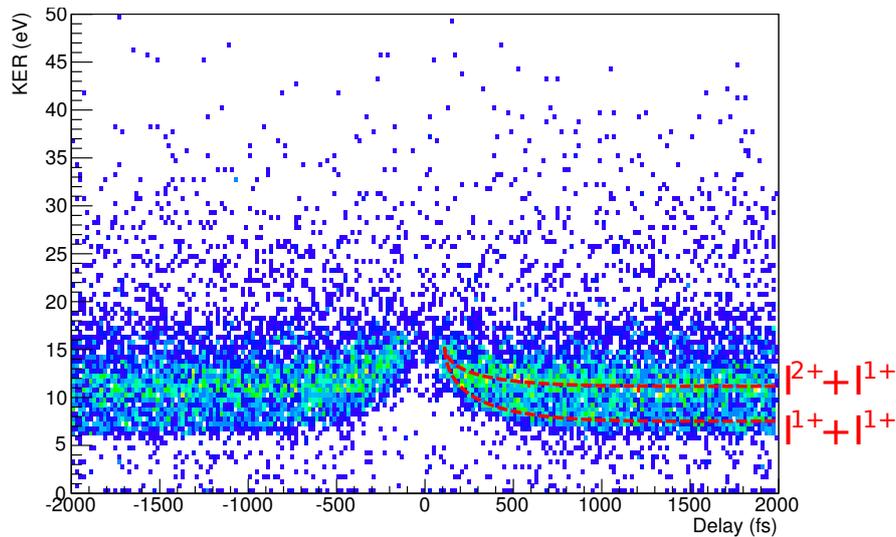


Figure 2: Measured kinetic energy release of coincident I^+ and I^{4+} ions as a function of the delay between pump and probe pulse. Two different precursor dissociation channels $I^+ + I^+$ and $I^+ + I^{2+}$ can be identified.

larger yield for non-dissociated molecules. In conclusion, dissociating the molecule 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. As is shown exemplarily for the $I^+ + I^{4+}$ dissociation channel, we observe the internuclear-distance dependent formation of ion pairs with asymmetric charge sharing: No counts are observed at time delays below 150 fs. This can be understood such that the two nuclei have to be separated by approximately 13 a.u. to allow the creation of this asymmetric charge pair.