CFEL SCIENCE

SEMINAR

1st July 2021 - 10:00 h

Zoom virtual meeting https://desy.zoom.us/j/91202137161

(Meeting-ID: 912 0213 7161, Password: 845021)

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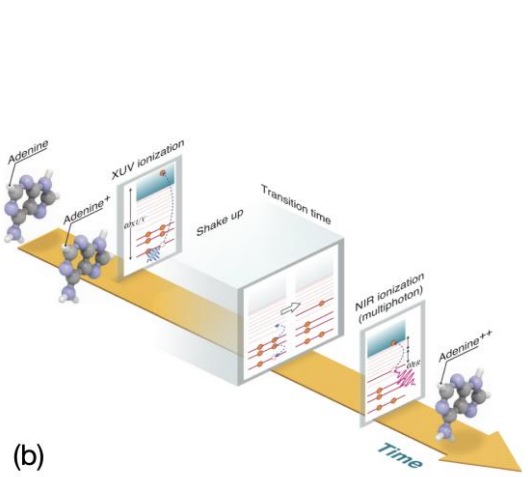
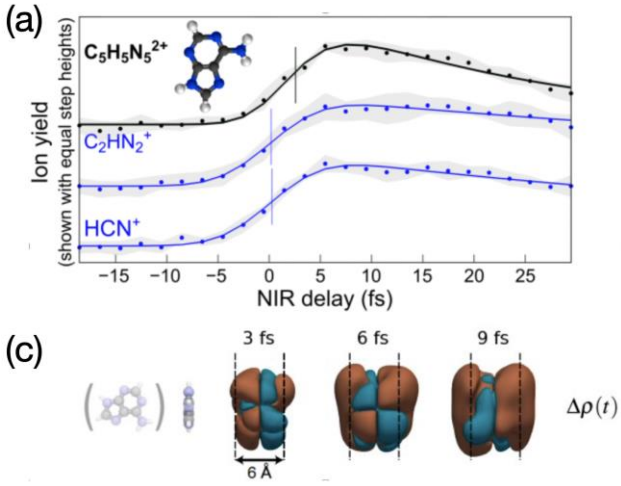
Sub-3f Correlation-Driven Charge Migration in Ionized Adenine

I will present a time-resolved study of photo-fragmentation following ionization by an isolated attosecond XUV pulse (20–35 eV) of the nucleobase adenine, one of the building blocks of DNA. The most intriguing experimental finding is the direct observation of an XUV induced sub-3fs dynamics that leads to the formation of doubly ionized adenine. This can be seen in Fig. (a), which shows the mass spectra as a function of time-delay with respect to an NIR probe pulse. Without the properly timed NIR pulse, the singly or doubly photoionized adenine mainly undergoes fragmentation, as confirmed by theoretical simulations based on Time-Dependent Density Functional Theory combined with Ehrenfest dynamics. We explain the need for a delay between NIR and XUV pulses as the time it takes for electronic correlation to bring the molecule into a state where the NIR pulse is able to further ionize the molecule, Fig. (b).

We find that the production of intact doubly charged adenine, via a shortly-delayed laser-induced second ionisation event, represents the signature of a charge inflation mechanism resulting from many-body excitation, Fig. (c). We obtain qualitatively agreeing time scales from a simple rate equation approach and from ab-initio many-body time-dependent dynamics based on Green's function theory. We identify the relevant mechanism as a shake-up process to an excited state with a delocalised electronic distribution enhancing the cross section for NIR photoionization, from which 2-NIR-photon ionization is energetically feasible. In summary we found that an ultrafast correlation-driven electronic process can determine the destiny of the nucleobase reactivity on much longer time scales.





Hosts: Francesca Calegari / CFEL Molecular and Ultrafast Science Seminar