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Seeing chemical interactions and dynamics with time-resolved X-ray spectroscopy

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Photochemically activated molecules catalyze chemical reactions and a number of time-resolved probes is commonly used for detailed insight into the reaction mechanisms. An orbital-level understanding, however, of how molecules transform and how short-lived and reactive intermediates catalyze reactions has remained limited to date. This is due to the limitations of established experimental probes for characterizing electronic-structure changes, down to the orbital level, as a function of time during chemical reactions.

The aim of this lecture is to discuss the new opportunities provided by time-resolved X-ray spectroscopy at X-ray free-electron lasers and with synchrotron as well as with lab-based short-pulse X-ray sources to reveal the dynamical evolution of chemical interactions. It will be demonstrated how time-resolved X-ray spectroscopy enables probing chemical interactions on atomic length and time scales of Ångströms and femtoseconds. The question about “Where are the electrons?” during a chemical reaction can be answered by addressing the valence electrons or valence orbitals and their evolution as the reaction proceeds. This, in turn, gives us the opportunity to test predictions of quantum-chemical calculations for the first time.

Examples span from small molecules in the gas phase to metal complexes and photo-catalysts in solution and they are used to demonstrate how nuclear dynamics and transient electronic structures are coupled in the electronic excited states of molecular systems and what it is that determines photocatalytic activity.