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Pushing time-resolved extreme UV and soft X-ray spectroscopy towards bigger molecules and condensed phase

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Abstract

Time-resolved spectroscopy has proven invaluable in tracking short-lived intermediates in chemical reactions. Since formation and breaking of chemical bonds happens on the time-scale of vibrational motions, femtosecond time-resolution is required to monitor this

event. Various types of pump-probe spectroscopy with such a capability are used nowadays to elucidate the course of chemical transformations. They typically employ an ultrashort laser light pulse ("pump") to initiate a photochemical process and another laser pulse ("probe") to interrogate the sample after a given time. However, the multitude of processes happening in the first picoseconds after light absorption, e.g., energy re-distribution within the molecule and dissipation to the surrounding, pose difficulty to the unambiguous identification of intermediates. Broadband detection schemes help to disentangle these processes, yet various examples of controversial assignments can be found in the literature.

In this talk, we will show how probe light in the extreme ultraviolet and soft x-ray spectral range can unequivocal identify photochemical processes and chemical species since electronic and nuclear coordinates can readily be distinguished – contrary to the commonly used visible and infrared light. This selectivity stems from the participation of core orbitals in the transitions monitored. In contrast to other groups, we work with low laser field intensities and apply this technique to the study of chemical processes in condensed phase.

Biography

Jakob Grilj has obtained his Ph.D. from University of Geneva (Switzerland) with Eric Vauthey studying the ultrafast excited state dynamics of radical ions in liquid solution. He was awarded a Marie Curie fellowship to work with Markus Guehr at Stanford University (USA) and Majed Chergui at EPFL (Switzerland) on time-resolved extreme ultraviolet and soft x-ray spectroscopy applied to non-Born-Oppenheimer dynamics of coordination compounds.