Exploring Ultrafast Molecular Dynamics in Gas- and Liquid-Phase with Water Window Soft-X-Ray Attosecond Pulses

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Attosecond spectroscopy has revolutionised the study of ultrafast dynamics by enabling direct observation of electronic motion – the fastest mechanism in chemistry – at its intrinsic timescale. Initially confined to the extreme ultraviolet regime, recent advances have extended attosecond pulses into the soft X-ray "water window" (282–533 eV), combining unparalleled temporal resolution with atomic-site specificity detection. In this talk, I will present recent achievements at ETH Zürich using soft-X-ray attosecond transient absorption spectroscopy (ATAS).

I will highlight experiments in gas-phase systems, including the measurement of ultrafast conical intersections in ethylene, revealing electronic relaxation occurring significantly faster than a single vibrational period. Furthermore, I will discuss pioneering liquid-phase experiments, exemplified by proton-transfer dynamics in ionised urea dimers in aqueous solutions. Utilising the site-selective capabilities of tabletop water-window X-ray absorption spectroscopy, supported by advanced quantum-mechanical simulations, I will demonstrate how these ATAS experiments enable the detection of ultrafast charge transfer motion, structural rearrangements, and associated electronic reconfigurations with atomic resolution.

Lastly, I will describe current efforts aimed at achieving the ultimate temporal resolution for initiating and probing electronic coherence, *i.e.*, charge migration. Specifically, I will highlight recent advancements in generating sub-cycle transient strong-field pulses *via* soliton self-compression. This innovative approach expands the toolkit of laser technology, enabling precise control over coherence in matter. When integrated with ATAS, these transient pulses allow the intrinsic complexity of ultrafast electronic processes in molecular systems to be unravelled by simultaneously observing dynamics at multiple atomic edges.