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Probing interfacial electron dynamics with time-resolved X-ray spectroscopy

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Time-resolved core-level spectroscopy techniques using laser pulses to initiate and short X-ray pulses to probe photo-induced processes have the potential to provide electronic state- and atomic site-specific insight into fundamental electron dynamics at complex interfaces. We describe the implementation of femto- and picosecond time-resolved photoelectron spectroscopy at the Linac Coherent Light Source (LCLS) and at the Advanced Light Source (ALS) in order to follow light-driven electron dynamics at dye-semiconductor interfaces on femto- to nanosecond timescales, and from the perspective of individual atomic sites. A distinct transient binding-energy shift of the Ru3d photoemission lines originating from the metal centers of N3 dye-molecules adsorbed on nanoporous ZnO is observed 500 fs after resonant HOMO-LUMO excitation with a visible laser pulse [1]. This dynamical chemical shift is accompanied by a characteristic surface photo-voltage response of the semiconductor substrate [2]. The two phenomena and their correlation will be discussed in the context of electronic bottlenecks for efficient interfacial charge-transfer and possible charge recombination and relaxation pathways leading to the neutralization of the transiently oxidized dye following ultrafast electron injection. First steps towards *in operando* time-resolved X-ray absorption spectroscopy techniques to monitor interfacial chemical dynamics will be presented.

[1] Siefermann *et al.* J. Chem. Phys. Lett **5**, 2753 (2014)
[2] Neppl *et al.* Faraday Discuss. **171**, 219 (2014)