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Electron Dynamics at Dye-Semiconductor Interfaces probed with Picosecond Time-Resolved XPS STEFAN NEPPL, ANDREY SHAVORSKIY, IOANNIS ZEGKINOGLOU, MATTHEW FRAUND, MIQUEL SALMERON, JINGHUA GUO, HENDRIK BLUHM, OLIVER GESSNER, Lawrence Berkeley National Laboratory — Picosecond time-resolved core-level spectroscopy using laser pulses to initiate and short X-ray pulses to probe photo-induced processes have the unique potential to provide electronic state- and atomic site-specific insight into fundamental electron dynamics in complex systems. Up to now, however, most of these experiments have concentrated on the electronic and structural dynamics in isolated or solvated molecules. Here we report preliminary results of a time-resolved X-ray photoelectron spectroscopy (TRXPS) study with the goal to follow the light-driven electron dynamics of N3 dye molecules adsorbed on a nano-structured ZnO semiconductor substrate – a technologically pertinent system for dye-sensitized solar cells – on the pico- to nanosecond time scale from the perspective of individual atomic sites at this complex interface. A distinct evolution of the molecular C1s photoemission line shape is observed as a function of time delay between a visible (532 nm) laser pump pulse (resonant with the N3 HOMO-LUMO gap) and the X-ray probe pulses. The observed changes in the C1s TRXPS spectra will be discussed in the context of possible charge recombination and relaxation processes leading to the neutralization of the transiently oxidized dye following ultrafast photo-induced electron injection.

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