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Towards time-resolved ambient-pressure X-ray photoelectron spectroscopy

Light-induced charge carrier separation, transport, trapping and recombination at semiconductor electrodes are of central importance for the functioning of many photo-electrochemical devices like dye-sensitized solar cells. Here we present a setup that is capable of capturing the dynamics of these processes directly in the time domain, with element specificity and under realistic working conditions using time-resolved ambient-pressure X-ray photoemission spectroscopy (APXPS) techniques at beamline 11.0.2 of the Advanced Light Source.

By combining time-tagging and pump-probe approaches, we are able to take advantage of the full X-ray flux available during multi-bunch operating mode. This highly efficient detection scheme will enable time-resolved APXPS experiments at high background pressures which intrinsically suffer from low photoemission signal levels.

The viability of the time-resolved APXPS setup is demonstrated by resolving transient charge carrier dynamics in nano-porous ZnO electrodes in response to visible laser excitation: for bare ZnO substrates, ultrafast impurity-state mediated electron-hole pair recombination on the nanosecond timescale is observed at the surface, whereas charge carrier relaxation in dye-sensitized electrodes evolves much slower over several microseconds.

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