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Time-resolved X-ray absorption studies on charge carrier dynamics in aqueous TiO2 nanoparticles

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Significant effort has been put into understanding electron/hole dynamics in electronically excited TiO2 nanoparticles (NPs) and how the kinetics of the charge carriers are affected by changes in the electronic and structural properties of such systems. This is necessary in order to understand the underlying mechanisms occurring in photocatalysis and in dye-sensitized solar cells, for which TiO2 is the main material. Here we present a new approach investigating the charge carrier dynamics in nanoparticles using our recently developed high repetition-rate pump-probe XAS setup at the Swiss Light Source, pumping at 355 nm and probing the spectral changes at the Ti K-edge (4.9 keV), which show how the electron dynamics affect not only the electronic but also the structural properties of colloidal TiO2 NPs. The transient Ti K-edge spectrum shows significant changes in the pre-edge region below 4.981 keV, which contain information on the conduction band states of the semiconductor NP. There is a contribution due to the shift of the edge to lower energies, pointing to a change from Ti4+ to Ti3+, and indicating the presence of a trapped electron at the Ti site. The EXAFS region also indicate a change of structure around the Ti atom, which points to a transition from anatase towards a more disordered local geometry. These results underscore the correlation between electronic relaxation and the electronic and geometrical structural changes in the NPs.

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