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Time-resolved X-ray probing of charge transfer dynamics in plasmonic light-harvesting systems with single-electron sensitivity MATTHEW FRAUND, MARIO BORGWARDT, JOHANNES MAHL, FELIX BRAUSSE, Lawrence Berkeley Laboratory, FRIEDRICH ROTH, WOLFGANG EBERHARDT, Center for Free-Electron Laser Science, OLIVER GESSNER, Lawrence Berkeley Laboratory — The use of plasmonic metal nanoparticles (NPs) to improve the solar light-harvesting efficiencies of semiconductor (SC) systems is an attractive approach towards renewable energy technologies, such as the use of photoelectrochemical (PEC) water splitting to produce storable fuels. Improving the economic viability and efficiency of these systems requires a more complete understanding of underlying photo-induced electron transfer processes. We have used picosecond time-resolved X-ray photoemission spectroscopy (tr-XPS) to study photo-induced electron dynamics in a nanoporous TiO2 film sensitized with 20 nm gold NPs. This technique enables quantitative probing of transient charge distributions from the elementally specific perspectives of donor (AuNP) or acceptor (TiO2) sites. An injection efficiency of ˜2 electrons per NP, corresponding to a photon-to-charge conversion efficiency of ˜0.1% has been observed, lower than previously reported for similar systems. Back electron transfer was also observed, proceeding through a predominant, fast recombination channel on a 60 10 ps timescale, followed by a less prominent pathway, which is completed after approximately 1 ns. Implications for the design of nanoplasmonic light-harvesting systems will be discussed as well as an outlook toward studying PEC water splitting dynamics by in situ tr-XPS.

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