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Ultrafast Carrier Dynamics in Water-Splitting Photocatalysts ELIZABETH CARROLL, OWEN COMPTON, MICHAEL SARAHAN, FRANK OSTERLOH, DELMAR LARSEN, Department of Chemistry, University of California — Semiconductors that photocatalytically split water into hydrogen and oxygen using solar energy represent promising renewable energy technologies. Charge carrier dynamics on sub-nanosecond timescales dictate the overall efficiency of charge separation, and consequently, photocatalytic activity of these materials. Using transient absorption spectroscopy, we investigated photoexcited carrier trapping, recombination, and interfacial charge transfer dynamics in water-splitting niobate nanosheets. $Ca_2Nb_3O_{10}$ generated H_2 from water with 0.22% quantum yield, but no O_2 was formed. Carriers relaxed with second-order kinetics on a sub-nanosecond time scale that depended on the nanosheet size. Methanol was used as a sacrificial donor to separate electron and hole dynamics and identify absorption spectra for the trapped carriers. In aqueous methanol, hole scavenging was observed within 100 ps in direct competition with electron-hole recombination. The nanosheets were also functionalized with metal and semiconductor nanoparticles to form novel photocatalyst nanostructures. Colloidal IrO_2 was attached to $Ca_2Nb_3O_{10}$ to sensitize the catalyst for visible light absorption. Interfacial electron transfer stabilized the charge separation in IrO_2, and O_2 was generated.

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