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Ultrafast Structural Dynamics of Photoactive Metal Complexes in Solar Energy Conversion.[1]

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The photoexcited states of metal complexes are precursors for many important photochemical processes in solution phase. Using laser-initiated time-resolved x-ray absorption spectroscopy (LITR-XAS), transient metal oxidation states, coordination geometry, and atomic rearrangements that closely reflect photochemical processes can be probed, which complements with ultrafast optical laser spectroscopic studies for kinetics and coherence among different excited states as well as intra- and intermolecular energy/charge transfer processes associated with solar energy conversion. We have studied by LITR-XAS combined with transient absorption spectroscopy excited state structures, such as metalloporphyrins and platinum(II) complexes, in solution. Direct evidence of photoinduced redox reactions and coordination geometry changes have been observed. These experimental studies are combined with time-dependent density functional theory (TDDFT) calculations to rationalize the excited state structural nuclear changes with electronic configuration changes that may be responsible for the reactivity of the molecules. These studies will have a great impact in fundamental understanding of solar fuel production.

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