Structural Dynamics Challenges in Photosynthesis and Molecular Solar Fuels Catalysis

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Central challenges in solar energy research lie in deciphering the fundamental mechanisms operating in biological and biomimetic modular architectures, and understanding how these are used to tune local site function and to optimally couple photosynthetic modules for efficient solar energy conversion function. Synchrotron x-ray light source developments have offered new opportunities to resolute ground and excited-state structural dynamics for biomimetic photosynthetic assemblies using a range of elastic, inelastic, and anomalous in-situ scattering techniques. We have demonstrated that wide-angle scattering provides a sensitive monitor of supramolecular conformation in solution that can be quantitatively compared to molecular dynamics and other simulation to provide a coordinate model of the coordinate landscape present in experiment. These capabilities have been extended by developing pump-probe, monochromatic x-ray scattering techniques with 100 ps time-resolution Advanced Photon Source. We have used this approach to measure time-dependent inner-and outer-sphere reorganization associated with excited-state charge transfer in donor-acceptor complexes designed for function in solar photocatalysis applications. This work suggests current light source limitations and opportunities for structural dynamics enhancement through development of light sources for photo-excited structure mapping using short pulsed x-rays across a broad x-ray energy range, including tunable hard x-ray energies for combined high-resolution spectroscopy, anomalous scattering experiments, and high-energy pulsed x-rays for high-resolution time-resolved pair distribution function (PDF) analyses.