

APR10-2009-020081

Abstract for an Invited Paper
for the APR10 Meeting of
the American Physical Society

Light Driven Energy Research at LCLS: Planned Pump-Probe X-ray Spectroscopy Studies on Photosynthetic Water Splitting

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Arguably the most important chemical reaction on earth is the photosynthetic splitting of water to molecular oxygen by the Mn-containing oxygen-evolving complex (Mn-OEC) in the protein known as photosystem II (PSII). It is this reaction which has, over the course of some 3.8 billion years, gradually filled our atmosphere with O₂ and consequently enabled and sustained the evolution of complex aerobic life. Coupled to the reduction of carbon dioxide, biological photosynthesis contributes foodstuffs for nutrition while recycling CO₂ from the atmosphere and replacing it with O₂. By utilizing sunlight to power these energy-requiring reactions, photosynthesis also serves as a model for addressing societal energy needs as we enter an era of diminishing fossil hydrocarbon resources. Understanding, at the molecular level, the dynamics and mechanism of how nature has solved this problem is of fundamental importance and could be critical to aid in the design of manufactured devices to accomplish the conversion of sunlight into useful electrochemical energy and transportable fuel in the foreseeable future. In order to understand the photosynthetic splitting of water by the Mn-OEC we need to be able to follow the reaction in real time at an atomic level. A powerful probe to study the electronic and molecular structure of the Mn-OEC is x-ray spectroscopy. Here, in particular x-ray emission spectroscopy (XES) has two crucial qualities for LCLS based time-dependent pump-probe studies of the Mn-OEC: a) it directly probes the Mn oxidation state and ligation, b) it can be performed with wavelength dispersive optics to avoid the necessity of scanning in pump probe experiments. Recent results and the planned time dependent experiments at LCLS will be discussed.