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Taking Snapshots of Water Oxidation Reaction in Photosystem II at X-ray Free Electron Lasers

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The development of XFELs has opened up opportunities for studying the dynamics of catalysis and biological enzymes. Intense XFEL pulses enable us to apply both X-ray diffraction and X-ray spectroscopic techniques to dilute systems or small protein crystals. By taking advantage of ultra-bright femtosecond X-ray pulses, one can also collect the data under functional conditions of temperature and pressure, in a time-resolved manner, after initiating reactions, and follow the chemical dynamics during catalytic reactions and electron transfer.

We have developed spectroscopy and diffraction techniques necessary to fully utilize the capability of the XFEL X-rays for a wide variety of metalloenzymes, and to study their chemistry under functional conditions. One of such methods is simultaneous data collection for X-ray crystallography and X-ray spectroscopy, to look at the overall structural changes of proteins and the chemical changes at metal catalytic sites. The sample is photochemically or chemically activated at various time delays to capture reaction intermediates with crystallography and spectroscopy.

We have used the above techniques to study photochemical activation of the water oxidation reaction of the Photosystem II (PSII) multi subunit protein complex, in which the Mn_4CaO_5 cluster catalyzes the reaction. We report the light-induced structure and electronic state changes of the intermediates during the catalysis. The current status of this research and the mechanistic understanding of this metalloenzyme based on the X-ray techniques is presented.

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