

X-ray Transient Absorption (XTA) Spectroscopy: from Molecular Dynamics to Solar Energy Research

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Transient molecular structures along chemical reaction pathways are important for predicting molecular reactivity, understanding reaction mechanisms, as well as controlling reaction pathways. During the past decade, x-ray transient absorption spectroscopy (XTA), analogous to commonly used optical transient absorption spectroscopy, has been developed. XTA uses a laser pulse to trigger a fundamental chemical process, and an x-ray pulse(s) to probe transient structures as a function of the time delay between the pump and probe pulses. Using x-ray pulses with high photon flux from synchrotron sources, transient electronic and molecular structures of metal complexes have been studied in disordered media from homogeneous solutions to heterogeneous solution-solid interfaces. Examples from the studies at the Advanced Photon Source (APS) in Argonne National Laboratory are summarized, including excited state metalloporphyrins, metal-to-ligand-charge-transfer (MLCT) states of transition metal complexes, and charge transfer state of metal complexes at the interface with semiconductor nanoparticles. Recent developments of the method are briefly described followed by a future prospective of XTA. We envision that concurrent developments in x-ray free electron lasers and synchrotron x-ray facilities as well as other table-top laser driven fs x-ray sources will make many breakthroughs and realize our dreams of visualizing molecular movies and snapshots, which ultimately enable us to control chemical reaction pathways.

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