

Ultrafast dynamics in light-excited transition metal complexes revealed with hard X-ray spectroscopy

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Abstract

Photo-induced transformations of molecular systems are ubiquitous in various branches of chemistry, physics, molecular biology, and materials science. They are essential in photosynthesis and photocatalysis, and have high potential for molecular storage or switching devices, and light-harvesting systems. In order to design better performing functional molecules, understanding the elementary steps and the formation of transient species in related molecular reactions, phase transitions or biochemical function is inevitable. However, the traditional tool set of pump-probe experiments has several limitations, preventing us from capturing many relevant details needed to fully understand the underlying ultrafast dynamics.

In principle all IXS tools can be utilized to probe the ultrafast dynamics in pump-probe experiments [1, 2]. Nevertheless, synchrotron-based studies make use of picosecond-long pulses, and thus lack the necessary time resolution to unveil the ultrafast processes. The intense femtosecond X-ray pulses of X-ray free electron lasers permit us to exploit the X-ray spectroscopy tools with the appropriate time resolution, offering direct access to the changes in the charge, spin and nuclear degrees of freedom during the elementary physical processes of a chemical reaction, photophysical transformation, or biological function. We report on the implementation of hard X-ray spectroscopies in such time-resolved experiments, as well as their combination with X-ray diffuse scattering, which allows us to simultaneously address both the electronic and structural dynamics. Results obtained on light-excited transition-metal-based model systems for photoswitchable or light-harvesting functional molecules will be shown as examples.[3, 4]

References

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