Picosecond and Femtosecond X-Ray Absorption Spectroscopy of Photoexcited Solutes in the Condensed Phase

Christian Bressler

Laboratoire de Spectroscopie Ultrarapide, Ecole Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland

Abstract: Time-resolved x-ray absorption fine structure (XAFS) spectroscopy with picosecond temporal resolution is a new method to observe electronic and geometric structures of short-lived reaction intermediates. It combines an intense femtosecond laser source synchronized to the x-ray pulses delivered from a synchrotron. We present key experiments on charge transfer reactions as well as spin-crossover processes in coordination chemistry compounds next to solvation dynamics studies of photogenerated aqueous atomic radicals. These examples emphasize the new observables at hand using XAFS techniques, which include the density of states, full and even partial changes in oxidation state, and internuclear distances with milli-Angström accuracy. New results featuring femtosecond studies on the ultrafast magnetization dynamics of aqueous $Fe(bpy)_3$ reveal how complex spin-flip processes involving two (or more) electrons occur in real-time. Finally, an outlook towards biologically relevant systems stress the high potential of XAFS methods using new femtosecond x-ray sources like free electron lasers (XFELs).