

Ultrafast structural dynamics of solvated organometallic complexes and prospects for studying coherent bi-molecular reactions

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ABSTRACT

Iron pentacarbonyl, $\text{Fe}(\text{CO})_5$, forms weakly bound complexes with a single solvent molecule in alcohol solutions at room temperature. This “pre-assembly” of iron pentacarbonyl with a solvent molecule, in principle, enable concerted ligand substitution reactions, i.e. the simultaneous bond-formation with the associated solvent molecule and the simultaneous dissociation of a carbonyl ligand after photo-initiation of a chemical reactions. Thus, this system may permit measurements of the ultrafast structural dynamics of *bi*-molecular reactions under ambient conditions *without* a diffusive reactant encounter after photo excitation. Because of the likely absence of the reactant diffusion which causes dephasing of photo-induced coherent atomic motions, it might be possible to observe the evolution of coherent atomic wave packets during bi-molecular reactions in solutions at room temperature. Measuring such coherent motions using structural tools is fundamentally important because it would be the first observation of coherence transfer from reactants to product molecules. As a consequence, it might be possible to coherently control not just half-reaction, such as dissociations or isomerizations, but entire bi-molecular reactions under ambient conditions in solution while observing the structural dynamics response.

Such experiments will require a temporal resolution of 100 fs. As a first step toward the goal introduced above, we recently carried out ultrafast x-ray absorption experiments at the Advanced Photon Source, Argonne National Laboratory, 7ID-C. While the x-ray pulse length produced by the synchrotron is about 100 ps, a temporal resolution of 2 ps has been achieved by detecting the x-radiation transmitted through the sample with a streak camera. The iron K-edge shift of a sample of $\text{Fe}(\text{CO})_5$ solvated in ethanol was measured at a repetition rate of 1 kHz. The experimental arrangement is described and the experimental results are discussed. Furthermore, the results demonstrate that single picosecond temporal resolution can be achieved at a synchrotron beam line.