



SEMINAIRE ISMO

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Ultrafast laser and X-ray absorption study of solvation dynamics and metal organic molecules

We applied ultrafast x-ray absorption spectroscopy to study solvation dynamics of aqueous iodide and spin-crossover process in $[\text{Fe}^{\text{II}}(\text{bpy})_3]^{2+}$. In the solvation dynamics study, a femtosecond laser pulse extracts an electron of the iodide ion impulsively then an X-ray pulse, whose time delay can be tuned with respect to the first pump pulse, will take snapshots of the hydration structural changes from iodide to iodine. The structural changes from picosecond to femtosecond during the transition from hydrophilic (I^-) to hydrophobic (I^0) are extracted.

Light-induced spin crossover study of $[\text{Fe}^{\text{II}}(\text{bpy})_3]^{2+}$ using femtosecond X-ray absorption spectroscopy revealed the ultrafast relaxation cascade of excited states from singlet metal-to-ligand-charge-transfer ($^1\text{MLCT}$) to quintet 5T. We unambiguously determined that 5T state is directly populated by triplet MLCT ($^3\text{MLCT}$) in 150 fs. This time scale of ~ 150 fs is also the period of Fe-N vibration period in the HS state, as determined by Raman spectroscopy.

We present our study of multielectron transfer of catalyst-sensitizer assembly using sequential excitation method. Previous studies of catalyst $[\text{Ru}^{\text{II}}(\text{tpy})(\text{bpy})(\text{OH}_2)]^{2+}$ linked to chromophore $[\text{COOEt}]_2(\text{bpy})_2(\text{phen})\text{Ru}^{\text{II}}]^{2+}$ using single excitation transient absorption spectroscopy show that when the chromophore moiety of molecule absorbs photon with energy in MLCT band, it transfers an electron to a neighbour electron acceptor molecule, enabling electron transfer from the catalyst moiety to photooxidized chromophore, thus activating catalytic reaction. Sequential excitation of the molecule is expected to generate Ru^{4+} by charge accumulation at catalyst site. Our preliminary results show interesting and complicated reaction pathways. Future experiment using laser pump X-ray probe technique on this compound will be discussed.

Mardi 3 mars 2015 à 11h
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