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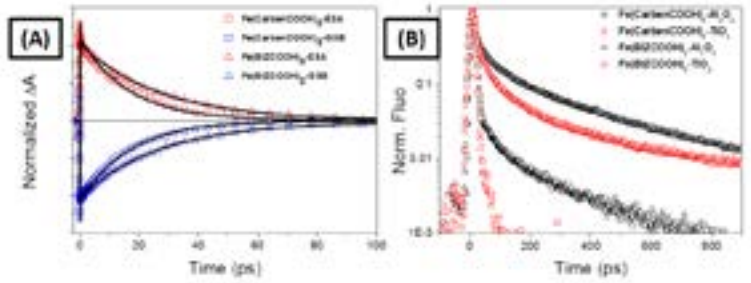
Excited state lifetime of isolated iron(II) complexes and electron injection into TiO_2 film

Over last decades, transition metal complexes (Pt, Ir and Ru) have received great attention due to their long-lived electronic excited states, for applications such as OLEDs and dye-sensitized solar cells (DSSCs). However, the replacement of these expensive, scarce and toxic metals by earth-abundant metals such as Fe or Cu is still extremely challenging due to very short excited state lifetimes of the metal-to-ligand charge transfer states (MLCT), limited, in the case of Fe, by spin cross-over into metal-centered high spin states as observed in Fe-pyridine complexes. A possible way to stabilize the lowest energy triplet ^{3}MLCT energetically and thus temporally is to increase the ligand field strength by chemical design. Recently, the use of N-heterocyclic carbenes (NHC) ligands was reported to lead to a prolonged ^{3}MLCT lifetime (9 ps) which we were able to extend up to 16.5 ps due to carboxylation of the ligands (fig. A). Very recently, the Wärnmark group working on the same carboxylated complex, reported efficient electron injection into TiO_2 by using THz spectroscopy, but also deleterious charge recombination. We have designed new complexes where iron was coordinated by benzimidazolylidene-based (Biz) which affords a new record ^{3}MLCT lifetime of 26 ps in MeCN. Streak camera fluorescence experiments on complexes grafted on TiO_2 and Al_2O_3 substrates indicate electron injection through shorter ^{3}MLCT state lifetime on TiO_2 than on high-band gap Al_2O_3 (fig. B), but also surprisingly long ^{3}MLCT state lifetime components induced only through binding on the semiconductor surfaces. A detailed modeling of the effect of surface attachment on the excited state ordering is in progress.



(A) Kinetic traces of excited state absorption (ΔA>0, red) and ground state bleach (ΔA<0, blue) GSB of Iron complexes. ^{3}MLCT lifetimes are 16.5 ps ($Fe(CarbenCOOH)_2$) and 26ps ($Fe(BizCOOH)_2$) in MeCN. (B) Kinetic traces of ^{3}MLCT of Iron complexes grafted on TiO_2 and Al_2O_3.

Biography

Stefan Haacke is a Professor in Physics at Strasbourg University and Director of the Institute of Physics and Chemistry of Materials Strasbourg. His research on ultrafast spectroscopy of biologic molecules and organic nanostructures has been published in 114 papers. He is Coordinator of the French-German project Femto-ASR.

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