Theoretical Evidence of Solvent-Mediated Excited-State Dynamics in a Functionalized Iron Sensitizer - DTU Orbit (25/02/2019)

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The solvent-mediated excited-state dynamics of the COOH-functionalized Fe-carbene photosensitizer $[\text{Fe(bmicp)}_2]^{2+}$ (bmicp= 2,6-bis(3-methyl-imidazole-1-ylidine)-4-carboxy-pyridine) is studied by time-dependent density functional theory, as well as classical and quantum dynamics simulations. We demonstrate the crucial role of the polar acetonitrile solvent in stabilizing the metal-to-ligand charge transfer (MLCT) states of the investigated molecule using the conductor polarizable continuum model. This leads to dynamics that avoid sub-ps back electron transfer to the metal and an exceptionally long-lived 1MLCT state that does not undergo sub-ps $^{1}\text{MLCT} \rightarrow ^{3}\text{MLCT}$ intersystem crossing as it is energetically isolated. We identify two components of the excited-state solvent reorganization process: an initial rotation (∼300 fs) and diffusional dynamics within the local cage surrounding the rotated solvent molecule (∼2 ps). Finally, it is found that the relaxation of the solvent only slightly affects the excited-state population dynamics of $[\text{Fe(bmicp)}_2]^{2+}$.

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