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Research Summary

The photophysical properties of transition metal-based chromophores, such as ruthenium(II)tris(2,2'-bipyridine), make them a key group of compounds for application in light harvesting processes such as use in Dye Sensitized Solar Cells and photocatalysis. When ruthenium(II)tris(2,2'-bipyridine) absorbs a photon, it is promoted into a metal-to-ligand charge transfer (MLCT) state which creates a separation of charge capable of driving these processes.¹ With light harvesting being a very material intensive process and ruthenium being among the rarest materials on the planet, finding suitable, more abundant alternatives to ruthenium becomes a worthwhile endeavor. A focus of our research group is to study iron(II)-based chromophores as this potential replacement. While iron(II) is isoelectronic to ruthenium(II) and very abundant, iron(II)tris(2,2'-bipyridine) has one major issue; its MLCT state lifetime is not long lasting enough to drive electron transfer or photocatalysis.² This reduction in MLCT lifetime is the cause of the primogenic effect reducing the ligand field splitting in iron(II)-based complexes, which causes metal-centered ligand field excited states to fall below the MLCT state energetically. This allows the MLCT state to rapidly decay to high spin excited states in less than 200 femtoseconds.² One way we seek to resolve this issue is through synthetic design that will disrupt and slow down the relaxation kinetics.³ My research utilizes ultra-fast transient absorption spectroscopy to study the excited state relaxation of various iron(II)-based chromophores to see how various factors, such as altering the π -donating nature of the ligand via substitution, impacts relaxation. By utilizing variable temperature transient spectroscopy, I can determine various factors related to relaxation from the high spin excited state to the low spin ground state, such as activation energy and reorganization energy.⁴ This information can be used to design ligand structures that will disrupt the reaction coordinates that are kinetically relevant to MLCT relaxation.

References:

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