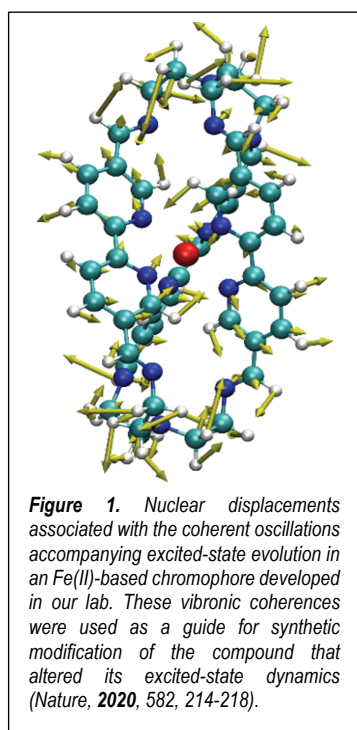


Tailoring the Photophysics of First-row Transition Metal-based Chromophores for Applications in Light-to-Chemical Energy Conversion: Challenges and Opportunities

Professor James K. McCusker

Department of Chemistry, Michigan State University

The interconversion of light and chemical energy is one of the most fundamental processes on Earth. Research on solar energy conversion, for example – which will ultimately lead to the next generation of solar energy technologies – has sought to replicate Nature’s solution through the realization of artificial



constructs that mimic various aspects of photosynthesis. Whether it is the creation of potential gradients to generate current (i.e., photovoltaics) or more recent efforts coupling photo-generated electrons and holes to catalysts (e.g., photoredox catalysis), the critical first step is the absorption of light and the subsequent separation of charge. Transition metal-based chromophores are particularly well-suited for use in such schemes by virtue of the charge-transfer excited-states that a majority of them possess. Indeed, tremendous advances have been made through use of compounds such as $[\text{Ru}(\text{bpy})_3]^{2+}$ and $\text{Ir}(\text{ppy})_3$ in areas ranging from solar energy conversion to photoredox catalysis. Despite the obvious advantages of ruthenium- and iridium-based chromophores, the fact that these elements are among the rarest in the earth’s crust raises legitimate questions concerning cost and scalability of processes reliant on such chromophores. Such issues, coupled with the possibility of unlocking new chemistry, has motivated exploring the possibility of replacing these compounds with

chromophores based on earth-abundant first-row transition metals that can carry out analogous excited-state reaction chemistry. With these opportunities come significant challenges due to inherent differences in the electronic structures of first- versus second- and third-row metal complexes that profoundly impact the ability of such compounds to engage in the desired chemistry. The focus of our research program is therefore to understand the factors that determine the dynamics associated with charge-transfer excited-states of first-row transition metal-based chromophores, with the ultimate goal of circumventing and/or redefining their intrinsic photophysics in order to make feasible their use in a variety of light-driven applications. This seminar will outline the key scientific issues defining this challenge as well as discuss recent examples from our lab illustrating how these challenges can be met and, ultimately, overcome.