

The McCusker Group is interested in the physical and photophysical properties of transition metal complexes. Our approach relies on a confluence of synthetic chemistry, a host of physical techniques ranging from magnetism to femtosecond time-resolved spectroscopy, and high-level theory. The simultaneous examination of chemical problems on all three of these fronts places us in a unique position to explore the physical chemistry of inorganic compounds.

**Ultrafast Spectroscopy of Transition Metal Complexes.** Our research efforts in this area concern the short time scale photo-induced dynamics of transition metal complexes. By “short time scale”, we refer to processes occurring between the time a photon is absorbed by a molecule and the point at which that molecule is fully relaxed in its lowest-lying excited state. Some of the questions we are addressing with this research include the following: (1) what is the general time scale for excited-state evolution in transition metal complexes? (2) what is the mechanism of this process? (3) how do the geometric and electronic structures of the compounds, the surrounding medium, and other factors couple to and/or influence this process? and (4) to what extent can we use this information to control excited-state dynamics? Certain of these questions are very fundamental in nature, whereas others are geared toward work on solar energy conversion. What distinguishes the group, we believe, is our ability to carry out both the synthesis and spectroscopic characterization of a wide range of inorganic molecules. This enables us to systematically examine chemical perturbations to excited-state electronic and

geometric structure, and in so doing develop a comprehensive picture of how transition metal chromophores absorb and dissipate energy.

**Spin and Spin Polarization Effects on Excited-state Dynamics.** Electron spin is a fundamental property of Nature. Although many of the more common physical observables linked to spin are well documented (e.g., magnetism), the degree to which spin and spin polarization influences the chemistry of molecular systems is not as clear. We are pursuing the design and development of chemical systems that will allow us to determine whether there exists a cause-and-effect relationship between the physical and photophysical properties of molecules and their innate spin properties, and if so, to what extent can we exploit this connection in order to manipulate the chemistry of molecular systems. Much of this work centers on the study of so-called donor-acceptor assemblies wherein energy and electron transfer processes are being examined in systems containing spin-coupled paramagnetic fragments. Through careful synthetic manipulation of these compounds, correlations between the observed excited-state reactivity and the involvement of spin-polarized electronic states of the donor and/or acceptor can be realized. Coupled to this experimental work are theoretical studies that exploit recent advances in density functional theory. We believe that these combined efforts will forge an important link between magnetism and electron/energy transfer processes, thereby allowing us to establish a new paradigm in the emerging field of molecular spintronics. 🌐



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## Synthesis and Spectroscopy of Transition Metal Complexes

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### SELECTED PUBLICATIONS

*Leveraging Excited-state Coherence for Synthetic Control of Ultrafast Dynamics*, B.C. Paulus, S.L. Adelman, L.L. Jamula, and J.K. McCusker, *Nature* **2020**, in press.

*Outer-sphere Effects on the Ligand-field Excited-state Dynamics: Solvent Dependence of High-spin to Low-spin Conversion in  $[\text{Fe}(\text{bpy})_3]^{2+}$* , J.N. Miller and J.K. McCusker, *Chem. Sci.* **2020**, *11*, 5191 - 5204.

*The Influence of Electrolyte Composition on Ultrafast Interfacial Electron Transfer in Fe-sensitized  $\text{TiO}_2$ -based Solar Cells*, C.R. Tichnell, J.N. Miller, C. Liu, S. Mukherjee, E. Jakubikova, and J.K. McCusker, *J. Phys. Chem. C* **2020**, *124*, 1794 - 1811.

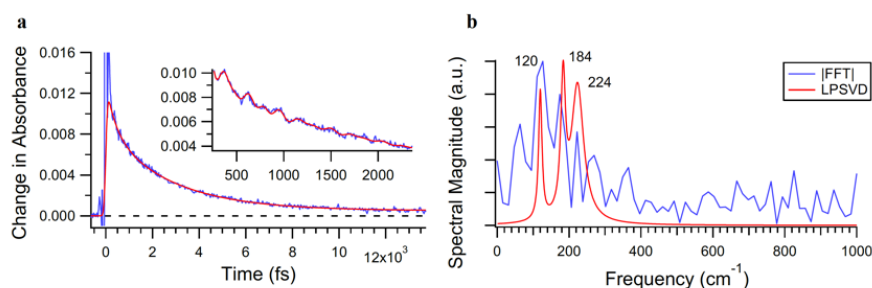
*Electronic Structure in the Transition-metal Block and its Implications for Light Harvesting*, J.K. McCusker, *Science* **2019**, *363*, 484-488.

*Insights into the Excited-state Dynamics of Fe(II) Polypyridyl Complexes from Variable-temperature Ultrafast Spectroscopy*, M.C. Carey, S.L. Adelman, and J.K. McCusker, *Chem. Sci.* **2019**, *10*, 134-144.

*Utilizing Coherence to Enhance Function in Chemical and Biophysical Systems*, G.D. Scholes, G.R. Fleming, L.X. Chen, J.K. McCusker, et al., *Nature* **2017**, *543*, 647-656.

*Photosensitized, Energy Transfer-mediated Organometallic Catalysis through Electronically Excited Ni(II)*, E.R. Welin, C. Le, D.M. Arias-Rotondo, J.K. McCusker, and D.W.C. MacMillan, *Science* **2017**, *355*, 380-385.

*The Photophysics of Photoredox Catalysis: A Roadmap for Catalyst Design*, D.M. Arias-Rotondo and J.K. McCusker, *Chem. Soc. Rev.* **2016**, *45*, 5803-5820 (cover article).



**c** Coherent Vibrational Modes of **2**

Frequency, $\text{cm}^{-1}$	116 (3)	183 (2)	220 (4)
Dephasing Time, fs	700 (190)	770 (300)	580 (300)

