**Beyond Two Component Fe-Catalyzed Cross-Couplings**

**Abstract:** Despite advances in high-throughput screening methods leading to a surge in the discovery of catalytic reactions, our knowledge of the molecular-level interactions in the rate- and selectivity-determining steps of catalytic reactions, especially those involving highly unstable and reactive open-shell intermediates, is rudimentary. These knowledge gaps prevent control, suppression or enhancement, of competing reaction channels that can drive development of unprecedented catalytic reactions. In this talk, I will focus on our use of high-level quantum mechanical calculations, rigorously calibrated against experimental data, to interrogate the mechanisms of asymmetric iron-catalyzed C(sp2)-C(sp3) cross-coupling reactions. Then, I will focus on how our group used this combined experimental and computational approach to quickly develop a vast array of multicomponent cross-coupling reactions.