Abstract: Metalloenzymes facilitate some of the most thermodynamically and kinetically challenging chemical transformation known, and in contrast to our industrial methods, they do so under extremely mild conditions. What are they doing that we are not? One aspect of metalloenzyme reactivity that is ubiquitous in nature yet often overlooked in the development of synthetic systems is the role that conformational gating steps play in mediating metallocofactor reactivity. Inspired by these enzymes, research in the Olshansky lab is focused on testing the hypothesis that conformational control represents an efficient means with which to control metal ion reactivity, promote electron and proton transfer steps, and mediate the interconversion of different forms of energy. However, naturally occurring metalloenzymes are extremely complex and studying them directly, it is often impossible to disentangle the interplay between conformation and reactivity, so instead, the Olshansky lab prepares and studies simplified model systems encapsulating this interplay. Through the construction of switchable artificial metalloproteins and coordination complexes, we are working to dissect the kinetic and thermodynamic consequences of conformational control, and we are preparing an array of switchable systems for applications in solar energy conversion, catalysis, and biomedicine.