Special Seminar

Thursday, Dec. 9th
1:30 p.m., Seminar Hall

Dr. Jillian Dempsey

California Institute of Technology

"Mechanisms of Hydrogen Evolution Catalyzed by Cobaloximes"

Hydrogen is likely to be a key component of our future energy economy, but a low cost and energy efficient generation of this high energy molecular fuel is still a significant challenge. Cobalt(II) complexes with diglyoxime ligands (cobaloximes) are recognized as one of the most promising classes of molecules capable of efficiently catalyzing the reduction of protons to H_2. Several catalytic reaction mechanisms have been proposed that begin with protonation of a reduced Co^{I} complex to form $Co^{III}H$; H_2 evolution can occur via protonation of $Co^{III}H$ or upon bimolecular combination of two $Co^{III}H$ species. Alternatively, $Co^{III}H$ can be reduced further to form $Co^{II}H$, which can react via similar heterolytic or homolytic routes. Laser flash-quench and excited-state proton transfer triggering methods, together with time-resolved spectroscopic monitoring, have allowed us to map out the kinetics of key steps and to identify reactive intermediates in the catalytic cycle. These experiments have allowed us to glean key insights into the reaction mechanisms at play, and opened the way for the design and construction of a second generation of more active catalysts.