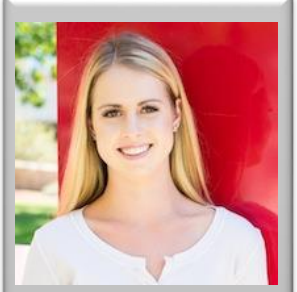

COMPUTATIONAL INVESTIGATION OF QUANTUM MOTION IN SIMPLE AND COMPLEX HYDROGEN CONTAINING COMPLEXES

Jessica Anne Schulze (Ryan Steele)

Department of Chemistry

The objective of this research project is to investigate the role of quantum mechanical molecular motion. Motion was investigated originally in a simple molecule, methanol, and then later investigated in a potential hydrogenation catalyst, $FeH(H_2)(PH_3)_4^+$. Motion is imperative to describing chemical reactions. Hydrogenation is one of the most ubiquitous chemical processes in both industrial and academic chemical synthesis, yet fundamental questions remain unanswered regarding hydrogenation mechanisms. In particular, the inherent molecular motion of metal hydrides (M-H) and "sigma" complexes (M-H_2)—as well as complexes that straddle these two limiting regimes—is conjectured to play a critical role in the efficacy of hydrogenation catalysts but is often challenging to measure experimentally. This project will, instead, utilize high-accuracy computer simulations of methanol and one representative complex, $FeH(H_2)(PH_3)_4^+$, in order to determine (a) the inherent quantum molecular motion and, in the former case, (b) the role of this motion in affecting hydrogenation activity. The iron complex is an example of a molecule that exists as both a metal hydride as well as a sigma complex, and past studies have suggested the possibility of rapid interconversion. Using computer modeling of quantum chemical dynamics, the motion was fully investigated for methanol, and ongoing research continues into the iron complex. For methanol, the alcohol group was found to rotate around the methyl group slowly at room temperature and quite freely at high temperatures. Methanol also exhibited some quantum tunneling effects at lower temperatures, which contribute to its motion. As for the iron complex, the phenomenon of a hydrogen shuttling was found to be a possible motion, as well as dihydrogen rotation and hydride stretching. Further research is being conducted to produce a full picture of all motion, both classical and quantum, which is contributing to this particular iron complex.