Announcing the Final Examination of Dave Austin for the degree of Doctor of Philosophy in Physics

Date: July 5, 2024
Time: 12:00 p.m.
Zoom: zoom link
Dissertation title: FIRST PRINCIPLES STUDIES OF NANOSCALE PHENOMENA AT SURFACES: FROM CHARACTERISTICS OF SINGLE ATOM CATALYSTS TO MOLECULAR STRUCTURE FORMATION

Abstract:
The dissertation focuses on gaining a theoretical understanding of selected phenomena at surfaces that benefit from the novel electronic structure induced at surfaces through the adsorption of atoms and molecules. Of note are the physical and chemical properties of singly dispersed metal atom sites on oxide surfaces that have the potential to be cost-effective catalysts for reactions of technological and fundamental importance. Also of interest are hybrid organic-inorganic interface-driven novel molecular structure formations that display evidence of electron confinement. It utilizes density functional theory (DFT) based calculations to predict and simulate atomic-scale behaviors. The results aim to contribute to understanding reaction mechanisms and factors that enhance catalytic activity and the engineering of functional nanostructures.

One study examines the pathways for water-gas shift reaction for singly dispersed platinum atoms coordinated with a 10-phenanthroline-5,6-dione ligand adsorbed on a titanium oxide surface and compared it to that in the absence of the ligand. Vacancies in the titanium oxide surface are found to be important, and their role in controlling the reaction pathway is delineated. It is shown that the ligand helped stabilize the platinum atom, thus reducing carbon monoxide poisoning and promoting a more efficient reaction that would otherwise have been the case. In another set of studies, vibrational frequencies of carbon monoxide are used to identify the local geometric and electronic structure of singly dispersed platinum atoms on ceria calcinated at two different temperatures, which display marked different propensities for carbon monoxide and ammonia oxidation.

Another study focuses on the electronic structure of bistable molecules with potential applications as a molecular switch. This is motivated by STM measurements, which show two stable configurations of the organic molecule diazodiphenlethane on Cu(111). DFT calculations establish the existence of the two stable configurations, one electrically conductive and the other not. It is shown that these two configurations have very different electronic structures, one showing characteristics of strong hybridization with the surface and the other retaining features of the gas phase molecule.

Similarly, DFT calculations lead to the formation of striking patterns for the overlayer of the organic molecule 4,7-dibromobenzo[c]-1,2,5-thiadiazole on Au(111), whose calculated images provide a rationale for those observed by scanning tunneling microscopy. DFT calculations of the band structure and electronic-structure analysis help unravel a unique pattern known as a Kagome lattice. The molecule's modulation of the Au surface is traced to be the reason for the electron confinement despite a weak interaction between the molecular layer and the Au surface. The above examples lead to the main thesis that the local atomic environment and the accompanying electronic structure are responsible for the ensuing novel properties of these intriguing systems.

Outline of Studies:
Major: Physics

Educational Career:
M. S. University of Central Florida, 2022
B. S. College of Charleston, 2018

Committee in Charge:
Dr. Talat Rahman (Chair)
Dr. Sergey Stolbov
Dr. Mihai Vaida
Dr. Fudong Liu(External Committee Member)

Approved for distribution by Dr. Talat Rahman, Committee Chair, on June 18, 2024.

The public is welcome to attend.