Announcing the Final Examination of Tao Jiang for the degree of Doctor of Philosophy in Physics

Date: Friday, November 8, 2019
Time: 4:00 p.m.
Room: PSB 161
Dissertation title: Catalytic Properties of Defect-Laden 2D Material from First-Principles

Abstract:

Two dimensional (2D) materials offer excellent opportunities for application as catalysts for energy needs. Their catalytic activity depends on the nature of defects, their geometry and their electronic structure. It thus important that the characteristics of defect-laden 2D materials be understood at the microscopic level. My dissertation focuses on theoretical and computational studies of several novel nanoscale materials using state-of-the-art techniques based on density functional theory (DFT) with the objective of understanding the microscopic factors that control material functionality.

My work has helped establish defect-laden hexagonal boron nitride (*dh*-BN) as a promising metal-free catalyst for CO_2 hydrogenation. Firstly I showed how small molecules (H_2, CO, CO_2) interacting with several kinds of defects in *dh*-BN (with nitrogen or boron vacancy, boron substituted for nitrogen, Stone-Wales defect). I analyzed binding energies and electronic structures of adsorption of molecules on *dh*-BN to predict their catalytic activities. Then by computational efforts on reaction pathways and activation energy barriers, I found that vacancies induced in *dh*-BN can effectively activate the CO_2 molecule for hydrogenation, where activation occurs through back-donation to the π^* orbitals of CO_2 from frontier orbitals (defect state) of the *h*-BN sheet localized near a nitrogen vacancy (V_N). Subsequent hydrogenation to formic acid (HCOOH) and methanol (CH_3OH), indicating *dh*-BN (V_N) an excellent metal-free catalyst for CO_2 reduction, which may serve as a solution for global energy and sustainability.

At the same time, I studied critical steps of the catalytic processes from carbon monoxide and methanol to higher alcohol on single-layer MoS_2 functionalized with small Au nanoparticle, indicating C-C coupling feasible on MoS_2-Au_{13}, which led to production of acetaldehyde (CH_3CHO). Whereas a bilayer 31-atom cluster of gold on MoS_2 show excellent catalytic

performance on CO hydrogenation to methanol through two effective pathways: CO* → CHO* → CH_2O* → CH_3O* → CH_3OH* and CO* → CHO* → HCOH* → CH_2OH* → CH_3OH*, where the former pathway showing relatively energetic favorable. In my thesis I discuss the validation of our predictions by experimental observations in the Blair group at UCF.

Outline of Studies:

Major: Physics

Educational Career:

M. S. University of Central Florida, USA, 2015

B. S. Northeastern University, China, 2013

Committee in Charge:

Dr. Talat S. Rahman (Chair)

Dr. Sergey Stolbov

Dr. Laurene Tetard

Dr. Richard G. Blair (External Committee Member)

Approved for distribution by Dr. Talat S. Rahman, Committee Chair, on Oct 31, 2019.