

Announcing the Final Examination of Naseem Ud Din for the degree of Doctor of Philosophy in Physics

Date: November 12, 2020

Time: 10:00 a.m.

Room: [Join Zoom Meeting](#)

<https://us02web.zoom.us/j/7628917416?pwd=OHdHYWk3MTlvV0dsTVh6Y3ZuM0wrdz09>

Meeting ID: 762 891 7416

Passcode: YTq0Z6

Dissertation title: Understanding and predicting properties of low dimensional functional materials from first principles

Abstract:

A periodic network with uniform single metal active site, in coordination with redox-active organic ligands, is a promising class of materials for next generation single atom catalysts. At the same time, two-dimensional transition metal dichalcogenides also offer interesting possibilities for tuning their properties for fundamental understanding and technological applications. This dissertation focus on the one hand on designing metal organic systems with single and multiple metal atom sites coordinated with different organic linkers, and on the other hand on understanding optical properties of two-dimensional molybdenum disulphide. For the first, we have carried out density functional theory (DFT) based calculations of the geometrical and electronic structure and magnetic properties of several transition-metal-organic-chains (TM-C) both in gas phase as well as on Au(111) surface. Of particular interest are dipyridyltetrazine (DT), Bis-pyrimidine (BP), and 1,10-phenanthroline5,6-dione, (PDO) ligands used to design the TM-C with several single TM atoms as the coordination center. We have screened several TM atoms to get their coordination geometry (stable structure) as well as analyzing their chemical activity through adsorption of small molecules on the TM center. Our results suggest that TM atoms with partially occupied d-orbitals exhibit strong affinity, while the TM atoms with fully occupied d-orbitals show weak affinity to the CO and O₂ molecule. We have also investigated the effect of the support (Au(111)) on the geometry and charge state for the V-BP and V-PDO systems to find that the support not only alters the local coordination of TM-Cs, but also has significant charge transfer from TM-C to Au(111). We find that the tetrazine-based ligand, DT, is only able to undergo a two-electron reduction, which limits the complexation to one metal per ligand. In a related study of the complexation of tetraethyltetraaza- anthraquinone (TAAQ) with elemental Fe, leading to complex metal–organic chains, we utilized the multiple binding pockets of TAAQ to achieve higher metal:ligand (M:L) ratios. Our results of various Fe:TAAQ ratio, suggests that thermodynamically one cannot create FeTAAQ species with higher than 2:1.

The second part of this dissertation deals with electronic structure and excitation spectrum of hydrogenated single layer and clean bilayer MoS₂. We calculate the excitation spectrum of single-layer MoS₂ at several hydrogen coverages by using Density-Matrix Time-Dependent Density-Functional Theory (TDDFT). Binding energies of the excitons of the hydrogenated MoS₂ are relatively large (few tens of meV), making their experimental detection facile and suggesting hydrogenation as a knob for tuning the optical properties of single-layer MoS₂. To examine ultrafast charge dynamics in bilayer MoS₂, we applied DFT+Liouville equation approach and found that in conjunction with electron-phonon interaction ultrafast charge dynamics has a strong

effect on the calculated emission spectrum. Our results reveal the importance of ultrafast charge dynamics in understanding photo emissive properties of a few-layer transition-metal dichalcogenide.

Outline of Studies:

Major: Physics

Educational Career:

M. S. University of Central Florida 2017

M.Phil. Quaid-I-Azam University Islamabad Pakistan, 2012

Committee in Charge:

Dr. Talat Rahman (Chair)

Dr. Sergey Stolbov

Dr. William Kaden

Dr. Fernando (External Committee Member)

Approved for distribution by Dr. Talat Rahman, Committee Chair, on November 5, 2020.

The public is welcome to attend [remotely](#).