Announcing the Final Examination of Shree Ram Acharya for the Degree of Doctor of Philosophy in Physics

**Date**: Wednesday, January 17, 2018

**Time**: 3.00 PM

**Room**: PSB 160

**Dissertation title:**

From Excited Charge Dynamics to Cluster Diffusion: Development and Application of Techniques beyond DFT and KMC

## Abstract:

The work presented in this dissertation focuses on developing reliable and accurate computational techniques which enable the examination of static and dynamic properties of various activated phenomena using deterministic and stochastic approaches. To explore the ultrafast electron dynamics in materials with strong electron-electron correlation, under the influence of laser pulse perturbation, an *ab initio* electronic structure method based on time-dependent density functional theory (TDDFT) in combination with dynamical mean field theory (DMFT) is developed and applied to: 1) single-band Hubbard model; 2) multi-band metal Ni; and 3) multi-band insulator MnO. The results for the time-dependent (femtosecond) demagnetization in Ni reveal the importance of non-adiabatic and correlation effects, as their inclusion leads to much closer agreement with experimental data than is achieved by the standard TDLDA method. The results for the d-electron dynamics in MnO identifies the main channels of the charge response. Furthermore, through numerical fitting of the DMFT-based exchange-correlation (XC) kernels for different strength of electron correlation, a simple analytical form is obtained which can be used readily in future applications, saving huge computational cost.

To investigate the temporal and spatial, size-dependent, evolution of homo- and hetero-epitaxial adatom islands on the (111) surface of several fcc transition metals, under thermal perturbation, I apply the self-learning kinetic Monte Carlo (SLKMC) method that explores dynamics for long time being unbiased by a set of diffusion processes chosen *apriori*. A number of novel multi-atom diffusion processes are revealed. Trends in the diffusion coefficients, as a function of island size and elemental composition of the adatom islands versus the substrate, point to the relative role of lateral interaction among the adatom islands and the binding energy of the island to the substrate. Moreover, analysis of a large database of activation energy barriers, obtained with SLKMC, for a multitude of diffusion processes for the variety of systems considered allows extraction of a set of descriptors, which are used to develop predictive models for energy barrier calculation using data-driven approaches.

Finally, for a phenomenon driven by externally controllable temperature and pressure, the kinetics of the industrially important methanol partial oxidation reaction on a model nanocatalyst is explored using the kinetic Monte Carlo (kMC) method supplemented by DFT energetics. The molecular static study explores the active surface sites for reaction components including different intermediates and energetics of competing probable reaction pathways, while kinetic study attends to the selectivity of products and its variation with external factors.

Comparison of the above results with experimental data, where available, validate the methodology that we have developed and implemented.


**Outline of Studies:**

**Major**: Physics

**Educational Career:**

M.Sc. Tribhuvan University, Nepal, 2007.

B.Sc. Tribhuvan University, Nepal, 2004.


**Committee in Charge:**

Dr. Talat S. Rahman (Chair)

Dr. Lee Chow

Dr. Sergey Stolbov

Dr. Annie Wu (External Committee Member)


Approved for distribution by Dr. Talat S. Rahman, committee Chair, on January, 12, 2018.


The public is welcome to attend.