CORE-CM SEMINAR
Michigan State University

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Mode Specificity and Bond Selectivity in Dissociative Chemisorption

Dissociative chemisorption is the initial and often rate-limiting step of many important heterogeneous catalytic processes such as ammonia synthesis, steam reforming, and water gas shift reaction. A complete understanding of these processes at the microscopic level is important for a predictive model of catalysis. Recent molecular beam experiments have indicated strong mode specificity and bond selectivity in the dissociative chemisorption of polyatomic molecules such water and methane. These observations suggest that all forms of energy are not equal in activating the reactions, which as a result cannot be treated with statistical models.

We will discuss quantum dynamical models for dissociative chemisorption of water and methane on high-dimensional potential energy surfaces constructed by accurate fits of many density functional theory points over the relevant configuration space. These first-principles quantum dynamical investigations successfully reproduced, and in some cases predicted, experimental observations, highlighting the importance of dynamics in the experimentally observed mode specificity and bond selectivity. In this talk, we will focus on our recent full-dimensional simulations of water dissociative chemisorption on Ni(111), and discuss the site-specific dynamics and dependence on incident angles.

We further discuss a simple model for predicting mode specificity and bond selectivity in reactions both in the gas phase and between gas molecules and surfaces. This Sudden Vector Projection (SVP) model is based on the premise that the collision is much faster than the intramolecular vibrational energy redistribution (IVR) in the reactants. It further assumes that the ability of a reactant mode in promoting the reaction is proportional to its coupling strength with the reaction coordinate at the transition state. This is approximated by the projection of the reactant normal mode vector onto the reaction coordinate vector. The SVP model is used to interpret and predict mode specificity and bond selectivity in dissociative chemisorption reactions.

Thursday, Sept. 24, 2015
12:00 NOON
Room 1400 – Biomedical & Physical Sciences
Professor Marcos Dantus – Host