

## **Colloquium**

### **Building the World's Greatest Microscope: Revealing the atomic scale dynamics of surface chemistry**

**Alec M Wodtke**

**Georg-August University of Göttingen & Max Planck Institute for Biophysical Chemistry, Germany**

In 1929, Nobel Laureate Paul Dirac made comments to the effect that Chemistry had been solved. With the advent of quantum mechanics “The underlying physical laws necessary for the mathematical theory of... ..the whole of chemistry are... ..completely known.... However, on a practical level computational chemistry is still in an early stage of development. Dirac went on: “the difficulty is only that the exact application of these laws leads to equations much too complicated to be soluble.” Despite electrifying advances in computational power since that time, Dirac is still right. The theory of chemistry requires approximations before theoretical descriptions and predictions of chemical reactions can be made.

The advent of the Born-Oppenheimer Approximation led to the development of the standard model of chemical reactivity where the electronically adiabatic potential energy surface for nuclear motion is derived and quantum motion of the nuclei on that surface can be calculated. For simple gas phase reactions, this approach has become an extraordinarily useful and reliable tool. For surface chemistry, additional approximations are commonly made: 1) classical mechanics for describing nuclear motion, 2) density functional theory (usually at the generalized gradient level) for calculating electronic states, 3) reduced dimensionality approximations and as before 4) the Born-Oppenheimer approximation to separate electronic and nuclear degrees of freedom. I call this collection of approximations the provisional model for surface chemistry as we in the field are still testing and improving it.

In this talk, I will describe how a fruitful interplay between experiment and theory can lead to accurate atomic-scale simulations of simple reactions at metal surfaces. I will describe the very significant challenges surface chemistry presents including the problems of high dimensionality and the common failure of the Born Oppenheimer approximation. I will present three concrete examples. In the first, results of a full dimensional experimentally validated theoretical approach to hydrogen atom adsorption at a metal surface that includes the effects of Born-Oppenheimer failure leads to an atomic scale view of H-atom adsorption at a noble metal as well as an explanation for chemicurrents. In the second, energy loss dynamics occurring on a 25-fs time scale can be inferred from the scattering of hydrogen atoms from a graphene surface where a transient C-H chemical bond is formed. The H interaction on graphene is strongly influenced by the choice of metal substrate upon which the graphene is grown. The third example - CO oxidation on Pt - brings us closer to the real world. Here, I will show some of the first comparisons of experimental rate constants to those predicted from first principles theory.

***Monday, Jan 8<sup>th</sup> 2018***

***4:00 PM (Tea/Coffee at 3:30 PM)***

***Auditorium, TIFR-H***