Tadashi Ogitsu, J Phys Chem Biophys 2018, Volume 8
DOI: 10.4172/2161-0398-C3-034

Atomistic insights into electrochemical interfaces by *ab initio* simulations and *in-situ* characterizations

Tadashi Ogitsu
Quantum Simulation Group - Lawrence Livermore National Laboratory, USA

Recent progresses in *ab-initio* computer simulation techniques as well as the advancement in high performance computing made direct simulations of complex systems such as electrochemical interfaces possible. Faithful modeling of electrochemical interface, however, is still very challenging partly due to lack of experimental probe that provides direct atomistic structural information. However, there are experimental characterization techniques that give us information about local chemical environment. In this presentation, we will first discuss about our recent experience in using *ab-initio* simulations for interpreting ambient-pressure X-ray photoemission spectroscopy (AP-XPS) results on III-V semiconductors (GaP/InP) exposed to chemical agents such as oxygen and/or water. XPS spectrum is usually analyzed based on comparison to reference information available in literature, which is valid if the system of our interest can be well approximated as a linear combination of well-defined reference problems, which is unlikely to be the case for electrochemical systems where local chemical environments tend to be dynamical in nature. *Ab-initio* simulations provide information regarding relation between thermodynamic stability of structural motifs and their spectroscopic signatures, therefore, peak assignment can be performed in a more rational and robust fashion. In addition, one may combine multiple theoretical and experimental spectroscopic information for the same system and may examine the consistency of each analysis. Development of accurate and realistic structural models of complex electrochemical systems will give us atomistic insights on electrochemical processes such as hydrogen/oxygen evolution and/or material corrosions, which in turn, can be used to improve the performances of energy conversion/storage devices.

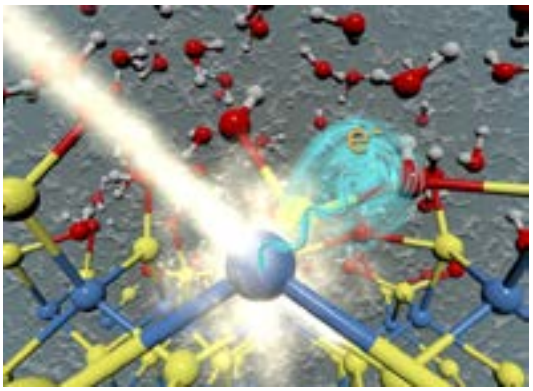

Figure 1: Schematic image of water-semiconductor interface. Some experimental techniques, such as AP-XPS, probe local chemical environment by measuring a perturbation on local electronic structure (ex. core-level binding energy shift). Systematic comparison between simulated spectroscopy for a given structural model and experimental results will enable us to develop reliable models of complex interfacial systems

Recent Publications

1. Pham T A et al. (2018) Integrating *ab initio* simulations and X-ray photoelectron spectroscopy: toward a realistic description of oxidized solid/liquid interfaces. Journal of Physical Chemistry Letters. 9(1):194-203.
2. Pham T A et al. (2016) Salt solutions in carbon nanotubes: the role of cation−π interactions. Journal of Physical Chemistry C. 120(13):7332-7338.
3. Esposito et al. (2015) Methods for photoelectrode characterization with high spatial and temporal resolution. Energy and Environmental Science. 8(10):2863-2885.
4. Wood B C et al. (2014) Surface chemistry of GaP(001) and InP(001) in contact with water. Journal of Physical Chemistry C. 116(2):1062-1070.

5. Wood B C et al. (2013) Hydrogen-bond dynamics of water at the interface with InP/GaP(001) and the implications for photoelectrochemistry. Journal of the American Chemical Society. 135(42):15774-15783.

Biography

Tadashi Ogitsu was awarded PhD in Materials Science from University of Tsukuba, Japan in 1994 and completed his Post-doctoral studies from University of Illinois at Urbana-Champaign in 2001. He is a Deputy Group Leader of Quantum Simulation Group at the Lawrence Livermore National Laboratory and is the point of contact for DOE/EERE HydroGEN consortium which is designed to facilitate sustainable hydrogen production R&D. He has his expertise in *ab-initio* simulations and computational spectroscopy, and is interested in applying these skills and investigates on fundamental aspect of electrochemical processes relevant for energy applications such as photoelectrochemical hydrogen production.

ogitsu@llnl.gov

Notes: