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Complex electrochemical processes elucidated by ab-initio simulations and in-situ characterizations

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Electrochemical processes are ubiquitously seen in problems relevant for our life, yet, extremely challenging to gain an accurate atomistic information due to its complex and dynamical nature. However, recent progresses on ab-initio computer simulation techniques combined with the advancement in high performance computing made direct simulations of complex electrochemical interfaces possible albeit connection to the corresponding real problem often is not apparent. On the other hand, there are many experimental probes that provides chemical and physical information of such problems albeit interpretation of such experimental data tends not to be straightforward. In this presentation, we will demonstrate how a combination of ab-initio simulations and in-operando characterization techniques such as ambient-pressure X-ray photoemission spectroscopy (AP-XPS) can be used to gain atomistic insights into electrochemical problems. We will discuss how AP-XPS data taken for oxidation of III-V semiconductors (GaP/InP) induced by chemical agents such as oxygen and/or water can be interpreted with the help of ab-initio simulations. Most importantly, ab-initio simulations provide information regarding relation between thermodynamic stability of structural models and their spectroscopic signatures that will give us confidence in interpreting the experimental data. This is particularly the case, when comprehensive set of spectroscopic information for the given problem is available. We show that one can effectively narrow down the candidate surface oxidation models based on systematic theory-experiments comparisons on O1s, P2p core-level binding energies, surface workfunction shift as well as their stabilities. Such a comprehensive theory-experiments comparison will lead to deeper understanding of electrochemical processes such as hydrogen/oxygen evolution and/or material corrosions that will facilitate improvements of the energy conversion/storage technologies.

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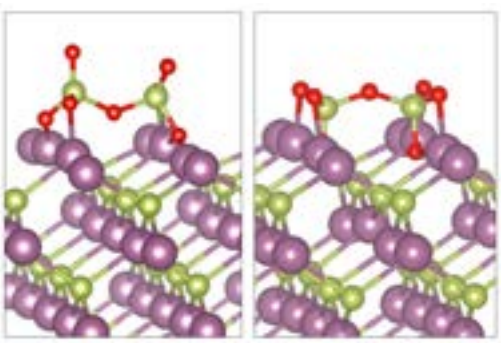

Figure 1: P2O7 isomers on InP(001) surfaces. The structure on the right provides good agreements with the experiments on O1s/P2p core-level binding energy shifts as well as workfunction shift simultaneously. In addition, the structure on the right is significantly more stable than one on the left. Systematic comparison between simulated spectroscopy for series of structural models and experimental results will enable us to understand complex interfacial processes.

Recent Publications

1. Zhang X et al. Revelation of the oxidation mechanism on InP(001) surface by integrated APXPS and ab-initio simulations, submitted.
2. Pham TA et al. (2018) Integrating Ab Initio Simulations and X-ray Photoelectron Spectroscopy: Toward A Realistic Description of Oxidized Solid/Liquid Interfaces, J. Phys. Chem. Lett 9, 194-203.
3. Esposito et al. (2015) Methods for Photoelectrode Characterization with High Spatial and Temporal Resolution, Energy Environmental Science 8, 2863-2885.
4. Wood BC et al. (2014) Surface chemistry of GaP(001) and InP(001) in contact with water, J. Phys. Chem. C 118, 1062-1070.
5. Wood BC et al. (2013) Hydrogen-Bond Dynamics of Water at the Interface with InP/GaP(001) and the Implications for Photoelectrochemistry, J. Am. Chem. Soc. 135, 15774-15783.

Biography

Tadashi Ogitsu has his expertise in ab-initio simulations and computational spectroscopy, and is interested in applying these skills and investigate on fundamental aspect of electrochemical processes relevant for energy applications such as renewable hydrogen production. He is a deputy group leader of Quantum Simulation Group at Lawrence Livermore National Laboratory and is the point of contact for DOE/EERE HydroGEN consortium (www.h2awsm.org), which is designed to facilitate sustainable hydrogen production R&D by providing highly diverse and complemental research capabilities.

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