Discovery of Fundamental Reaction Mechanisms at Electrochemical Interfaces by Quantum Simulations

Florian Deißenbeck, Stefan Wippermann, Mira Todorova and Jörg Neugebauer

Max-Planck-Institut für Eisenforschung Max-Planck-Str. 1, 40237 Düsseldorf, Germany
neugebauer@mpie.de

Direct experimental exploration of atomistic mechanisms governing electrocatalysis or wet corrosion at the solid-liquid interface is notoriously difficult. While fully parameter- free ab initio calculations have the potential to accurately elucidate such mechanisms, they encounter substantial conceptual challenges. Recent methodological breakthroughs, including the advent of efficient computational electrodes and the development of effective thermopotentiostats, overcame many of these limitations thus providing new research opportunities. This presentation outlines the fundamental concepts underlying these innovative methodologies and demonstrates their application in performing fully explicit calculations of structure and chemical reactions at electrified solid-water interfaces. The power of this approach in discovering new mechanisms will be demonstrated for fundamental electrochemical reactions such as metal dissolution and the hydrogen evolution reaction (HER). The identified mechanisms challenge existing understandings and underscore the active involvement of water molecules in electrochemical reactions, challenging the perception of them as passive bystanders.