

M2 Internship

Electrostatic Embedding for hybrid grand-canonical DFT/molecular mechanics simulations of electrified interfaces

Context

The theoretical chemistry group of the Laboratoire de Chimie (LCH) at the ENS de Lyon has a strong expertise in modelling heterogeneous electrocatalysis. In collaboration with experiments carried out at LEPMI (Grenoble) and LISE (Paris), we will advance the understanding of species at electrified interfaces via grand-canonical DFT (LCH) and experimental tip-enhanced Raman scattering data (LISE) on monocrystalline noble metal surfaces (LEPMI). The three partners have obtained an ANR project (TERS_NORMAL), which will provide funding for a PhD for molecular simulations.

Background

Tailoring new catalytic systems requires to understand their reaction mechanisms and thus fundamental studies on model surfaces such as those provided by metallic single crystals. In this project, we will build a library of electrocatalytic surfaces Pd@Pt and Pd@Au that resist CO poisoning. Local measurements by Tip Enhanced Raman Spectroscopy will detect and quantify formic acid decomposition products such as CO and OH adsorbates at different potentials and positions on the substrate. To obtain a global vision, a theoretical chemistry approach that takes into account the specificities of the electrochemical interface will be implemented. It will allow to reconstruct and better interpret experimental spectra. We will finally provide a multiscale description of the reaction.

Project

Computational modelling is ideally suited to study the surface state and reactivity of monocrystalline metallic surfaces under an applied electrochemical potential. LCH has ample experience in modelling electrocatalysis under operating conditions, and in particular including the effect of the electrochemical potential and the electrolyte. [Abidi et al. WIREs Comput Mol Sci. 2021, 11, e1499]. However, in order to improve the description of the electrified interfaces, an atomistic description of the electrolyte in combination with a suitable force-field for the water/metal interaction is preferable compared to the standard implicit solvation models. LCH has recently developed corresponding force fields which lead to good agreement for solvation energies at the metal/water interface [Clabaut et al. J. Chem. Theory Comput. 2020, 16, 6539]. In this internship, we will couple the accurate force fields with electrostatic embedding for GC-DFT. The scheme will be applied to the adsorption of pyridine on Au(111), a well-established benchmark system. [Steinmann and Sautet J. Phys. Chem. C 2016, 120, 5619]

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