PhD: AIMD/DFT-MD for electrochemistry at aqueous interfaces
and coupling to spectroscopies in operando conditions

Keywords : theoretical and computational chemistry, ab initio molecular dynamics, DFT-MD, AIMD, electrochemistry, spectroscopy, Sum Frequency Generation SFG, THz-IR, condensed matter, interfaces, materials

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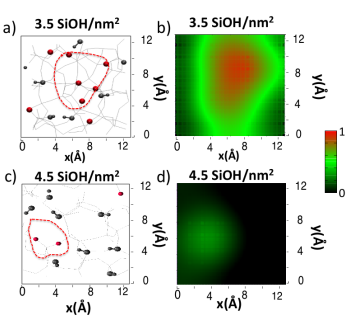
The PhD work will consist in applying ab initio molecular dynamics simulations AIMD/DFT-MD to simulate electrochemical aqueous interfaces in order to extract structural, dynamical and vibrational spectroscopic properties of the interfaces in operando conditions. The central issues in the AIMD simulations are the electrochemical conditions that are non trivial to set-up in the quantum representation, and for which some of the developed theoretical methods will be tested, and the calculation and interpretation of spectroscopic signals such as SFG (Sum Frequency Generation) and THz-IR for which our group is expert. Our simulations will be coupled to spectroscopic experiments performed by some of our collaborators: Prof P. Petersen at the University of Bochum-Germany, Prof M. Havenith at the University of Bochum-Germany, Prof W. Liu and Prof Y.R. Shen at the Fudan University in Shanghai-China and UC Berkeley-USA, and with the electrochemistry group of Prof K. Tschulik at the University of Bochum-Germany. The electrochemical systems of interest are typically related to the CO_2 reduction for improving this electrocatalysis process.
Codes developed in our group are used for the analyses and the extraction of structural/dynamical/spectroscopic/reactivity properties from the trajectories. For those interested, theoretical developments can be considered and/or development of codes for trajectory analyses.

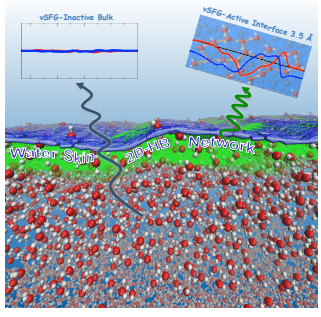
Our group is internationally recognized in AIMD/DFT-MD simulations of solid/liquid interfaces. The following references illustrate some of our recent original works and innovative developments in the field of interfaces and in the field of theoretical SFG spectroscopy [J. Am. Chem. Soc. 142:6991-7000 (2020), PNAS 116:1520(2019), Phys. Chem. Chem. Phys. 21:22188-22202 (2019), J.Chem.Phys. 150:041721(2019), Phys.Chem.Chem.Phys. 20:5190(2018), Minerals 8:305(2018), J.Phys.Chem.Letters 8:3133(2017), J.Phys.Chem.C. 120:14034(2016)].

Our group is composed of 5 permanent academic researchers, 1 Engineer in informatics, 4 PhDs (Chinese, Italians, French), 1 Post-Doc, 2-3 foreign visitors/year. Our group is internationally recognized and funded by several French national ANR and LABEX research schemes, e.g. bilateral ANR-NSF/USA, ANR-DFG/Germany & LABEX CHARM$_3$AT. Our University is a member of the larger University Paris-Saclay.
The city of Evry is located 45' from the center of Paris via RER D train line or 30' from Paris-Saclay via bus. The University is 2' walk from the RER station Evry-Courcouronnes. See https://www.univ-evry.fr/accueil.html for more practical details.

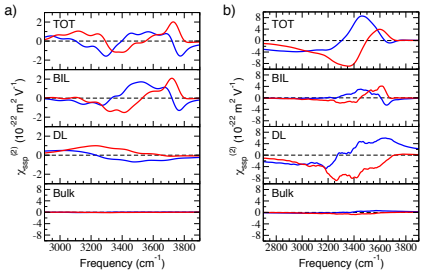
Some illustrations from recent works:



Inhomogeneous patches of hydrophilicity/hydrophobicity at (hydrophilic) SiO_2 silica/liquid water interfaces.
PNAS 116:1520(2019)



A water 2D-H-Bond-network unraveled at the air/water interface by AIMD/DFT-MD simulations.**J.Phys.Chem.Letters 8:3133(2017)**



Deconvolution of SFG signals at solid or air/water interfaces in terms of BIL (Binding Interfacial Layer), DL (Diffuse Layer) & Bulk contributions. **PCCP 20:5190(2018)**

PhD: AIMD/DFT-MD of tunable charged/hydrophilic/hydrophobic solid/water interfaces, in direct relation with interfacial vibrational spectroscopy

Keywords : theoretical and computational chemistry, ab initio molecular dynamics, DFT-MD, AIMD, spectroscopy, Sum Frequency Generation SFG, condensed matter, interfaces, materials,

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The PhD work will consist in applying ab initio molecular dynamics simulations AIMD/DFT-MD to extract structural, dynamical and vibrational spectroscopic properties of tunable charged/hydrophilic/hydrophobic solid/liquid water interfaces. One central issue for the simulations is the calculation of non-linear SFG (Sum Frequency Generation) vibrational spectra, and their interpretation and assignment in terms of structure of the water and of the solid surface at the interface between the two media, in terms of thickness of water being probed experimentally, in terms of chemical reactivity at the interface, especially in electrochemical conditions. As the pH conditions and the presence of electrolytes in the liquid are of high importance for the final structural organization and reactive properties of the interface, these conditions are especially investigated in our works. The theoretical work will be done on oxide/water interfaces, their hydrophilic/hydrophobic behaviors at the molecular level will be revealed as a function of pH, electrolytes concentration, and surface morphology. The DFT-MD simulations are systematically coupled with SFG experiments. These later will be obtained from our regular collaborators, Prof W. Liu and Prof Y.R. Shen at the Fudan University in Shanghai-China and UC Berkeley-USA, Prof P. Petersen at the University of Bochum-Germany, Prof E. Borguet at Temple University-USA. The package CP2K is used for the AIMD simulations, codes developed in our group are used for the analyses and the extraction of structural/dynamical/spectroscopic/reactivity properties from the trajectories. For those interested, theoretical developments can be considered and/or development of codes for trajectory analyses.

Our group is internationally recognized in AIMD/DFT-MD simulations of solid/liquid interfaces. The following references illustrate some of our recent original works and innovative developments in the field of interfaces and in the field of theoretical SFG spectroscopy [J. Am. Chem. Soc. 142:6991-7000 (2020), PNAS 116:1520(2019), Phys. Chem. Chem. Phys. 21:22188-22202 (2019), J.Chem.Phys. 150:041721(2019), Phys.Chem.Chem.Phys. 20:5190(2018), Minerals 8:305(2018), J.Phys.Chem.Letters 8:3133(2017), J.Phys.Chem.C. 120:14034(2016)].

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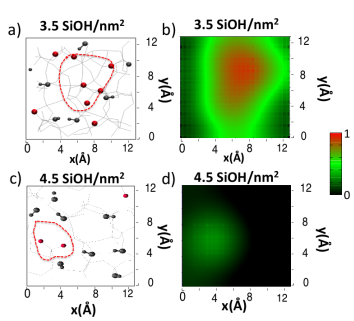

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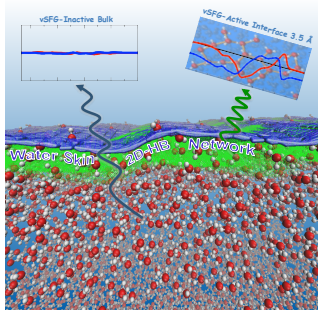

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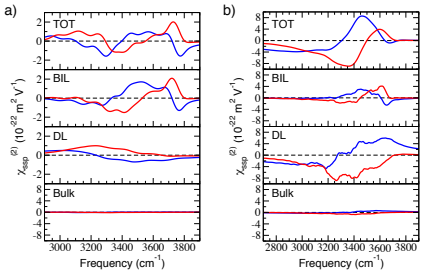

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