PhD: AIMD/DFT-MD for the simulation of 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 metal/liquid water interfaces in operando conditions. The electrochemical conditions are non-trivial to set-up in the quantum representation. There are currently few developed theoretical methods that will be tested and assessed in this work. Our central focus is to make the link between the structure at the interface between the metal surface and the liquid water with SFG (Sum Frequency Generation) and THz-IR spectroscopic signatures that can both be probed at interfaces. Our group is expert in the theoretical modeling of these linear and non-linear spectroscopic signals from AIMD/DFT-MD trajectories. 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 for the applications are typically related to the CO_2 reduction for improving this electrocatalysis process at metal/liquid water interfaces.
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. Developments in synergy with Graph Theory and AI are part of this project for those interested.

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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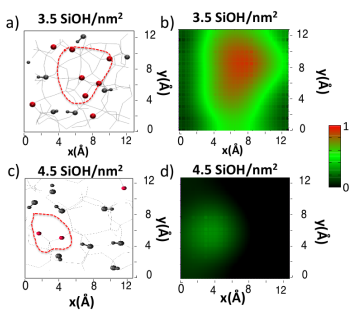
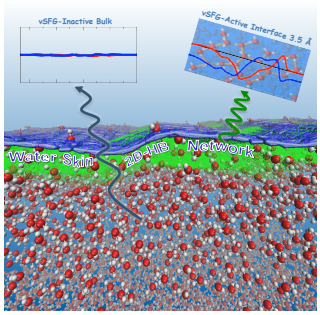
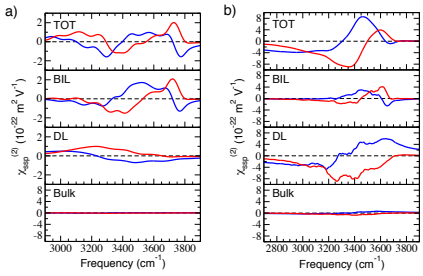
Some illustrations from recent works:



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 and Classical MD of tunable charged/hydrophilic/hydrophobic solid/water interfaces, direct relation with interfacial vibrational spectroscopy

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

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The PhD work will consist in applying ab initio AIMD/DFT-MD and classical CIMD molecular dynamics simulations in order to extract structural, dynamical and vibrational spectroscopic properties of tunable charged/hydrophilic/hydrophobic aqueous interfaces. One central focus 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 both the water and solid surface at the interface between the two media, in terms of thickness of water being probed experimentally and of the chemical reactivity at the interface.
The theoretical work will be done on oxide/liquid water interfaces and SAMs/liquid water interfaces (SAM: Self Assembled Monolayer), their hydrophilic/hydrophobic behaviors at the molecular level will be revealed as a function of pH, electrolytes concentration, and surface morphology. The DFT-MD and CIMD simulations will systematically be coupled with SFG spectroscopy (experiments by our collaborators & simulations by us). Part of this PhD work will be devoted to the development of a new scheme that will allow CIMD-SFG spectra to be of DFT-MD-SFG accuracy. SFG experimental spectra 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, GROMACS or LAMMPS for CIMD, 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. Developments in synergy with Graph Theory and AI are part of this project for those interested.

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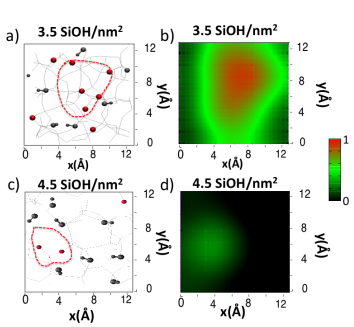

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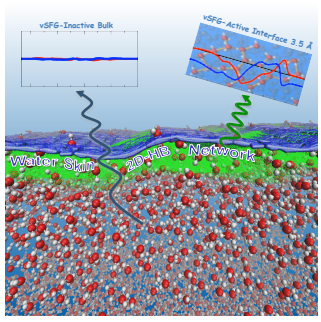

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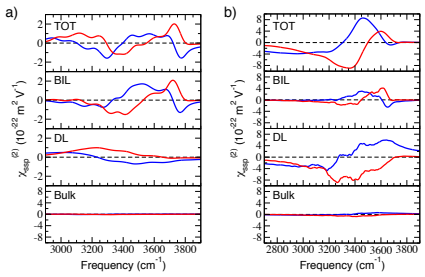

Deconvolution of SFG signals at solid or air/water interfaces in terms of BIL (Binding Interfacial Layer), DL (Diffuse Layer) & Bulk contributions. **PCCP 20:5190(2018)**

PhD: High-throughput theoretical decoding of vibrational spectra of gas phase glycans: coupling AIMD/DFT-MD simulations with Graph Theory and AI

Keywords : theoretical and computational chemistry, ab initio molecular dynamics, DFT-MD, AIMD, Graph Theory, Artificial Intelligence AI, machine learning, deep learning, spectroscopy, IR, Raman, mass spectrometry, materials

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The goal of modern analytical (bio)-chemistry is to sequence (bio-)molecules and reveal their 3D microscopic structures. Mass Spectrometry (MS), one of the essential analytical method in the gas phase domain, is commonly (orthogonally) coupled to techniques such as e.g. chromatography and electrophoretic separation, Ion Mobility Spectrometry (IMS), activation methods, and spectroscopies. Among these orthogonal couplings to MS, infrared (IR) action spectroscopy is one of the very few techniques providing direct information into atomistic 3D-structures, with the paradigm of 1-to-1 spectral fingerprints←→3D-structure relationships. The past 5 years have seen the MS-IR gas phase community embark on the challenge of high-throughput analytical chemistry of bio-oligomers of relevance in biological processes. Decoding IR signatures (i.e. band-positions, intensities and shapes) into a microscopic 3D-structure is however non-trivial and cannot be accomplished from experiments alone for complex flexible (bio)-molecules. Theoretical chemistry calculations have to be employed which is the expertise of the Gaigeot group by employing AIMD/DFT-MD simulations for the calculation and assignment of anharmonic vibrational spectra, for very diverse molecular systems, i.e. gas phase molecules and clusters, liquids, solid/liquid interfaces.
This PhD project focuses on the characterization of the 3D-structures of (very) large glycan oligomers (of high interest in biological recognition in general, an expertise in our lab) in order to make the systematic 1-to-1 assignments between structures and IR signatures. To that end, AIMD/DFT-MD simulations will be applied, together with the application of our developed methods for extracting and decoding the IR vibrational modes.
To be high throughput and applied routinely, i.e. applied in a few minutes only, the decoding of the IR signatures into a 3D-structure has however to be done in a completely different theoretical way. We are currently developing innovative theoretical methods based on graph theory, machine/deep learning, AI, from computer science, in our bi-disciplinary team made with the group of Prof Dominique Barth at the University Paris-Saclay (expert in graph theory & AI techniques). In the present PhD, the first applications of these new methods to the IR-decoding will be done. If interested, the PhD student will directly participate to the on-going developments of the graph theory and machine-/deep- learning methods.

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Our group is internationally recognized in AIMD/DFT-MD simulations of gas phase molecules, liquids and solid/liquid interfaces. The following references illustrate some of our recent original works and innovative developments for gas phase IR spectroscopy and Graph Theory methods [Chem. Rev. 120: 3233-3260 (2020), Faraday Discussions 217:67 (2019), J. Chem. Phys. 149:184102-15 (2018), J. Chem. Theory. Comput.,13:3802 (2017), Phys. Chem. Chem. Phys., 19:13778 (2017)].

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