Oxide/water interfaces characterized by Ab Initio Molecular Dynamics simulations: making the link between structure and SFG spectroscopy, and beyond

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In this presentation, we review our recent theoretical works on the characterization of oxide/liquid water interfaces, i.e. silica/water (crystalline and amorphous), alumina/water and cobalt/water interfaces, making a systematic link between structure and non-linear SFG (Sum Frequency Generation) experiments. Non-linear SFG spectroscopy in the 3000-4000 cm^{-1} spectral domain is indeed one method of choice to probe complex inhomogeneous solid/liquid and liquid/air interfaces. Although an extremely powerful technique, the detailed interpretation of the experimental signatures requires associated calculations. This is the challenge we have been tackling over the past 5 years, applying ab initio DFT-based molecular dynamics simulations (AIMD/DFT-MD).

We will illustrate here some of our recent works showing how to separate vibrational signatures arising from the different layers of water at the interface, how to provide a direct interpretation of the H-Bond networks at play at interfaces (water-water vs solid-water networks), including our recent works unraveling 2-dimensional interfacial water networks, how to define the only three spatial regions of interest at any charged interfaces (BIL, Binding Interfacial Layer; DL, Diffuse Layer; Bulk) and how to universally interpret/assign SFG vibrational bands from these three regions, including χ^2 and χ^3 contributions into the theoretical signals. We will hence unravel trends in going from hydrophobic to hydrophilic surfaces at the interface with liquid water, providing a clear and unambiguous knowledge of the oxide/water interfacial structures. Furthermore, our very recent works on reactive cobalt oxide/water interfaces of interest for electrocatalysis and water splitting will be presented, showing how the above structural and spectroscopic analyses can be used in electrochemical conditions.

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Some recent references:

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