Water at silica/ and alumina/liquid water interfaces investigated by DFT-MD simulations: structure & SFG vibrational signatures revealed

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Probing the microscopic structural organization of water at silica/ & alumina/liquid water interfaces, including electrolytes, is achieved here by first principles DFT-based molecular dynamics simulations (DFT-MD). SFG (Sum Frequency Generation) vibrational spectroscopy is one of the very few tools used to probe such buried interfaces, and such signals are extracted from our simulations and analyzed in details.

We show how water is organized in layers at these oxide interfaces, with certain specific orientations and H-bond networks that depend on the chemical nature of the surface oxides, we show where the electrolytes are located and how their location can be influenced by local and non local interactions. We demonstrate how these structural organisations give rise to specific SFG signals. We are not only providing a 1-to-1 comparison with experimental SFG data, of amazing quality/accuracy, but we are furthermore able to clearly understand/assign vibrational bands in terms of the underlying intertwined structural properties of the oxide surface, of the water layers & their properties, of the electrolytes and especially how these electrolytes form double layers.

Our work demonstrates how the structural arrangements of liquid water and electrolytes at silica/ & alumina/water interfaces modulate the final spectroscopic signatures, providing guidelines and marker-bands to be used by experimentalists.