DFT simulations of nanoconfined water between electrified gold surfaces using Non-Equillibrium Green's Functions

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Albeit water is the most common and best studied solvent, understanding its structure and properties at the surface of materials is still an open problem. Besides, important modifications of its structure and dynamics occur when the surfaces are electrified (as in electrochemical environments), and when the confinement space is nanometric. Density Functional Theory (DFT) simulations can deal with these issues, although imposing the external voltage in the simulation has proven difficult. Non-Equillibrium Greens functions (NEGF) techniques [1,2] as implemented in the SIESTA DFT package [3,4] are used here to address this problem, allowing first-principles molecular dynamics simulations of nanoconfined water in the presence of a finite voltage between the two confining surfaces. We will present proof of concept calculations of water between gold electrodes, showing the potential of our approach. Efforts for the massive parallelization of the simulations in large HPC infrastructure will be discussed, as well as the scaling of the CPU time with the system size. We also show how to increase the size of systems (in terms of number of atoms) and the simulation time which can be addressed by these simulations by using a quantum mechanics/molecular mechanics (QM/MM) approach coupled to the NEGF method, which will be the topic of the next oral presentation of this session, by Dr. Ernane Freitas.

References

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[4] García et al, J. Chem. Phys. 152, 204108 (2020).

Figures

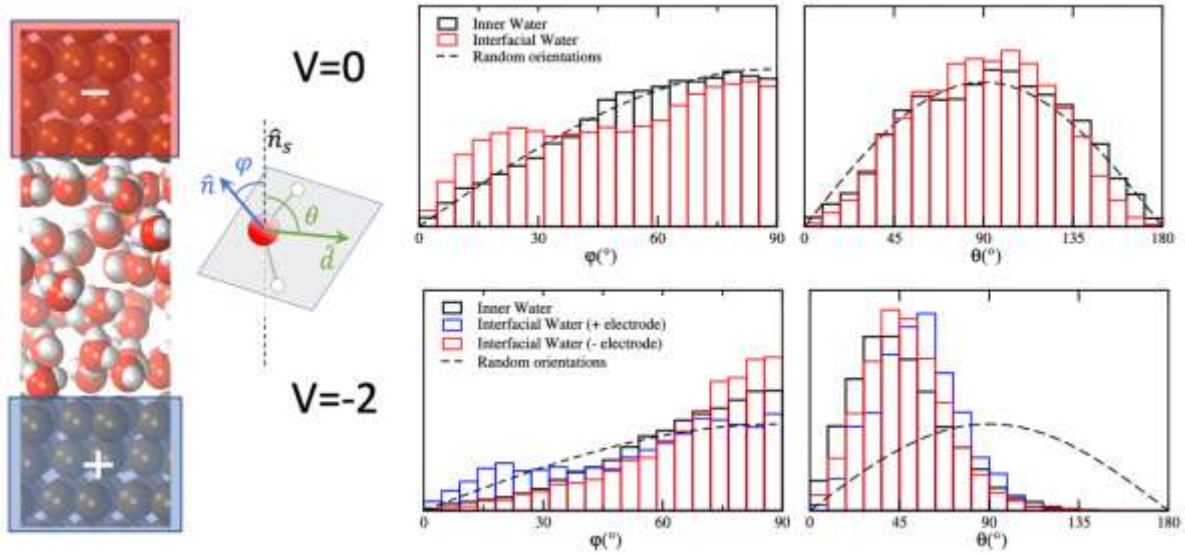

Figure 1: Distribution of angles for the H_2O molecules (both interfacial and inner) for water nanoconfined between Au electrodes, as shown on the representation on the left. The top graphs show the results when there is no voltage imposed between the electrodes, while ones in the bottom are for a voltage of V=2 Volt. We use two angles to characterize the molecular orientations, as shown in the second scheme on the left.