Water at electrified graphene interfaces: structure, dynamics, vibrational SFG spectroscopy and consequences for electron transfer reactions

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Water's properties at electrified interfaces are central in e.g. electrochemistry. We will present the main results of our theoretical study of liquid water/electrified semi-metallic graphene sheet interfaces. We find that – surprisingly – the interfacial water molecules' spatial structural and dynamical properties vary non-monotonically with the applied potential. We show that this behavior can be directly understood within the extended jump-model of water hydrogen-bond exchanges. We also indicate how simulated vibrational sum-frequency generation spectra of the water OH stretch depend on the contributions from the interfacial and the subsequent water layers. Finally, several key consequences for interfacial electron transfer reactions are discussed.