Machine learning-enabled ab initio studies of water solutions in bulk and at interfaces

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Machine learning methods have significantly expanded the accessible length and time scales of ab initio molecular dynamics (AIMD), enabling the exploration of previously inaccessible properties. In this talk, I will present our recent works using machine learning-enabled AIMD to investigate water solutions in bulk and at interfaces, which are critical for understanding processes fundamental to life and various energy applications.

Part 1: Dielectric permittivity of salt water [1]
The dielectric permittivity of salt water decreases with increased salt concentration, a phenomenon referred to as dielectric decrement. Our findings suggest that this decrement is due to the intrusion of ionic hydration shells into the hydrogen-bond network of the solvent water. This intrusion disrupts the hydrogen-bond network, suppressing the collective response of water molecules and consequently reducing the dielectric permittivity.

Part 2: Electrical double layer (EDL) at TiO_2-electrolyte interfaces [2]
We provide a molecular-scale picture of the EDL at TiO_2-electrolyte interfaces that demonstrates the limitations of standard mean-field models. We further develop a method to accurately calculate the electrostatic potential drop at the interface. The computed capacitance originating from the adsorbed charges and the potential drop agrees with experiments, supporting the reliability of our description of the EDL. The larger interfacial capacitance of basic relative to acidic solutions originates from the higher affinity of the cations for the oxide surface and gives rise to distinct charging mechanisms on negative and positive surfaces.

References
[1] Chunyi Zhang, Shuwen Yue, Athanassios Z. Panagiotopoulos, Michael L. Klein*, and Xifan Wu*. "Dissolving salt is not equivalent to applying a pressure on water." Nature communications 13, no. 1 (2022): 822.
[2] Chunyi Zhang, Marcos Calegari Andrade, Zachary K. Goldsmith, Abhinav S. Raman, Yifan Li, Pablo Piaggi, Xifan Wu, Roberto Car*, and Annabella Selloni*. "Electrical double layer and capacitance of TiO2 electrolyte interfaces from first principles simulations." arXiv preprint arXiv:2404.00167 (2024).