Enhancing electrochemical solvation with molecular dynamics and machine learning

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Efficient and accurate description of electrolytes is a central challenge in application of first-principles methods to electrochemical systems for energy storage and conversion. Implicit solvation models are quick, but can be rather inaccurate for detailed electrochemical modeling, while explicit solvation based on ab initio molecular dynamics simulations faces time and length scale issues, especially when including electrolyte ions explicitly. I will discuss the evolution of generations of implicit solvation models, particularly focusing on their accuracy for predicting the capacitance of electrochemical interfaces, which is important directly for electrochemical energy storage applications, and indirectly in electrocatalysis by connecting the electrode potential to the local electric field. Using molecular dynamics simulations of electrochemical and pure-solvent interfaces as a guide, I will outline the ingredients necessary for future implicit approaches that can capture all key features of experimental electrochemical capacitance profiles. Finally, I will also briefly introduce progress towards universal classical DFT solvation to bridge the gap between implicit and explicit solvation, leveraging machine learning both to map the response of arbitrary solvents/electrolytes in molecular dynamics and to capture that response within free energy functionals.