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First-principles simulations of charge storage at electrochemical interfaces in supercapacitors

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Supercapacitors store charge via polarization at the electrode-electrolyte interface. Many models of interfacial charge storage focus on the formation of the electric double layer (EDL) in the electrolyte, but it is often assumed that in the electrode, a shift in the Fermi level is the only notable response to interface polarization. In reality, the presence of the interface impacts the fundamental properties of both the electrode and the electrolyte, often in complex and nontrivial ways that are difficult to capture using simple models. I will discuss how including an applied bias potential in first-principles simulations allows one to directly simulate the process of charge storage at the electrode-electrolyte interface, and thereby to unravel the interplay between the electrode and the electrolyte. I will show how these more complex treatments lead to improved descriptions of intrinsic quantum and EDL capacitance contributions in graphene-based supercapacitors, which can be used to suggest engineering strategies for improved electrode materials. I will also discuss how combining theory with in operando X-ray spectroscopy can give insights into nanoscale chemical changes and mesoscale morphological changes in electrodes during charging. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under contract DE-AC52-07NA27344.