Machine learning of electronic charge densities in the simulation of electrochemical interfaces.

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Machine-learning methods are playing an increasingly prominent role in the atomistic simulation of materials. By inferring the relevant electronic properties from the sole knowledge of the spatial atomic coordinates, these data-driven techniques make it possible to attain the accuracy of quantum mechanics while sidestepping the computational burden of electronic-structure methods. When it comes to electrochemical systems, however, the effective implementation of these approaches presents unique challenges. Notably, the presence of a heterogeneous interface between an electrode material and an electrolyte solution under an applied electric bias requires predicting complex long-range interactions that drive the dynamics of the system at the nanoscale. In this context, I will introduce a machine-learning method suitable to treat the three-dimensional electron density of a system [1] and show how it can be used to predict the nonlocal electronic polarization of metallic electrodes that are embedded in concentrated ionic solutions [2]. I will then continue discussing the integration of this method within the MetalWalls molecular dynamics program in order to accurately simulate the electrostatic properties of model ionic capacitors [3]. By taking a gold/electrolyte interface as an example, I will show how these advancements allow us to overcome the shortcomings of classical approaches in the description of the electrical double layer, by matching the accuracy of density-functional QM/MM methods while reaching time scales of the order of nanoseconds.

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3. Andrea Grisafi and Mathieu Salanne, *Accelerating QM/MM simulations of electrochemical interfaces through machine learning of electronic charge densities*, The Journal of Chemical Physics 161, 024109 (2024) [https://doi.org/10.1063/5.0218379]