

Abstract Submitted
for the MAR15 Meeting of
The American Physical Society

Joint Density-Functional Theory for Atomically Detailed Structure of the Electrode/Electrolyte Interface KENDRA LETCHWORTH-WEAVER, CHRISTINE UMBRIGHT, RAVISHANKAR SUNDARARAMAN, T.A. ARIAS, Department of Physics, Cornell University, Ithaca, NY 14853 — Understanding the complex and inherently multi-scale interface between a charged electrode surface and a fluid electrolyte would inform design of more efficient and less costly electrochemical energy storage and conversion devices. Joint density-functional theory (JDFT) bridges the relevant length-scales by joining a fully *ab initio* description of the electrode with a highly efficient, yet atomically detailed classical DFT description of the liquid electrolyte structure. First, we introduce a universal functional to couple any quantum-mechanical solute system with a classical DFT for any liquid. This universal coupling functional captures aqueous and non-aqueous solvation free energies of small molecules with a mean absolute error of only 1-2 kcal/mol. We also present classical density-functionals for ionic species which reproduce the key features of ion-water correlation functions when combined in a mixture with existing functionals for water. Leveraging the above theoretical innovations and our framework to treat charged systems in periodic boundary conditions, we then predict the voltage-dependent structure and energetics of solvated ions at the interface between a graphene electrode and an aqueous electrolyte.

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Date submitted: 14 Nov 2014

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