Predicting the electronic properties of aqueous solutions from first-principles[1] ERIC SCHWEGLER, TUAN ANH PHAM, Lawrence Livermore National Laboratory, MARCO GOVONI, University of Chicago and Argonne National Laboratory, ROBERT SEIDEL, STEPHEN BRADFORTH, University of Southern California, GIULIA GALLI, University of Chicago and Argonne National Laboratory — Predicting the electronic properties of aqueous liquids has been a long-standing challenge for quantum-mechanical methods. Yet it is a crucial step in understanding and predicting the key role played by aqueous solutions and electrolytes in a wide variety of emerging energy and environmental technologies, including battery and photoelectrochemical cell design. Here we propose an efficient and accurate approach to predict the electronic properties of aqueous solutions, based on the combination of first-principles methods and experimental validation using state-of-the-art spectroscopic measurements. We present results for the photoelectron spectra of a broad range of solvated ions, showing that first-principles molecular dynamics simulations and electronic structure calculations using dielectric hybrid functionals provide a quantitative description of their electronic properties, including excitation energies, of the solvent and solutes. The proposed computational framework is general and applicable to other liquids, thereby offering great promise in understanding and engineering solutions and liquid electrolytes for a variety of important energy technologies.

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