Abstract Submitted
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Solvated ions as defects in liquid water: A first-principles perspective[1] ERIC SCHWEGLER, TUAN ANH PHAM, Lawrence Livermore Natl Lab, MARCO GOVONI, GIULIA GALLI, Institute for Molecular Engineering-The University of Chicago, and Argonne National Laboratory — Understanding the electronic properties of solvated ions is crucial in order to control and engineer aqueous electrolytes for a wide variety of emerging energy and environmental technologies, including photocatalytic water splitting. In this talk, we present a strategy to evaluate electronic energy levels of simple solvated ions in aqueous solutions, using a combination of first-principles molecular dynamics simulations and many-body perturbation theory within the GW approximation. We considered CO_3^{2-}, HCO_3^-, NO_3^-, NO_2^- ions and we show that by analogy to defects in semiconductors, these solvated ions may be classified as deep or shallow defects in liquid water. In particular CO_3^{2-} and NO_2^- ions behave as shallow defects, while HCO_3^- and NO_3^- as deep ones. We also show that the inclusion of many-body corrections constitutes significant improvement over conventional density functional theory calculations, yielding satisfactory agreement with photoemission experiments.

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