Abstract Submitted
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A novel theoretical probe of the $SrTiO_3$ surface under water-splitting conditions[1] KENDRA LETCHWORTH-WEAVER, DENIZ GUNCELER, TOMÁS ARIAS, Dept. of Physics, Cornell University, MANUEL PLAZA, XIN HUANG, JOEL BROCK, School of Applied and Engineering Physics, Cornell University, JOAQUIN RODRIGUEZ-LÓPEZ, HECTOR ABRUÑA, Dept. of Chemistry and Chemical Biology, Cornell University — Understanding the reaction mechanisms required to generate hydrogen fuel by photoelectrolysis of water is essential to energy conversion research. These reaction pathways are strongly influenced by the geometry and electronic structure of the electrode surface under water-splitting conditions. Electrochemical microscopy has demonstrated that biasing a $SrTiO_3$ (001) surface can lead to an increase in water-splitting activity. *In operando* X-ray reflectivity measurements at the Cornell High Energy Synchrotron Source (CHESS) correlate this increase in activity to a significant reorganization in the surface structure but are unable to determine the exact nature of this change. Joint Density-Functional Theory (JDFT), a rigorous yet computationally efficient alternative to molecular dynamics, provides a quantum-mechanical description of an electrode surface in contact with an aqueous environment, and a microscopically detailed description of the interfacial liquid structure. Our JDFT calculations determine the structure of the activated $SrTiO_3$ surface and explore why it is correlated with higher activity for water splitting. With no empirical parameters whatsoever, we predict the X-ray crystal truncation rods for $SrTiO_3$, finding excellent agreement with experiment.

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