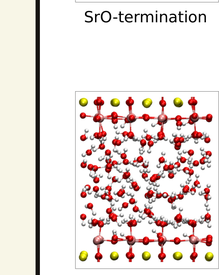
$SrTiO_3/H_2O$ interface from first-principles molecular dynamics and experiment



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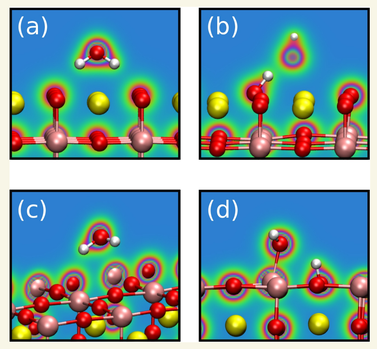
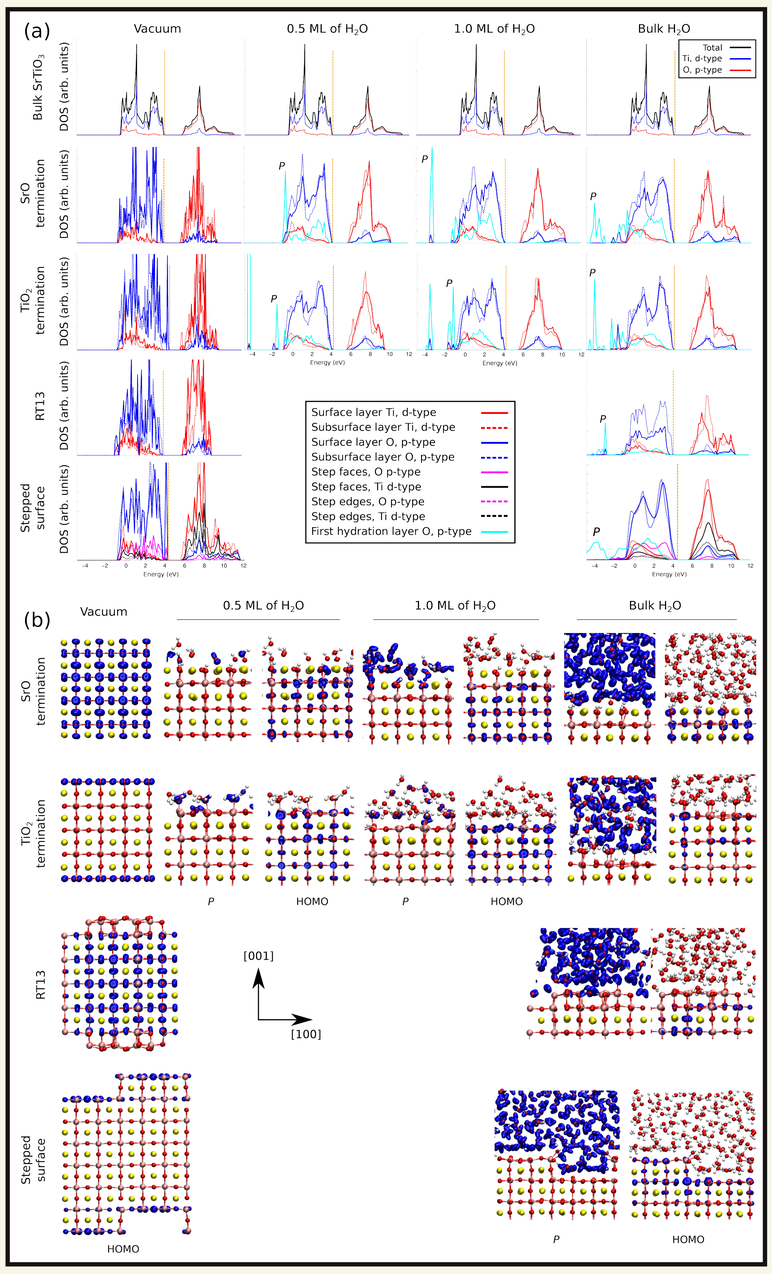
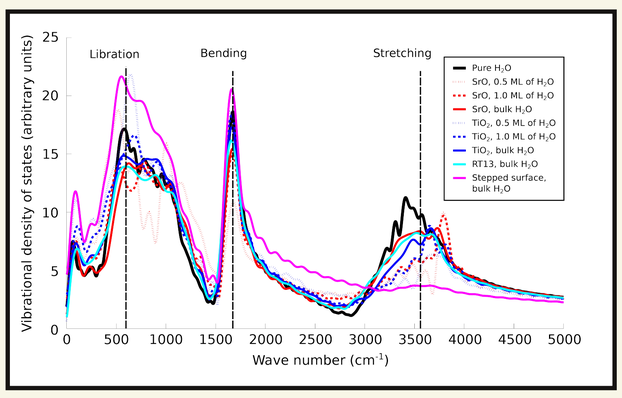
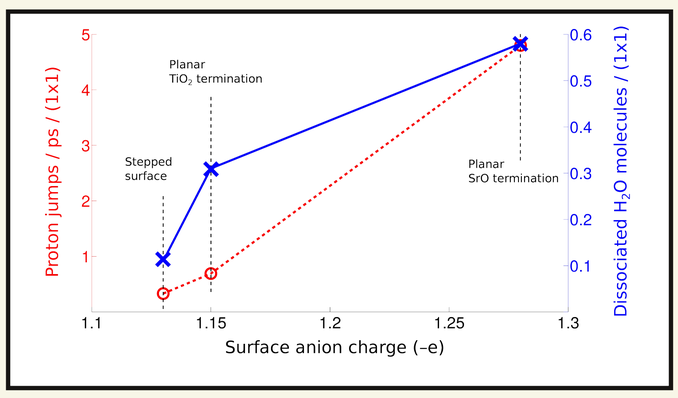
Strontium titanate ($SrTiO_3$) is a promising photocatalyst for hydrogen production through water-splitting. In order to realize this technological potential, a robust and detailed understanding of the $SrTiO_3/H_2O$ solid-liquid interface is necessary. We use density functional theory molecular dynamics simulations (DFT MD), classical MD, frequency-modulated atomic-force microscopy (FM-AFM), and solid-liquid contact angle measurements to understand the structural, electronic, and dynamical properties of this important metal-oxide/water interface.



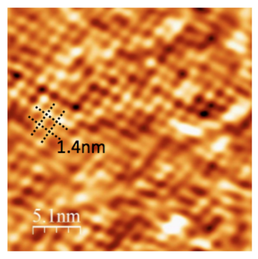
Left: Hydration structure of various $SrTiO_3$ surfaces at bulk H_2O coverage. **Right:** Electronic density of states (a) and orbital isosurfaces (b) for the same. **Top:** Most favorable molecular and dissociative adsorption geometries for H_2O on SrO-terminated (a,b) and TiO_2-terminated (c,d) $SrTiO_3$ (001).



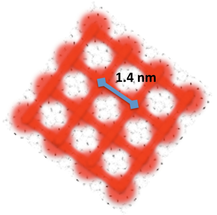


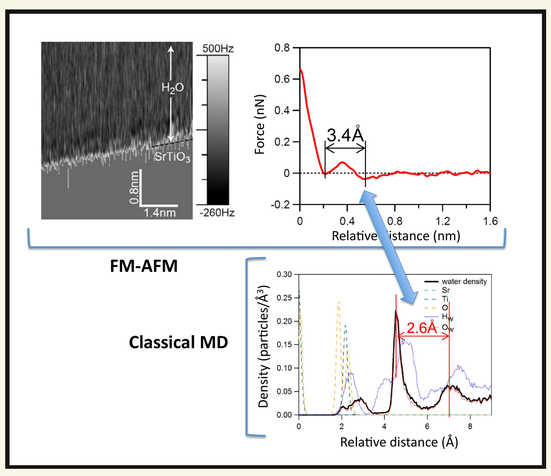


Processed FM-AFM topographic image of RT13 $SrTiO_3$ under 50 mM KCl aq.

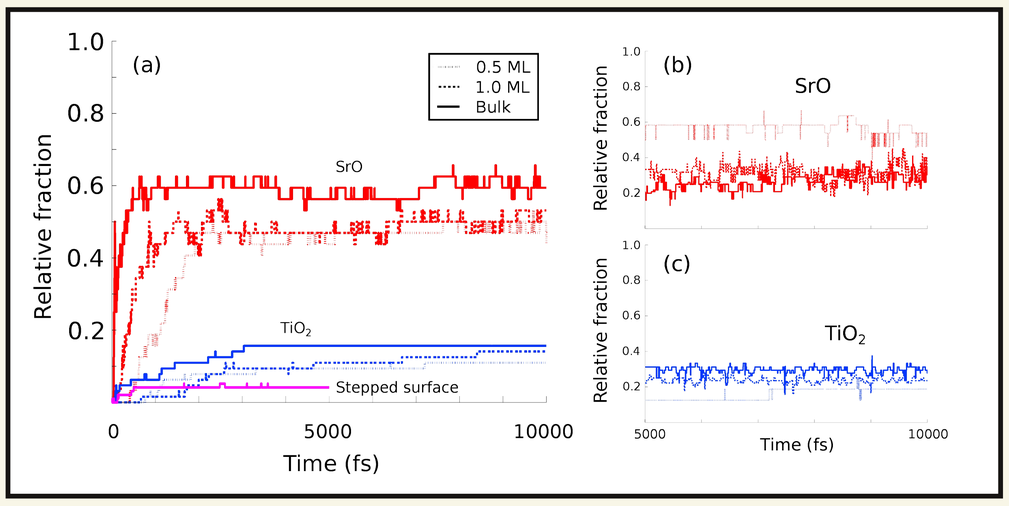


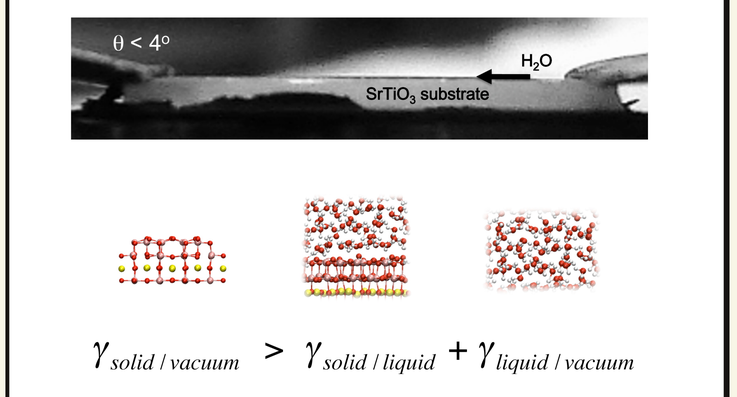
Hydration structure from classical MD





Above left: Vibrational density of states for water on $SrTiO_3$ from from DFT MD. **Above center:** Correlation between mean surface oxygen charge and degree of hydroxylation and mean proton transfer rate. **Above right:** Topographic FM-AFM image compared to classical MD of water on the RT13 surface.





Above: Force curve from FM-AFM compared to classical MD vertical density profile on RT13. **Far left:** Fraction of hydroxylated surface oxygens (a), and the fraction of dissociated water in the first hydration layer of (b) SrO- and (c) TiO_2-terminated $SrTiO_3$. **Left:** A droplet of H_2O spreads thin onto the RT13 surface in experiment, in agreement with DFT MD.

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