First Principles Structure Determination in Complex Environments: Leveraging Experimental Characterization and Machine Learning

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The interface between a solid surface and a liquid is relevant in applications such as water purification, corrosion, catalysis, and energy storage. The exact structure and composition of the solid surface crucially impacts ion adsorption, dissolution and intercalation and electron energy alignment between surface and reacting molecules. However, the structure of these surfaces is often challenging to probe experimentally under operating or growth conditions and the relevant reaction mechanisms are often unknown. Furthermore, even control of the termination of the bare surface via epitaxial growth is not always straightforward due to processes such as dynamic layer rearrangement. X-ray reflectivity (XRR) determines the electron density of an interface with high resolution but typically relies on model-dependent fitting to invert the data and obtain the corresponding atomic structure. First principles theory calculations of surface structure rely upon approximations which can be inaccurate for surfaces out of equilibrium and with defects. We will present software developments which integrate first principles theory of surfaces with experimental XRR measurements using global optimization algorithms, allowing us to more accurately determine interfacial structures than is possible through experiment or theory alone. Additionally, we will explore preliminary results utilizing a combination of first principles theory and machine learning to characterize local structure in high entropy oxides, which show promise as catalyst and battery materials.