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Nonlinear X-ray Spectroscopy: a novel probe for interfacial dynamics

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Chemistry at interfaces is central to applications spanning energy science, catalysis, energy storage, the water-energy nexus, and solar fuel generation. Understanding this chemistry at the molecular scale remains a grand challenge, due to the complex nature of buried interfaces. This is largely due to the lack of reliable in situ characterization tools with the chemical and interfacial sensitivity needed to track chemical transformations and transport at these interfaces. Recently, soft X-ray Second Harmonic Generation spectroscopy (SXR-SHG) was demonstrated for the first time on carbon films [1]. Aside of surface-specific X-ray spectroscopy, SXR-SHG is highly promising for studying buried interfaces due to the high penetration depth, elemental selectivity, and extremely high interfacial sensitivity (1-3 atomic layers) while providing electronic structure information that is sensitive to chemical bonding, symmetry, and weaker interactions such as hydrogen bonding. In recent experiments we were able to quantify the interfacial bond geometry of an organic-inorganic interface with a precision of 0.1Å [2]. In addition, we studied the second-order phase transition in a polar metal and monitored the displacement of the alkali metal ions within the unit cell of the atomic lattice using SXR-SHG [3] as well as performing SXR-SHG polarimetry for anisotropy analysis [4]. The inherent femtosecond nature of the required highly intense X-ray pulses provides excellent prospects for time-resolved interfacial studies and our most-recent findings show that SXR-SHG can be expanded to wide availability using laboratory sources [5].

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