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NANOSCIENCE COLLOQUIUM

Resolving chemical bond dynamics at an electrode surface

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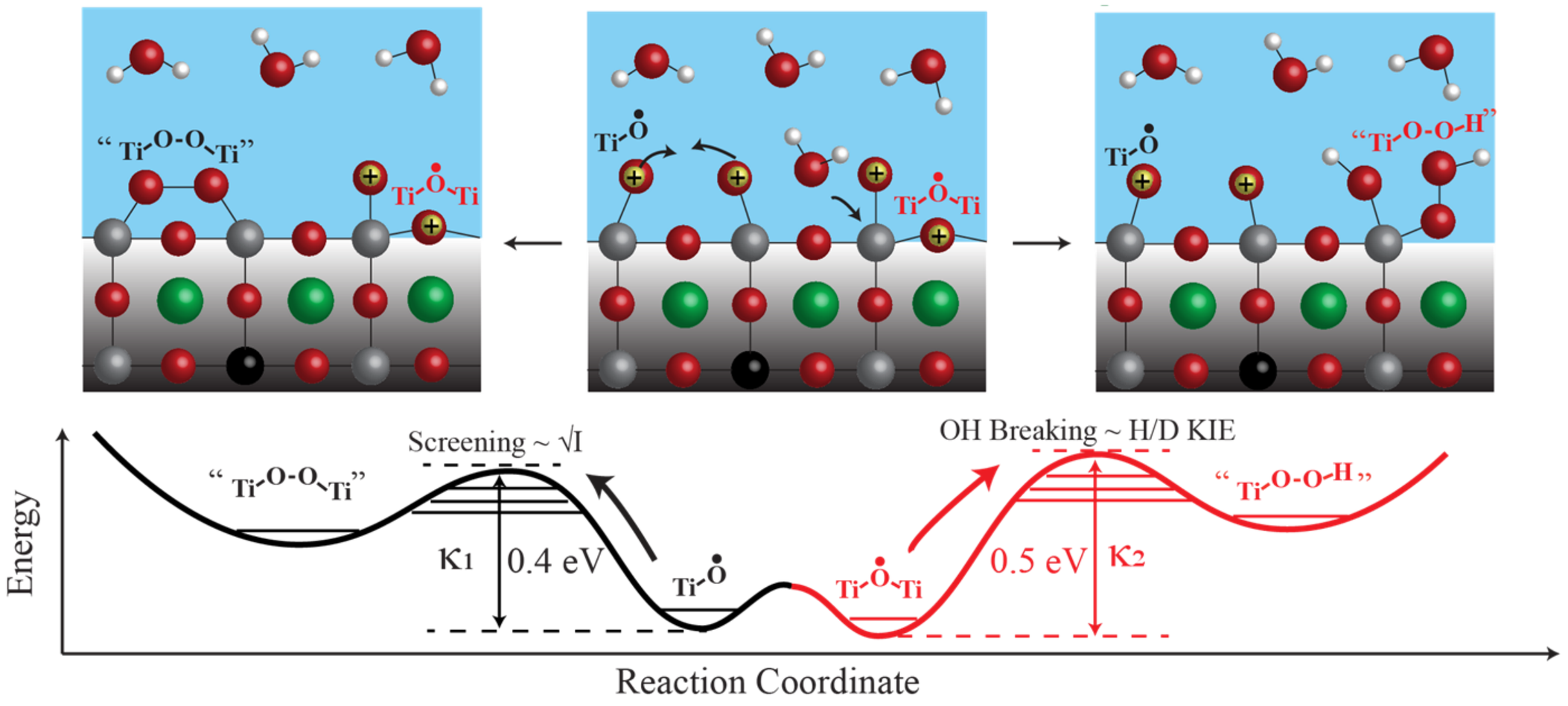
Abstract: Catalytic mechanisms at electrode surfaces guide the development of electrochemically-controlled energy storing reactions and chemical synthesis. Yet, only recently have the intermediate steps and how they lead to the chemical bonds of the product been directly observed. We employ time-resolved vibrational and electronic spectroscopy to interrogate the molecular and dynamic properties of the meta-stable intermediates and how they reach critical transition states. In order to do so, the highly selective water oxidation reaction is triggered at the semiconductor (SrTiO3)-aqueous interface by an ultrafast light pulse in an electrochemical cell. We capture the birth of charge-trapping intermediates (e.g. Ti-O• and Ti-O•-Ti) and their evolution towards the first chemical bond formation event, O-O. The research reveals ways by which the surface creates new paradigms of reactivity for heterogeneous catalysis at the electrode surface. The paradigms promise to explain and exploit the distinctive properties of heterogeneous catalysis in this environment, including its robustness to charge degradation and the existence of multiple and tunable reaction pathways.



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