Taming the complexity of solid catalysts with the modern modeling toolbox

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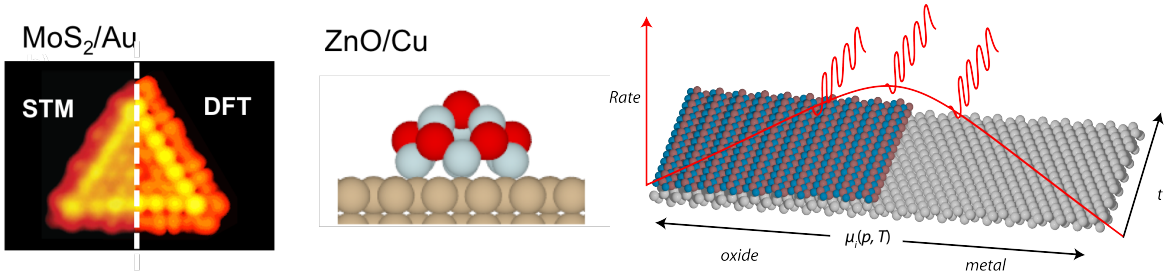
During the last decades, the improvement of solid catalysts has increasingly relied on a rational, knowledge-based approach. Computational modeling techniques based on quantum mechanics have largely contributed to this endeavor, allowing rationalizing, and even predicting, the catalytic properties of materials based on the electronic structure of their active sites.

Despite the success of studies using density functional theory (DFT) calculations, the models used in theoretical catalysis (and in many surface science studies) always represent simplified versions of the materials and conditions used in applied catalysis. Extended surfaces with regular structures are often studied instead of the nanoparticulate structures more frequently found in technical catalysts, and the effect of the typically harsh operating conditions on the structure and oxidation state of catalytic substrates is often ignored. These simplifications (referred to as materials and pressure gaps) limit the descriptive and predictive capacity of model catalysts and theoretical models, which often disregard effects from undercoordinated sites of nanoparticles, catalyst-support interactions, or operation under high pressures.

In my talk, I will present the results of three case studies where the complexity of different catalytic substrates is addressed by connecting accurately calculated atomistic observables from DFT calculations with macroscopic properties. These studies combine global optimization methods, simulated spectroscopy, (constrained) *ab initio* thermodynamics analysis, and first-principles microkinetic models.



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