

# **Modeling Single-Atom Catalysis**

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### Significance and Relevance

We present a computational approach to model single-atom catalysts. The approach is applied to a recently synthesized 2D material; "goldene" an Au(111) monolayer. We propose the system as a suitable support for SACs. Results indicate that SACs are stable in reaction conditions and may be active for hydrogen and oxygen evolution. Simple descriptors allow to rationalize and, in principle, to optimize the catalytic activity.

*Preferred and 2<sup>nd</sup> choice for the topic:* Multiscale modeling and advanced simulation aspects *Preferred presentation*: Oral only.

# **Introduction and Motivations**

Single Atom Catalysts (SACs) are emerging as new frontier in the field.<sup>1</sup> Computational chemistry offers a valid framework to access the atomistic details of catalytic processes and to rationalize of even predict novel systems.

In this work, the key ingredients to model SACs will be discussed. SACs differ substantially from metal surfaces and can be considered analogues of coordination compounds. In coordination chemistry, at variance with metal surfaces, stable complexes can form.<sup>2</sup> We show that the same can occur on SACs and their formation may change the kinetics of the process.<sup>3,4</sup> We propose an approach to predict the stability of SACs under working conditions of pH and applied voltage.<sup>5</sup> Needless to say, the adopted DFT functional affects the accuracy of the predictions, and we show evidence suggesting that self-interaction corrected schemes should be adopted.<sup>6</sup>

# **Results and Discussion**

DFT numerical simulations demonstrate that these ingredients may lead to different conclusions about the activity of SACs.<sup>7</sup> We applied the approach to a novel candidate of supporting matrix for SACs; *"goldene"* an Au(111) monolayer (1L), recently synthesized.<sup>8</sup> Our results indicate that goldene-based SACs are often stable object in reaction conditions, and they may be active for relevant reactions such as hydrogen and oxygen evolution. Importantly, the activity can be rationalized and optimized by introducing simple descriptors.

This work provides an example of the important analogies between the chemistry of SACs and that of coordination compounds, and underlines the key ingredients to be accounted when attempting to provide predictions with computational frameworks. Results also point as goldene-based SACs as promising systems deserving further attention from experimental and theory communities.





**Figure 1** Key ingredients to model a SAC (panel a), reaction energy profile of HER (panel b) and OER (panel c) on Au(111)-1L based SAC.

### References

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