Large-scale simulations of catalytic surface dynamics with first principles machine learning

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Abstract

Understanding atomic-level processes in surface science and heterogeneous catalysis is complicated by the wide range of time scales and length scales needed for simulations. To accelerate molecular dynamics calculations, we rely on machine learning methods to capture interatomic interactions with quantum accuracy. We then implement and deploy these models on parallel GPUs to reach billions of atoms in size or microseconds in time, and demonstrate this scale for a Pt hydrogen exchange catalyst at the micron scale. One method is a family of equivariant interatomic potential models (NequIP [1] and Allegro [2]) based on symmetry-preserving layer architectures that we use to achieve state-of-the-art accuracy and training efficiency for simulating atomistic. Another method (FLARE [3, 4]) enables autonomous selection of training sets for reactive systems, based on adaptive closed-loop algorithm that constructs accurate and uncertainty-aware Bayesian force fields on-the-fly from a molecular dynamics simulation. We examine the current limitations of machine learning models across relevant metal systems [5] and highlight the usefulness of ML-accelerated MD simulations to study surface reconstruction [6], dislocation dynamics [7], direct heterogeneous reactions [4, 8], nanoparticle shape changes [9], and full-scale catalyst modeling. These results demonstrate the power of first-principles based machine-learned force fields for the dynamic study of a variety of material classes, yielding previously inaccessible mechanistic and kinetic insights at relevant scale for comparison to experimental characterization techniques.

References

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