

Nearly Exact Molecular Dynamics with Quantum Chemistry and Machine Learning

Alexandre Tkatchenko

Physics and Materials Science Research Unit

University of Luxembourg

Email: alexandre.tkatchenko@uni.lu

Abstract:

Molecular dynamics (MD) simulations employing classical force fields constitute the cornerstone of contemporary atomistic modeling in chemistry, biology, and materials science. However, the predictive power of these simulations is only as good as the underlying interatomic potential. Classical potentials often fail to faithfully capture key quantum effects in molecules and materials. Here we enable the direct construction of flexible molecular force fields from high-level ab initio calculations by incorporating spatial and temporal physical symmetries into a gradient-domain machine learning (sGDML) model in an automatic data-driven way. The developed sGDML approach faithfully reproduces global force fields at quantum-chemical CCSD (T) level of accuracy and allows converged molecular dynamics simulations with fully quantized electrons and nuclei. We present MD simulations, for flexible molecules with up to a few dozen atoms and provide insights into the dynamical behavior of these molecules. Our approach provides the key missing ingredient for achieving spectroscopic accuracy in molecular simulations.