Machine Learning Surrogate Models of Electronic Structure

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Atomistic simulation based on quantum mechanics (QM) is currently being revolutionized by machine-learning (ML) methods. Many existing approaches use ML to predict molecular properties based on quantum chemical reference data. This has enabled molecular property prediction within vast chemical compound spaces and high-dimensional parametrization of energy landscapes for the efficient simulation of measurable observables. However, as all properties derive from the QM wave function, an ML model that can predict the wave function or the electronic Hamiltonian also has the potential to predict other properties. In this talk, I will explore ML approaches that deliver surrogate models of the electronic structure based on electronic structure Hamiltonians [1,2] with the aim to develop methods that use ML and QM in synergy. Using example systems from heterogeneous catalysis and organic electronics, I will discuss the challenges associated with encoding physical symmetries and invariance properties into linear and deep learning mappings of atomic configuration and composition onto electronic structure. Upon overcoming these challenges, integrated ML-QM methods within modern, modular software frameworks offer the combined benefits of data-driven parametrization and first-principles-based methods [3]. I will discuss several opportunities associated with building ML-augmented quantum chemical methods, including Inverse Chemical Design based on ML-predicted electronic structure and the development of efficient and accurate surrogate models to study materials chemistry [4,5].

References

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