Atomistic simulation based on quantum mechanics (QM) is currently being revolutionized by machine-learning (ML) methods. ML methods enable molecular property prediction within vast chemical compound spaces and the high-dimensional parametrization of energy landscapes for the efficient simulation of molecular dynamics. However, as all properties derived from the QM wave function, an ML model that is able to predict the wave function also has the potential to predict all other molecular properties.

In this talk, I will explore ML approaches that directly represent QM properties beyond energies such as wave functions, QM Hamiltonians, and their derivatives for the development of methods that can use ML and QM in synergy. Using examples from molecular dynamics, and heterogeneous catalysis, I will discuss the challenges associated with encoding physical symmetries and equivariance properties into ML models of electronic structure. Upon overcoming these challenges, integrated ML-QM methods offer many interesting new opportunities. I will discuss several applications of ML-augmented quantum QC, including Inverse Chemical Design based on ML-predicted wave functions, the development of accurate semi-empirical methods, and the prediction of electronic spectroscopy.