Quantum Machine Learning for Accurate and Low-Cost Computational Chemistry

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Quantum mechanical predictions of ground-state and excited-state potential energy surfaces and properties face a punishing balance between prediction accuracy and computational cost, creating demand for new methods and modeling strategies. Machine learning (ML) for electronic structure offers promise in this regard, although conventional approaches require vast amounts of high-quality data and offer limited transferability in chemical space. In the presented work [1-3], we focus on the goal of using ML to describe the post-Hartree-Fock (HF) correlation energy. Assuming willingness to incur the cost of a HF self-consistent field (SCF) calculation, we aim to describe the correlation energy associated with perturbation theory, coupled-cluster theory, or other post-HF methods. Our approach focuses on training not with respect to atom-based features, but instead using features based on the HF molecular orbitals, which have no explicit dependence on the underlying atom-types and may thus be expected to provide greater chemical transferability. Indeed, numerical results indicate that molecular-orbital-based machine learning (MOB-ML) yields striking accuracy and transferability across chemical systems at low computational cost.

[1] "Transferability in machine learning for electronic structure via the molecular orbital basis." M. Welborn, L. Cheng, and T. F. Miller III, J. Chem. Theory Comput., 14, 4772 (2018).

[2] "A universal density matrix functional from molecular orbital-based machine learning: Transferability across organic molecules." L. Cheng, M. Welborn, A. S. Christensen, and T. F. Miller III, J. Chem. Phys., 150, 131103 (2019).

[3] "Regression-clustering for improved accuracy and training cost with molecular-orbital-based machine learning" L. Cheng, N. B. Kovachki, M. Welborn and T. F. Miller III, *JCTC*, J. Chem. Theory Comput., 15, 6668 (2019).