Interpretable Graph-Network-Based Machine Learning Models via Molecular Fragmentation for Chemical Accuracy at DFT Cost

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Abstract

The failure of approximate methods, e.g., density functional theory (DFT), to compute accurate electronic structures and energies for complex systems is well documented. While the newest collection of dispersion corrected and long-range corrected density functionals offer improved results, the errors in thermochemical properties computed with DFT are often greater than 5 kcal/mol for many chemical processes. In this talk, we discuss a new fragmentation-based molecular representation framework "FragGraph" involving embedding fragment-wise fingerprints onto molecular graphs. Our QM/ML model is specifically designed for delta machine learning (Δ-ML) which learns the difference between properties calculated at two levels of theory. We aim to correct the deficiencies of approximate methods such as DFT to calculate molecular properties with an accuracy comparable to the most sophisticated and computationally intensive methods such as coupled cluster theory.

Our framework is based on many of the ideas from machine learning, fragmentation, and error-cancellation. Broadly, we combine the advantages of existing error-cancellation methods with standard molecular featurization techniques to develop a general framework for quantifying the molecular structure. More specifically, our method uses a molecular graph attributed with a collection of vectors representing each of the local environments of atoms based on fragments from the Connectivity-Based Hierarchy (CBH) of error cancellation schemes. The utility of our FragGraph representation is showcased in a QM/ML framework by using a state-of-the-art deep learning model to predict a variety of molecular properties. Each of the fragment-wise fingerprints are augmented by surrounding fragments through message-passing within a Graph Neural Network. Similar to standard fragmentation-based correction methods, each fragment ultimately provides a correction which, when summed, gives a total correction for a system. One of the key advantages of this approach is the inherent interpretability which allows the breakdown of the total Δ-ML corrections into individual group contributions elucidating the deficiencies of approximate levels of theory based on molecular fragments.