Making machine learning applications in density functional theory transferable

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Machine learning (ML) is increasingly extending the reach of quantum chemistry methods for molecular and materials simulations. However, in density functional theory, the primary workhorse for quantum simulations, using ML to address the limitations of human-designed density functional approximations (DFAs) remains elusive, as ML-based approximations suffer from severely limited transferability to unseen chemical systems. A prominent example is the DM21 functional which does not extrapolate to transition metals [1]. Improving transferability is essential if ML-based electronic-structure models are to support data-driven discovery across chemistry and molecular science. To address this challenge, we explore a real-space machine learning strategy for DFAs where energies are learned point-by-point in space through local energy densities. This training paradigm substantially expands the available learning signal, transforming a single molecular energy into thousands of local data points and thereby improving the data efficiency of neural network models. A key component of this approach is the derivation and implementation of energy densities from regularized perturbation theory [2], enabling real-space learning of energies [3]. In the presentation, we will illustrate how this framework improves transferability by presenting examples of reaction-energy predictions across chemically diverse systems, highlighting its ability to generalize beyond the chemical environments encountered during training.

References

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