Real-space machine learning of correlation density functionals

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Abstract: Machine learning (ML) has started to play a pivotal role in extending the reach of quantum chemistry methods for both molecules and materials. 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. We apply real-space ML of DFAs to address this challenge, where energy is learned point-by-point in space through energy densities. Central to our real-space learning is the derivation and implementation of correlation energy densities from regularized perturbation theory. This allows us to pursue two directions for real-space ML of DFAs from these energy densities. First, we introduce the Local Energy Loss (LES), where model energy densities are trained against our implemented counterparts at every point in space. By expanding each system's single energy data point into thousands, LES dramatically enhances the transferability of ML DFAs compared to traditional global energy training. Second, we use spin-resolution of our correlation energy density to build a real-space, ML-based, and regularized extension of Spin-Component-Scaled second-order Møller-Plesset perturbation theory. We then show how the resulting model opens new avenues for the construction of transferable ML DFAs.