The CIDER Framework for Machine Learning Exchange-Correlation Functionals

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This poster provides an overview of the CIDER framework, a machine learning (ML) approach for training exchange-correlation (XC) functionals for Density Functional Theory (DFT). The design of better XC functionals is a central challenge of modern electronic structure theory. However, current developments are limited by the mathematical form of the functional, with efficient semilocal functionals being inaccurate for many technologically important systems and the more accurate hybrid functionals being too expensive for large solid-state systems due to the use of the exact exchange operator. CIDER combines exact constraints and Gaussian process regression to design functionals that are both orbital-dependent and nonlocal, but which can be evaluated at roughly the cost of semilocal functionals and are significantly faster than hybrid DFT in plane-wave codes. To demonstrate this approach, we train an exchange functional and substitute it into existing hybrid functionals to achieve hybrid-DFT accuracy on thermochemical benchmark sets. We also extend the CIDER framework to explicitly fit band gaps and other properties of the DFT eigenvalues. Using this approach, we train an ML functional for the exchange energy that predicts molecular energy gaps and reaction energies of a wide range of molecules in excellent agreement with reference hybrid DFT calculations. To demonstrate the potential of our approach to address challenging materials science problems, we use CIDER functionals to study point defects in semiconductors and polarons in ionic crystals, two types of systems that are plagued by self-interaction error when conventional semilocal functionals are used. Due to its transferability and computational efficiency for both molecular and extended systems, the CIDER framework makes significant progress on overcoming the cost-accuracy trade-off between semilocal and hybrid DFT, and our general approach provides a feasible path toward a universal exchange-correlation functional with post-hybrid DFT accuracy and semilocal DFT cost.