Machine Learning Kohn-Sham Density Functionals from Molecules

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Kohn-Sham density functional theory (DFT) [1] is widely used to simulate electronic structures of materials owing to the balanced computational cost and accuracy. Accuracy of the DFT depends critically on the accuracy of its approximated exchange-correlation functional. Conventional strategies to construct functionals are, however, consist of complicated and heuristic combination of physical conditions, leading to difficulty in systematic improvement of their accuracy. Also, available physical conditions are often insufficient to determine the intrinsically complicated structure of functional. In order to overcome those difficulties and establish systematic way to improve DFT, we present a new strategy to construct a functional, which is not relying on the complicated use of physical conditions: application of the machine-learning (ML) scheme.

Here, Neural-Network (NN), which is flexible ML model with a large number of parameters, forms the functional structure. Recently, the NN has successfully constructed the exchange-correlation functional for a model system [2]. We demonstrate NN-based constructions of functionals for real materials within several semi-local approximation, including local spin density approximation (LSDA), generalized gradient approximation (GGA), and meta-GGA (Fig. 1) [3]. The NNs in those functionals are optimized (trained) to reproduce training data, composed of densities and energies of a few molecules obtained by accurate wave-function theories such as the coupled cluster method.

The trained NN-based functionals show comparable accuracies to existing functionals, even for hundreds of molecules which are not included in the training database. Also, with the same ML procedure, we have successfully constructed a non-local functional, which is conventionally difficult to construct due to lack of physical conditions. Those results imply that our ML scheme is effective to overcome existing problems in DFT, with utilizing the advantages of the data-dependent functional construction.

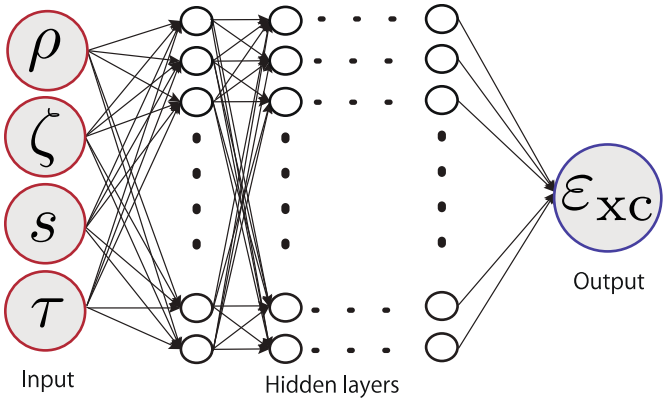

Figure 1: Structure of NN-based meta-GGA functional.

References

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