Analysis of Uncertainty in Machine Learned Density Functionals

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Summary

Density Functional Theory(DFT) is one of the most popular and successful methods for quantum mechanical simulations of matter because of its relatively lower computational costs. While it is theoretically exact, approximations of exchange correlation(XC) functionals have to be made. These calculations are highly time consuming and scale poorly with system size. The prospect of combining computer vision and deep learning is a fundamentally new approach to designing these XC functionals.

This approach combines the intuitive power of physical insight with the flexibility of machine learning and high-quality training data in order to develop new routes to approximating exchange-correlation energies. A parameterized function is first used to the data and the resulting residuals are used for bootstrap aggregating via an ensemble of neural networks. This two-stage method provides robust uncertainty quantification on the predicted XC energies and can potentially be automated for many systems without significant manual intervention.

Density Functional Theory

Accurate solutions to Schrodinger's Equation produce good predictions of the properties of molecular systems. The computational cost for calculating the Coulomb interaction amongst electrons rises exponentially with the number of electrons N. This makes direct solutions computationally intractable for all practical purposes (molecules with $N > 10$).

Kohn Sham DFT produces reliable predictions for such systems by mapping them onto a fictitious system of noninteracting electrons. However, the accuracy for KS DFT depends on the approximation of the XC functional. The XC functionals used for chemically accurate predictions are highly empirical and suitable only for a small class of molecular systems they are fitted to.

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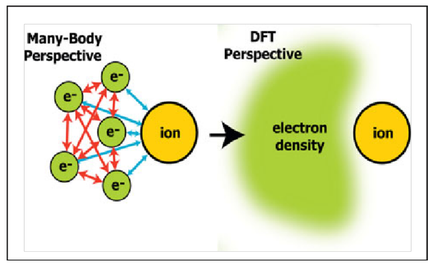

Figure 1. Traditional perspective vs. DFT.

Machine Learning

Machine learning (ML) is a powerful tool for finding patterns in high-dimensional data. Neural Networks are a class of ML algorithms loosely based on biological neural networks. With enough training data, neural networks act as universal function approximators. While these excel at classification tasks, single neural networks need a large amount of data to perform accurate regression in non-linear systems and do not produce uncertainty estimates. This is addressed by using an ensemble of neural networks through bootstrap aggregating (bagging). Bagging allows us to estimate uncertainty and helps avoid overfitting.

Training Workflow

According to KS DFT, a unique mapping exists between the electron density and the exchange correlation potential. We use the XC potentials derived from empirical XC functionals for multiple systems to train an ensemble of neural networks. This trained learner acts as a more generalized surrogate functional which can be used to estimate the XC energy of multiple systems.

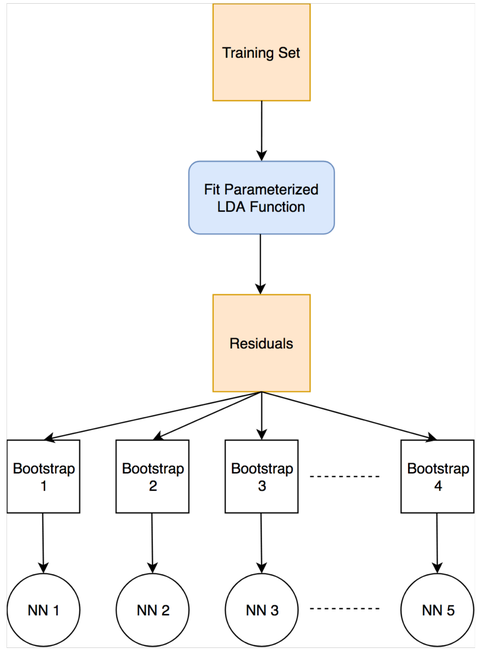

Figure 2. Ensemble Training Workflow.

Results Workflow

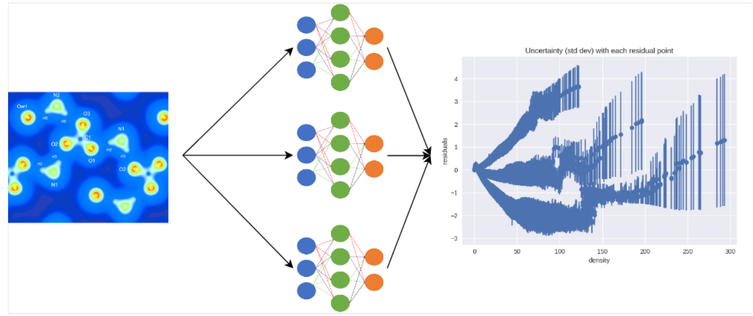

Figure 3. Prediction workflow. Produces Uncertainty Estimates.

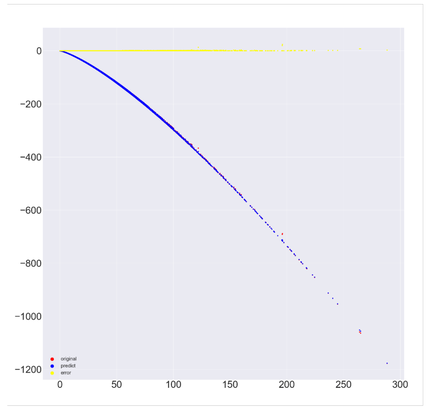

Figure 4. Electron Density Residual vs Potential plot. Error shown in yellow.

Conclusion

We demonstrated the viability of this workflow on a dataset consisting of 15 molecules resulting in a surrogate functional that generalized across all systems. For this demonstration, we used an ensemble for weak learners. While we did obtain the desired accuracy, we successfully estimated the uncertainty of predictions from our surrogate functional.

In the future, we hope to build on this workflow and achieve chemical accuracy by using an ensemble of more sophisticated learners.

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References

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