Density Functionals from Deep Learning

Jeffrey M. McMahon

Department of Physics and Astronomy, Washington State University

Introduction

Kohn–Sham density-functional theory (KS-DFT)[1,2] is a standard computational technique for studying materials, used in condensed matter physics and most branches of chemistry and materials science.

The accuracy and computational time of KS-DFT are limited by the approximations of density functionals that appear in the equations.

Density functional approximations are often based on:

- Nonempirical ones derived from quantum mechanics
- Empirical ones containing parameters fit to improve the accuracy on particular chemical systems

We propose an alternative approach to density-functional approximation, based on deep (machine) learning.

Deep Learning Model

We have developed a machine-learning model[3], based on a deep belief network (DBN)[4], to find a nonlinear mapping F:

$$F : \mathbf{n} \mapsto \mathbf{Z}$$

parameterized by the weights $\mathbf{W}$ of the DBN, which maps the input vector space of densities $\mathbf{n}$ to its feature space $\mathbf{Z}$.

Note that $\boldsymbol{F}$ is initialized in an entirely unsupervised way.

We then considered a probabilistic linear regression model with Gaussian noise to find a mapping from $\mathbf{Z}$ to a functional energy E:

$$E = f(\mathbf{z}) + \varepsilon \quad , \quad \varepsilon \sim \mathcal{N}(0, \sigma^2)$$

where the (density) function(al) $f(\mathbf{z})$ is distributed according to a Gaussian process (GP)[5].

Model System

We considered N noninteracting spinless electrons confined to a 1D box with a continuous potential. Our goal was to approximate the kinetic-energy density functional.

Performance statistics were selected so as to give a comprehensive assessment of a given model, as well as to allow a direct comparison between different ones:

- Normalized mean squared error (NMSE)
- Normalized mean bias factor (NMBF)
- Square of the sample correlation coefficient (r^2)

Kinetic-energy Density Functional

Performance for $N = 2$ to 8:

N	NMSE ($\times 10^{-6}$)	NMBF ($\times 10^{-4}$)	r^2
2	3.1(7)	−1.6(6)	0.977(4)
3	0.34(7)	−1.0(2)	0.93(1)
4	0.035(5)	−0.06(6)	0.960(5)
5	0.0076(8)	0.15(3)	0.951(5)
6	0.0017(3)	−0.07(1)	0.959(5)
7	0.0007(1)	0.002(8)	0.948(7)
8	0.00015(2)	−0.015(4)	0.970(3)

Performance for $N = 4$, using self-consistent densities:

NMSE ($\times 10^{-6}$)	NMBF ($\times 10^{-4}$)	r^2
0.46(3)	-4.0(2)	0.81(1)

The Mapping F

Improving the *representational power* of F:

n_{h1}-n_{h2}	NMSE ($\times 10^{-6}$)	NMBF ($\times 10^{-4}$)	r^2
25−10	0.13(2)	−0.3(2)	0.87(2)
25−25	0.059(7)	−0.4(1)	0.932(8)
50−25	0.034(3)	−0.2(1)	0.962(3)
125−50	0.020(3)	−0.17(5)	0.976(3)

Improving the *resolution* of F:

M_{ul}	NMSE ($\times 10^{-6}$)	NMBF ($\times 10^{-4}$)	r^2
100	0.046(4)	−0.37(6)	0.948(4)
200	0.043(5)	−0.23(7)	0.950(6)
500	0.034(3)	−0.2(1)	0.962(3)
1000	0.028(3)	−0.24(7)	0.970(3)

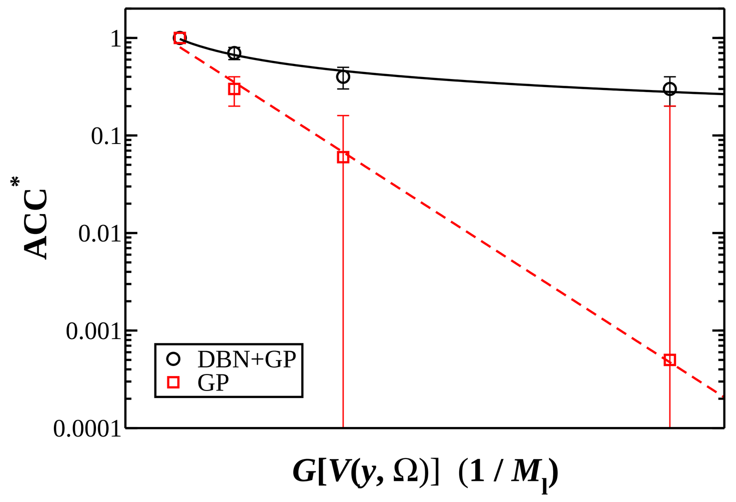
The Function(al) f

Improving the accuracy of f:

M_{l}	NMSE ($\times 10^{-6}$)	NMBF ($\times 10^{-4}$)	r^2
20	0.044(3)	−0.5(1)	0.951(3)
50	0.034(3)	−0.2(1)	0.962(3)
100	0.020(2)	−0.16(4)	0.975(3)
200	0.014(1)	−0.10(2)	0.983(2)

Comparison to Other Approaches

Example: Normalized accuracies (ACC*), in comparison to the use of a GP directly, as a function(al) of target variability $G[V(y, \Omega)]$:



Summary and Open Questions

Summary:

- A deep learning model was developed and applied to the problem of density-functional prediction
- The model performed well on approximating the kinetic-energy density functional for noninteracting electrons in a 1D box
- Advantages of the model were discussed

Open Questions:

- Can this approach be used in actual KS-DFT calculations? Perhaps in a self-consistent way?
- Can this approach be used in other problems for which invariance and sensitivity are needed — e.g., approximating potential-energy surfaces?

Acknowledgments

Start-up support:

Washington State University

Department of Physics and Astronomy

References

1. P. Hohenberg and W. Kohn, *Phys. Rev.* **136**, B864 (1964)
2. W. Kohn and L. J. Sham, *Phys. Rev.* **140**, A1133 (1965)
3. J. M. McMahon, *In Preparation* (2016)
4. G. E. Hinton, S. Osindero, and Y.-W. Teh, *Neural Comput.* **18**, 1527 (2006)
5. C. E. Rasmussen and C. K. I. Williams, *Gaussian Processes for Machine Learning (Adaptive Computation and Machine Learning)* (The MIT Press, 2005)

https://labs.wsu.edu/mcmahon/ jeffrey.mcmahon@wsu.edu