Constructing and Representing Exchange-Correlation Holes Through Artificial Neural Networks

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Kohn-Sham density functional theory (KS-DFT) is the most popular approach to calculate the electronic properties of matter. While KS-DFT is in principle exact, the functional for the exchange-correlation (XC) energy E_{xc} is unknown and must be approximated. Often, expressions for E_{xc} are based on a model for the XC hole $\varrho_{xc}(\mathbf{r},\mathbf{r'})$, since many of its properties are accurately known. $\varrho_{xc}(\mathbf{r},\mathbf{r'})$ represents the reduction of the probability density to find an electron at a position **r'** when a reference electron is at **r**. For the exchange (X) part, various successful hole models, such as the Becke-Roussel [1] or Ernzerhof-Perdew [2], were constructed by satisfying known physical constraints. However, only a few constraints can be addressed due to the complexity of finding simple functions that fulfill multiple conditions. An artificial neural network (NN), which can approximate almost any function, provides a tool to add more constraints. NN are already being used to develop XC functionals, they are trained with accurate theoretical/experimental data and automatic differentiation, implemented in the NN algorithms, provides access to the potentials [3]. In our work, we pursue an alternative approach by representing the exchange hole $\varrho_x(\mathbf{r},\mathbf{r'})$ with an NN. The X hole is determined by maximizing its information entropy, while satisfying various physical constraints. This is a variation of label free supervised learning [4]. Despite not using any experimental/computational data to optimize the NN, we obtain encouraging results. Furthermore, inspired by the correlation factor ($f_c(\mathbf{r},\mathbf{r'})$) models ($\varrho_{xc}(\mathbf{r},\mathbf{r'})=f_c(\mathbf{r},\mathbf{r'})\ \varrho_x(\mathbf{r},\mathbf{r'})$) developed by our group, we used a factor for a model X hole to consider additional constraint. For instance, a factor can adjust a X hole to reproduce a given exchange energy density while preserving other properties. Our work shows that neural networks are powerful tools to develop new approximations for the XC hole in KS-DFT.

[1] Ernzerhof and Perdew, J. Chem. Phys. 1998.
[2] Nagai, Akashi and Sugino, Npj Comput. Mater. 2020.
[3] Stewart and Ermon, Proceedings of the AAAI Conference on Artificial Intelligence. 2017.
[4] Roy, Cuierrier and Ernzerhof, J. Chem. Phys. 2020.