# Learning Exchange-Correlation Functionals from Nature

S.M. Vinko[1,2], S. Azadi[1], M.F. Kasim[3]

[1]*Department of Physics, Clarendon Laboratory, University of Oxford, Oxford OX1 3PU, UK*
[2]*Central Laser Facility, STFC Rutherford Appleton Laboratory, Didcot OX11 0QX, UK*
[3]*Machine Discovery Ltd, John Eccles House, Oxford Science Park, Oxford OX4 4GP, UK*

Recent advances in combining density functional theory (DFT) calculations of the electronic structure with machine learning are paving a new path towards the construction of chemically-accurate exchange-correlation (xc) functionals [1-7]. These advances promise to have broad application in the predictive modelling of real systems across chemistry, material science, and high energy density physics. In this context, differentiable programming is proving to be a formidable tool to help learn the behaviour of the xc functional, using a combination of higher-level simulations and experimental data, all within the physics-based constraints of the Kohn-Sham framework. Here we provide a brief outline of some recent developments in the field, and present the first fully-differentiable 3D density functional theory simulator (DQC – Differentiable Quantum Chemistry) where the exchange-correlation functional can be efficiently represented by a trainable deep neural network [8]. We demonstrate how this approach helps construct highly accurate exchange-correlation functionals using heterogeneous experimental data even for extremely limited datasets: using only eight experimental values on diatomic molecules, the trained exchange-correlation networks enable improved prediction accuracy of atomization energies across a collection of 104 molecules containing new bonds, new atoms, and new molecules not present in the training dataset [3]. The generalization power of this approach is particularly promising in the context of building accurate functionals for the exploration of extreme states of matter at high densities and temperatures.

[1] Nagai, Akashi, Sugino, Computational Materials 6, 43 (2020).
[2] Li et al., Phys. Rev. Lett. 126, 036401 (2021).
[3] Kasim, Vinko, Phys. Rev. Lett. 127, 126403 (2021).
[4] Dick, Fernandez-Serra, arXiv:2106.04481 (2021).
[5] Chen et al, J. Chem. Theory Comput. 17, 170 (2021).
[6] Kalita et al., arXiv:2110.14846v2 (2021).
[7] Nagai, Akashi, Sugino, arXiv:2111.15593v2 (2021).
[8] Kasim, Lehtola, Vinko arXiv:2110.11678 (2021). DQC code: https://github.com/diffqc/dqc/