Upscaling of Density Functional Theory calculations using Neural Networks

Martin Pimon, Johannes Gugler, Matthias Ebert, Paul Worm, and Peter Mohn

Center for Computational Material Science, Institute of Applied Physics, TU Wien

Summary: Currently, simulations of solids on a quantum level using Density Functional Theory (DFT) incur huge computational cost. More specifically, the simulated system's size has an upper limit of $\sim 10^3$ atoms on modern high performance clusters. Recent developments in the machine-learning (ML) domain have shown great promise alleviating this problem. Using modern ML libraries in combination with DFT training data has proven to significantly reduce the simulation time at similar levels of accuracy for large systems or long time simulated time scales. This is vital to study advanced materials containing difficult chemical compositions, phase boundaries or structural defects like dopants.

Description: Although DFT introduces astonishing simplifications [1] that make many-body calculations exceptionally fast compared to other methods with similar accuracy, it poses the problem of its time consuming quantum mechanical calculations. Nonetheless, there is still a limit to system size or time steps in a molecular dynamics (MD) run, which is well beyond the description of macroscopic phenomena for many real world scenarios.

However, the current workflow of DFT calculations is prone to throwing away important information, that could be used to achieve significant savings of computational power. As an example, the local surrounding of an atom in a MD run after a certain time step might be nearly identical to a previous step, which would mean that the atomic forces could be approximated using known information and the dynamics could be performed using classical mechanics only. Therefore, the algorithm would enhance DFT by saving known information to a database, making the simulation software *remember*.

A way to accomplish this is employing neural networks, which estimate the system's energy based on the generated DFT data of atomic positions. Accounting for the local surrounding of each atom only makes this approach versatile and transferable to arbitrary systems. Behler introduced a set of symmetry functions, which enable the unique identification of neighboring atoms by calculating so-called atomic fingerprints [2].

In order to conveniently use this approach in any scenario, the method has to be able to estimate if the current configuration is already known or if an expensive quantum mechanical DFT calculation has to be performed. In the second case, the algorithm would use the result to learn the neural network on-the-fly, improving the predictive performance of the neural network at each step.

Contents: In this talk we are going to present in detail the theoretical concepts underlying the method discussed above. First, we will give a short recap to machine learning, artificial neural networks and density functional theory. Then we discuss the sophisticated implementation of neural networks proposed by Behler and how it can aid the treatment of novel domains in system sizes or time scales. Finally, we present a method of estimating the error the neural network would make if it would predict the system's energy. Finally, we show our plan on how to construct the interface to a density functional theory code.

References

[1] Capelle, arXiv:cond-mat/0211443, (2006)

[2] Behler, International Journal of Quantum Chemistry **115**, 1032 (2015).