The road to accuracy: machine-learning-accelerated silicon ab initio simulations

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Simulating experimentally relevant situations ab initio is often computationally intense. Using hybrid approaches between ab initio methods such as density functional theory (DFT) and machine learning, models retain quantum accuracy while being computationally faster by several orders of magnitude. The methods are applied to Si.

Introduction

Ab initio models offer the ability to make high quality predictions of nanoscale properties at a rate much faster than is typically possible in experiment. This offers the possibility of creating large-scale ab initio databases using high-throughput virtual screening methods which can provide a useful guideline for more directed experiments. In practice, however, a tradeoff must often be made between the quality and quantity of the results, due to the computational time involved. Machine learning models offer a way to store quantum mechanical information from the relevant chemical space in a computationally efficient model and greatly accelerate the screening process. In this paper we consider two examples of such models, applied to silicon.

Active learning

The electronic behavior of silicon is both controlled and affected through the occurrence of native and foreign point defects. These sites can be occupied by many different elements in a variety of charge states which, combined with their mutual interactions, quickly increases the number of configurations. Active learning is a method where the DFT simulations are gradually replaced by a surrogate model. Figure 1 shows an example of a surrogate model to tasked to find the lowest energy pairs of substitutional defects, constructed from 72 elements. As expected, the most favorable defects, shown as blue in Figure 1(a), are pairs from group III-V. Figure 1(b) shows the uncertainty on these results provided by the Gaussian process model, blue regions are most certain. It shows that the model is most certain in the low energy regions, as desired. The predictions in the rest of the search space are still sufficiently adequate to observe the global trends, which is non-trivial since only 10% of the space was truly simulated with DFT.

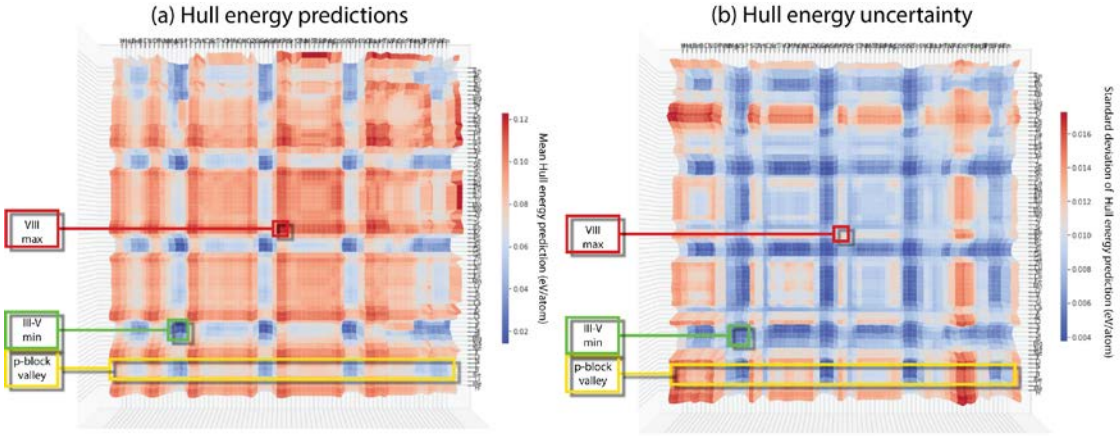

Figure 1: Gaussian process results for the hull energy of substitutional complexes in Si. (a) shows mean predictions, (b) shows the standard deviations on these predictions.[1]

Deep learning potentials

The models used in the active learning approach focus on constructing a simple model, quickly, using a limited amount of input data. Because the geometry does not change, using compositional information is enough to create a reliable model. Such a model, however, is not transferrable to new situations where the geometry has changed. Ideally, one would like to construct a model which can also process geometrical information, similar to an empirical potential, but which is capable of retaining the complexity of the quantum mechanical potential energy surface (PES). This would allow not only static energy evaluations, but also full molecular dynamics simulations. A new family of models based on deep learning offers the potential of achieving this. These methods directly fit the PES to the geometric inputs, bypassing the costly optimization of the electron density. Evaluating these models is several orders of magnitude faster than DFT, but achieve chemical accuracy. The only caveat is that the amount of input data required to train such a model is significantly larger than for a purely compositional search space.

Conclusions

Machine learning models offer great potential to accelerate computationally costly ab initio calculations. Our active learning approach was able to provide both an acceptable global overview and a precise result for our target configurations 10 times faster than a classical screening approach. Deep learning potentials offer the potential to extend this approach also to geometrical search spaces, retaining high precision at a low computational cost.

References

[1] M. Sluydts, M. Pieters, J. Vanhellemont, V. Van Speybroeck, and S. Cottenier, “High-Throughput Screening of Extrinsic Point Defect Properties in Si and Ge: Database and Applications,” *Chem. Mater.*, vol. 29, no. 3, 2017.