Moving Between Levels of Theory by Predicting Terms in the Many-Body Expansion for Lattice Energy Calculations

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Poster Abstract

The expense of high level ab initio calculations remains a bottleneck in many areas of computational chemistry, giving rise to a number of approximate methods to balance computational cost with accuracy. This is particularly a problem for crystal structure prediction, where the exploration of high-dimensional surfaces of periodic systems requires thousands of lattice energy calculations. Force fields and Density Functional Theory are typically used as a result, which can lead to ambiguity in identifying the most thermodynamically stable structures[1]. To address this, we introduce an approach to systematically improve low level calculations by using machine learning methods to predict terms in the many body expansion. Using the WilliamsRev6311 polarised force field and periodic PBE-D2/500 eV as a test case, we show that even only replacing two-body force field terms leads to significant convergence to the higher level of theory, and that these terms can readily be predicted by a neural network to a sub kJ/mol accuracy. We then describe a general framework for systematically selecting the best descriptor, model, and number of terms in the many-body expansion for a given system.

References

1. A.M. Reilly et al. , *Acta Crystallographica Section B Structural Science*, Vol 72, 439-539, **2016**