Automated uncertainty analysis and quantification for high-precision DFT calculations

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Methodological as well as computational progress in ab initio based atomistic simulations have substantially improved the predictive power in materials design over the last few years. A key aspect in achieving this high predictive power was to ensure that the large number of controllable parameters, which we have in state-of-the-art density functional theory calculations, guarantee a sufficiently high convergence. For finite temperature calculations, this commonly amounts to a convergence of better than 1 meV/atom in parameters such as supercell size, k-points, mesh sizes, energy cutoff etc. Doing this manually is from a scientific perspective routine work and thus not very appealing but expensive in labor and requires often months to train researchers adequately.

To automatize the complex uncertainty analysis and control we study and develop algorithms, which take the precision in the derived quantity as a convergence goal and automatically determine the convergence parameter to achieve it. The algorithms are implemented and tested using pyiron [1] (http://pyiron.org) – an integrated development environment (IDE) for computational material science. This tool provides an efficient and user-friendly environment for rapid prototype development and implementation of complex simulation protocols. Our investigations revealed that many of the commonly used rules of the thumb for choosing convergence parameters become invalid for high precision calculations.

References

[1] J. Janssen, S. Surendralal, Y. Lysogorskiy, M. Todorova, T. Hickel, R. Drautz, J. Neugebauer, Comp. Mater. Sci. **163**, 24 (2019).