Accelerating high-accuracy electronic structure theory

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Studying the electronic structure of systems containing more than a few atoms is computationally very demanding. For the most accurate methods, doubling the system size leads to over a 100-fold increase in the time and resources required. Thus, to investigate the behaviour of large-scale problems accurately requires the development both of new theories, and new technical approaches to their computation. Much research has recently been put in to finding linear-scaling coupled cluster approaches, with increasing success [1].

We present details on how our recently developed ALMO+RPA method [2] for intermolecular interactions can be extended to arbitrary levels of accuracy through stochastic methods. Moreover, we demonstrate how this lends the method to massive parallelisation, and the concomitant speed-ups this generates. In this way, we find it is possible to deal with solvated complexes of thousands of small molecules with coupled cluster level accuracy, using minimal computational resources. Extensions in the future will look towards using multilayer approaches to deal with even larger systems.

References

[1] M. Schwilk, Q. Ma, C. Köppl, and H.-J. Werner, J. Chem. Theory Comput. 13, 3650-3675, 2017

[2] R. A. Shaw, and J. G. Hill, *In preparation.*