Efficient Electronic Structure Theory via Scale-Adaptive Coupled-Cluster Formalism

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Coupled-cluster formalism is a *de facto* standard of accuracy in molecular electronic structure theory. Unfortunately, even the most basic approximations in coupled-cluster hierarchy, for example, CCD and CCSD, have a rather steep computational cost, $O(N^6)$ in this case, where N is the number of simulated particles. To overcome this problem, a plethora of the so-called local, or sometimes linear-scaling, approximations has been suggested, all of which are based on either fragmenting the underlying chemical system or introducing orbital interaction domains of bounded size or both, thus necessarily truncating the interaction scale and being unable to accurately describe non-local phenomena. We present an alternative approach based on the recently introduced formalism of scale-adaptive tensor algebra which imposes a hierarchical structure on many-body tensors. In contrast to the existing computationally efficient coupled-cluster approximations, in our formalism all parts of the chemical system interact with each other, thus enabling the description of non-local phenomena. Yet, the asymptotic memory requirements of the method are only $O(N)$ bounded and the computational cost is only $O(N\log N)$, thus making the method applicable to large chemical systems. Additionally, we also devised a way of including higher than double excitations in our scale-adaptive coupled-cluster ansatz without increasing the asymptotic memory requirements and computational cost, thus extending the applicability of the method to multireference problems. Furthermore, our formalism can also be extended to excited electronic states via the equation-of-the-motion approach with the same (low) computational complexity.

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