Rank-Reduced representations of the connected triples in Coupled Cluster theory

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Coupled cluster (CC) has proven to be one of the most successful quantum chemical methods, however, it is plagued by the presence of high-rank tensors: the two-electron integrals and the T (cluster) amplitudes. This high dimensionality leads to steep polynomial scaling. Reducing the scaling via rank- reduction presents two main difficulties: the lack of a clear prescription for the factorized (rank-reduced) form of the T amplitudes, and the complexity and extreme non-linearity of the rank-reduced coupled cluster equations. Despite these difficulties, many methods have been proposed, such as the singular- value decomposition coupled cluster with triple excitations (SVD-CCSDT) of Lesiuk, and the rank-reduced coupled cluster method of Parrish et al. (RR-CCSD and related methods). We discuss recent developments connecting rank-reducing factorizations of the integrals and doubles amplitudes (as in density fitting/resolution-of-the-identity and RR-CCSD), a Tucker-decomposed form of the triples amplitudes (as in SVD-CCSDT), and robust Approximation. We also explore graph-based diagrammatic techniques and knowledge-based algorithmic search through the Design-by-Transformation methodology in order to produce optimal working equations. Such automated techniques also enable new methods beyond the complexity of hand-derivation such as rank-reduced CCSD(T), SVD-CCSDTQ, etc. Reduced-scaling implementations of such methods will enable new model chemistries suitable for accurate thermochemistry (at the ~1kJ/mol scale) of molecules with as many as 12 first- or second-row atoms.