Parallel implementation of coupled-cluster methods based on the resolution-ofidentity approximation for two-electron repulsion integrals

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We present an algorithm for the parallel implementation of the coupled-cluster singles, doubles, and perturbative triples method [CCSD(T)] within the GAMESS suite of quantum chemistry programs.[1] The resolution-of-identity (RI) approximation is employed for the two-electron repulsion integrals (ERIs), which reduces the storage requirements of the conventional four-index ERIs by factorizing them as products of three-index ERIs via an auxiliary basis set.[2,3] The terms in coupled-cluster equations are evaluated in an integral-direct fashion by assembling the four-index ERIs from the three-index integrals, in this way bypassing the storage of the four-index ERIs completely. We employ a hybrid distributed memory/shared memory parallel programming model, which combines the General Distributed Data Interface[4] (GDDI) available in GAMESS with the thread-based parallelization scheme based on OpenMP. The GDDI model partitions *N* compute processes into *n* groups. The number of compute processes within each group is kept small, which permits a larger amount of shared memory to be associated with each of them. All requisite quantities for the CCSD(T) calculations are replicated on each group. Within each group, the three-index ERIs are distributed over the associated compute processes. The distribution of the computational workload follows the distribution of the ERIs. The workload on each compute process is further parallelized via large teams of OpenMP threads. One important advantage of using this hybrid GDDI/OpenMP model[5] is the scope of combining the RI-CCSD(T) implementation directly with the fragment molecular orbital (FMO) approach,[6] which is also based on the GDDI model. This opens up the possibility of applying the FMO approach in conjunction with a highlevel treatment of electron correlation effects to extended systems, e.g., biologically relevant molecules and materials.

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