Resolution-of-the-Identity Coupled-Cluster Algorithms for Heterogeneous Computing Platforms

Dipayan Datta and Mark S. Gordon
Department of Chemistry, Iowa State University, Ames, Iowa 50011, USA

We present an implementation of the coupled-cluster singles, doubles, and approximate triples [CCSD(T)] method within the GAMESS quantum chemistry program[1] for heterogeneous computing platforms consisting of CPUs and graphics processing units (GPUs). The resolution-of-the-identity (RI) approximation[2,3] is employed to factorize the four-index two-electron integrals as products of three-index two-electron integrals, which eliminates major memory bottlenecks of the CCSD(T) method. The Distributed Data Interface (DDI) implemented within GAMESS is employed to distribute computational workload over multiple compute nodes and multiple GPUs on each node. The offloading constructs of the OpenMP API are used for copying data between hosts (CPUs) and devices (GPUs) and for loop parallelization. Tenor contractions are performed with a GPU-accelerated math library, such as cuBLAS or hipBLAS. Large matrices are split into tiles along the row (or column) dimensions to fit big data into the relatively small device memory (16-32 GB). All of the time-consuming steps in the CCSD iteration and the non-iterative triples correction have been offloaded to GPUs. The GPU adaptation leads to 3x up to 7x speedup of the CCSD(T) computations depending on the system size compared to the recent CPU-only MPI/OpenMP parallel RI-CCSD(T) implementation[4] within GAMESS. Performance of the algorithm on heterogeneous compute nodes consisting of NVIDIA Tesla V100 GPUs is demonstrated.

1. Barca, G. M. J.; Bertoni, C.; Carrington, L.; Datta, D.; Leang, S. S.; Piecuch, C.; Rendell, A. P.; Ruedenberg, K.; Schmidt, M. W.; Slipchenko, L.; Gordon, M. S. *et al.* Recent developments in the general atomic and molecular electronic structure system. *J. Chem. Phys.* **2020**, *152*, 154102.
2. J. L.Whitten, *J. Chem. Phys.* **1973**, *58*, 4496-4501.
3. M. W. Feyereisen, G. Fitzerald, and A. Komornicki, *Chem. Phys. Lett.* **1993**, *208*, 359-363.
4. Datta, D.; Gordon, M. S. *J. Chem. Theory Comput.* **2021**, *17*, 4799-4822.