A Relativistic Fock-Space Coupled Cluster Method: Towards Efficient Execution on GPUs

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Relativistic Fock-space coupled cluster method (FS-RCC) seems to be the most promising tool for high-precision modelling of electronic structure of molecules containing heavy elements, especially in their excited electronic states. Such calculations are of crucial importance for modern molecular spectroscopy, chemistry and physics of ultracold matter as well as breakthrough research in fundamental physics. The computer implementation of the method is rather straightforward: the most time- and memory-consuming part of the FS-RCC algorithm may be reduced to the sequence of matrix-matrix multiplications and transpositions of multidimensional arrays. This fact makes it extremely attractive to parallelize calculations. A few GPU-parallelized CC codes implemented to the date aimed at the parallelization of nonrelativistic single-reference version of the method and were restricted to the CCSD model only (see, for example, [1]).

Here we present the way for performing FS-RCC calculations using GPUs for the most time-consuming operations (transpositions of multidimensional arrays and tensor contractions); the algorithm is able to work with operators of an arbitrary excitation rank and is optimized for complex arithmetic. The new coupled cluster program (called EXP-T) employing CUDA [2] was developed; the first results of the performance measurements are presented in Table 1.

Table 1. Time (in seconds) for the FS-RCCSD calculations of the Rb atomic energy levels (182 one-electron spinors) performed with the use of $C_{\infty v}$, C_s, and C_1 subgroups of its point group, measured for the new EXP-T coupled-cluster program on AMD FX-8320 (8) @ 3.497GHz / NVIDIA GeForce GTX TITAN Black (compared with the DIRAC17 program package [3]).

Symmetry	DIRAC17, 1 thread	EXP-T, 1 thread	EXP-T, 2 threads	EXP-T, CUDA
$C_{\infty v}$	387	334	255	1189
C_s	9931	3087	2323	2300
C_1	37302	9726	7290	4825

The dramatic increase of GPU performance for lower symmetries is due to the decreased overhead for manipulations with small, but very numerous blocks of integrals. This result clearly demonstrates the prospectivity of using the GPUs for relativistic coupled-cluster calculations.

References

1. DePrince III A.E., Hammond J.R. Coupled Cluster Theory on Graphics Processing Units I. The Coupled Cluster Doubles Method. // J. Chem. Theory Comput. 2011. Vol. 7, P. 1287–1295.

2. Nickolls J., Buck I., Garland M., Skadron K. Scalable Parallel Programming with CUDA. // ACM Queue. 2008. Vol. 6, No. 2. P. 40–53. DOI: 10.1145/1365490.1365500.

3. Visscher L. et al. DIRAC, a relativistic ab initio electronic structure program. http://www.diracprogram.org