Efficient and Cost-Effective Excitation Energy Calculations using Third-Order Cluster Perturbation Method

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Recently introduced Cluster Perturbation (CP) Theory [1] can be applied to overcome conventional Coupled Cluster Singles and Doubles (CCSD) excitation energy calculation steep scaling by avoiding explicitly solving the doubles eigenvalue problem, which is required to obtain the full spectrum of excitation energies. Instead, excitation energies from Coupled Cluster Singles (CCS) calculations are corrected one at a time based on the perturbation series in orders of the fluctuation potential up to the point where CCSD quality results are achieved. This series of corrections has been termed as the CPS(D) series and has already been applied out to the 6^{th} order correction. [2] Calculations have shown that the third-order correction, CPS(D-3), gives results in excitation energies of the CCSD quality in a fraction of computational time. Here we present further improvements of our massively parallel CPS(D-3) method implementation within the LSDalton program. [3] The CPS(D-3) can be formulated in an algorithm particularly suitable for a parallel computer. Furthermore, our implementation makes effective use of the GPU hardware available on Summit supercomputer. The resulting implementation may be applied to calculate excitation energies for system sizes previously only accessible using DFT methods. Additionally, due to inherit parallelism of the CPS(D-3) method combined with new high throughput and GPU-accelerated implementation the method may be applied in training neural networks for predicting excitation energies of CCSD quality.

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