Massively Parallel and GPU-Enabled Large Scale Excitation Energy Calculations from Cluster Perturbation Method

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The Cluster Perturbation (CP) Theory [1] can be applied to overcome conventional Coupled Cluster Singles and Doubles (CCSD) excitation energy calculation steep scaling. Within CP theory the explicit solution of the doubles eigenvalue problem can be avoided. 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 6th order correction.[2] Importantly, the 3rd order correction, CPS(D-3), can be formulated in an algorithm particularly suitable for parallel computers. Utilizing LSDalton program[3] built-in capabilities a massively parallel CPS(D-3) implementation can be achieved very rapidly. The implementation uses Scalable Tensor Library (ScaTeLib) for tensor contractions with GPU-offloading and the inherit parallelism of the CPS(D-3) to devise the method capable predicting excitation energies of CCSD quality for large molecular systems. With our new implementation systems of up to 1000 basis functions can be calculated in an hour of wall clock time. The scaling is very close to ideal up to 800 NVIDIA V100 GPUs.

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