

The divide–expand–consolidate (DEC) coupled cluster method. A linear–scaling approach with energy–based error control

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We present the Divide-Expand-Consolidate (DEC) coupled cluster (CC) model, where a CC calculation on a full molecular system is carried out in terms of calculations on small orbital fragments of the total molecular system. The fragmentation does not involve non-physical bond cuts and represents only an efficient way of dividing the calculation on a full molecular system into calculations on small orbital fragments. The sizes of the orbital fragment spaces are optimized during the calculation in a black box manner to ensure that the fragment energies are determined to a preset threshold. This in turn defines the total correlation energy as a sum of fragment energies to a preset threshold compared to a full molecular calculation. The number of independent fragment calculations scales linearly with the system size, and the method is therefore linearly scaling and embarrassingly parallel.

The fragmentation of orbital spaces relies on using a set of local Hartree-Fock (HF) orbitals. We use our recently developed orbital localization strategy where powers of the orbital variances are minimized to yield a set of local occupied and virtual HF orbitals.

DEC calculations are presented for the energy using second order Møller-Plesset perturbation (MP2) theory and the coupled cluster singles doubles (CCSD) models to demonstrate the performance of the DEC model. Calculations of the molecular gradient for MP2 will also be presented.