

Despite an immense progress in recent decades, a precise treatment of strongly correlated molecular systems still remains a challenge as of today. To help solve this problem, we have developed a massively parallel implementation of DMRG, called MOLMPS. For nonrelativistic systems requiring accurate treatment of both static and dynamical correlation, we have extended MOLMPS by the means of the almost linear scaling DLPNO-TCCSD method. In relativistic domain, we are the first who employed the 4c-CCSD to add dynamical correlation on top of DMRG, yielding the 4c-TCCSD method. When tested on benchmarks like the π -conjugated anthracene tetramer with CAS(63,63) and the FeMoco cofactor with CAS(113,76). We showed a good parallel performance on up to about 2000 CPUs. On the example of Iron(II)-Porphyrin model, we showed that the DLPNO-TCCSD captures 99.9% of TCCSD correlation energy. Our spectroscopic study on heavy diatomics showed that the 4c-TCCSD approach increases the precision of underlying CCSD to the order of CCSD(T) and that it is a promising approach. The thesis discusses three different implementations of quantum chemical methods based on QC-DMRG.