Pushing the limits of computations of strongly correlated molecules with the density matrix renormalization group method

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The quantum chemical version of the density matrix renormalization group (DMRG) method [1] has established itself as one of the methods of choice for calculations of strongly correlated molecular systems [2, 3]. Despite its great ability to capture strong electron correlation in large active spaces, it is not suitable for computations of dynamical electron correlation, which is, however, crucial for chemical accuracy.

In this talk, we will firstly introduce the combined approach [4] to the electronic structure problem of strongly correlated molecules, in which DMRG is responsible for a proper description of strong correlation, whereas dynamical correlation is computed via the adiabatic connection (AC) technique [5]. The great advantage of AC is that it requires only up to two-body active space reduced density matrices.

Secondly, with the aim of treatment of extended molecules with strongly correlated fragments, we have developed the projection-based DMRG-in-density functional theory (DFT) embedding [6], which comprises embedding of computationally demanding DMRG calculations in less-costly density functional theory (DFT) environment. The performence of both methods will be illustrated on selected benchmark problems.

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