NEW ELECTRON CORRELATION THEORIES FOR TRANSITION METAL COMPOUNDS

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Electronic structure theory faces many computational challenges in transition metal chemistry [1]. Usually, density functional theory is the method of choice because it is the only feasible one, although its results are not systematically improvable. By contrast, multireference ab initio methods could provide a correct description of the electronic structure, but are limited to small molecules because of the tremendous computational resources required. Considering the capabilities of transition metal compounds as catalysts [2] or molecular magnets [3], this long-standing dilemma deserves the continuous search for a solution.
In recent years, conceptually new ab initio methods emerged that turned out to be promising for theoretical coordination chemistry. We review and discuss two efficient parameterization schemes for the electronic wave function, the matrix product states and the tensor network states. The former parameterization is obtained in the density matrix renormalization group (DMRG) algorithm (for a recent review see [4]), while we have implemented a version of the latter for the first time for the full quantum chemical Hamiltonian [5]. The rather unexpected applicability of DMRG for compact electronic structures such as transition metal complexes and clusters had already been demonstrated in 2008 [6]. Only recently we have studied a cobalt tetraqua complex with the complete-graph tensor network approach [7]. Especially, tensor network states might provide the key to accurately describe strongly correlated and magnetic molecular systems in transition metal chemistry. But also DMRG calculations hold promises as an accurate electronic structure benchmark method, if the scaling-determining parameter, namely the number of renormalized system states, can be kept as small as possible, for which concepts from quantum information theory are valuable [8]. We also showed how a CI wave function can be efficiently reconstructed from a DMRG state [9].

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