From Electronic and Vibrational Structure to Quantum Dynamics with Matrix Product States

Markus Reiher

Lab. f. Phys. Chem., ETH Zurich, Vladimir-Prelog-Weg 2, 8093 Zurich, Switzerland
markus.reiher@phys.chem.ethz.ch

Tensor network states and in particular matrix product states, which can be efficiently optimized with the density matrix renormalization group algorithm (DMRG), turned out to be a versatile and efficient way to parametrize a many-particle wave function [1], which otherwise suffers from the curse of dimensionality in many-particle quantum mechanics. We have developed a second-generation, i.e., matrix product operator based DMRG program [2], which allows for a fast implementation of new Hamiltonians. For instance, we were able to quickly turn the 'fermionic' program for electronic structure problems into one that can treat vibrational structures [3] and even quantum dynamics [4]. Already these pilot developments turned out to be on a par with the best traditional methods of quantum chemistry. We were able to drive these calculations in a fully automated manner by developing the first protocol that enables the fully automated selection of active orbital spaces [5] that was later turned into the only software available for this purpose [6]. Other advancements concern the development of transcorrelated methods [7] and multi-particle quantum theories [8]. In my talk, I will present a basic introduction to these approaches and then demonstrate their power at challenging chemical problems.

[1] A. Baiardi, M. Reiher, The density matrix renormalization group in chemistry and molecular physics: Recent developments and new challenges. J. Chem. Phys. **2020**, *152*, 040903.

[2] S. Keller, M. Dolfi, M. Troyer, M. Reiher, An efficient matrix product operator representation of the quantum chemical Hamiltonian. J. Chem. Phys. **2015**, *143,* 244118.

[3] A. Baiardi, C. J. Stein, V. Barone, M. Reiher, Vibrational Density Matrix Renormalization Group. J. Chem. Theory Comput. **2017**, *13,* 3764.

[4] A. Baiardi, M. Reiher, Large-Scale Quantum Dynamics with Matrix Product States. J. Chem. Theory Comput. **2019**, *15,* 3481.

[5] C. J. Stein, M. Reiher, Automated Selection of Active Orbital Spaces. J. Chem. Theory Comput. **2016**, 12, 1760.

[6] C. J. Stein, M. Reiher. AUTOCAS: A Program for Fully Automated Multiconfigurational Calculations. J. Comput. Chem. **2019**, *40*, 2216.

[7] A. Baiardi, M. Reiher. Transcorrelated density matrix renormalization group. J. Chem. Phys. **2020**, *153*, 164115.

[8] A. Muolo, A. Baiardi, R. Feldmann, M. Reiher. Nuclear-electronic all-particle density matrix renormalization group. J. Chem. Phys. **2020**, *152*, 204103.