Computational Strategies for the Characterization of Strongly Correlated Chemical Systems

Alberto Baiardi, Markus Reiher

ETH Zürich, Laboratorium für Physikalische Chemie, Zürich, Switzerland
alberto.baiardi@phys.chem.ethz.ch

The full configuration interaction (full CI) method is the primary route towards exact simulations of strongly-correlated molecular systems, although its computational cost grows exponentially with the system size. The density matrix renormalization group (DMRG)[1] tames this prohibitive computational scaling and enables simulating complex molecular problems with full CI. In this talk, we will describe three new classes of DMRG-based quantum-chemical methods.[1] We will first introduce DMRG-based algorithms for calculating molecular vibrational properties both within[2–4] and beyond[5,6] the Born-Oppenheimer approximation. We will then apply the time-dependent DMRG method to simulate non-equilibrium phenomena in complex molecular systems.[7,8] Finally, we will introduce an explicitly-correlated DMRG variant based on the transcorrelated approach.[9]
In the last part of the talk, we will introduce a quantum computing method to solve the vibrational full-CI problem on quantum computers[10] and identify the hardware requirement to observe a quantum advantage over DMRG-based methods.

References

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

[2] Baiardi, A.; Stein, C. J.; Barone, V.; Reiher, M. Vibrational Density Matrix Renormalization Group. *J. Chem. Theory Comput.* **2017**, *13*, 3764–3777.

[3] Baiardi, A.; Stein, C. J.; Barone, V.; Reiher, M. Optimization of highly excited matrix product states with an application to vibrational spectroscopy. *J. Chem. Phys.* **2019**, *150*, 094113.

[4] Baiardi, A.; Kelemen, A. K.; Reiher, M. Excited-state DMRG made simple with FEAST. *J. Chem. Theory Comput.* **2021**, *18*, 430.

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

[6] Feldmann, R.; Muolo, A.; Baiardi, A.; Reiher, M. Quantum Proton Effects from Density Matrix Renormalization Group Calculations. *J. Chem. Theory Comput.* **2022**, *18*, 250.

[7] Baiardi, A.; Reiher, M. Large-scale quantum-dynamics with matrix product states. *J. Chem. Theory Comput.* **2019**, *15*, 3481–3498.

[8] Baiardi, A. Electron Dynamics with the Time-Dependent Density Matrix Renormalization Group. *J. Chem. Theory Comput.* **2021**, *17*, 3320–3334.

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

[10] Ollitrault, P. J.; Baiardi, A.; Reiher, M.; Tavernelli, I. Hardware efficient quantum algorithms for vibrational structure calculations. *Chem. Sci.* **2020**, *11*, 6842–6855.