Tensor Train Methods for Stationary and Dynamical General Many-Particle Problems

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In my talk, I will discuss some of the latest developments in my group on matrix product states in theoretical chemistry. These developments address rather diverse targets, which extend the general applicability of this type of tensor network states [1]:

We introduced transcorrelated DMRG (tcDMRG) [2] to address the nagging dynamic correlation problem. tcDMRG optimizes the right eigenvectors of non-Hermitian Hamiltonians with the imaginary-time TD-DMRG algorithm [7]. We found that tcDMRG applied to the two-dimensional Fermi-Hubbard Hamiltonian enhances convergence of DMRG with respect to the bond dimension [2]. The first implementation of tcDMRG for molecular systems has also been presented with density fitting to leverage the calculation of the two- and three-body integrals [3].

Furthermore, we introduced DMRG[FEAST] [4] which exploits the FEAST algorithm for the calculation of excited-state MPS wave functions. Although the demonstration of DMRG[FEAST] was in the context of vibrational DMRG (vDMRG) [5,6], it can be straightforwardly extended to electronic structure theory.

In the first implementation of tangent-space time-dependent DMRG for molecular simulations [7] we focused on vibronic and excitonic dynamics. Tangent-space TD-DMRG was then also employed in electronic structure theory to calculate absorption spectra with the correlation function formalism, dynamical response properties beyond the linear-response approximation, and ionization dynamics [8].

Moreover, we introduced the nuclear-electronic all-particle DMRG method [9] with stochastic optimization of all wave function parameters to obtain ground and excited state energies and electron-nuclear orbital entanglement [10].

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