A DMRG-based Framework for Large-scale Quantum Many-body Calculations

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Quantum chemical algorithms based on tensor factorizations are continuously expanding the scope of wave function-based molecular simulation methods. By leveraging very compact many-body wave function parametrizations, tensor network-based methods such as the density matrix renormalization group (DMRG) [1] can tame the computational cost of full configuration interaction (CI)-type calculations for strongly correlated molecular systems. While the DMRG algorithm is routinely applied to ground-state electronic structure problems, we present a versatile framework enabling its application to a broader range of many-body quantum problems [2,3]. We demonstrate the capabilities of our DMRG-based framework by introducing the *n*-mode vibrational DMRG method [4], which allows for an accurate treatment of the correlated nuclear problem for a reliable characterization of strongly anharmonic molecules described by complex potential energy surfaces. By expressing the vibrational CI wave function as a matrix product state, *n*-mode vDMRG can target systems with up to 30 fully coupled vibrational modes.

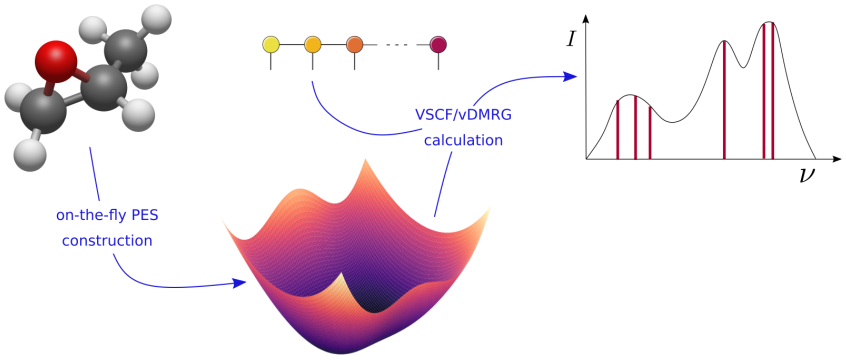


Figure 1: Illustration of the computational workflow of a *n*-mode vDMRG calculation

We further extend our framework to excited-states algorithms [5] enabling the large-scale calculation of both
low- and high-energy excitations. The combination of powerful tensor-based wave function representations
and efficient eigenstate targeting algorithms paves the route towards the accurate characterization of a
large variety of high-dimensional quantum systems.

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