New tensor product state approximations for strongly correlated molecules

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Developing accurate and robust ab initio methods capable of describing strong electron correlation remains a critical outstanding challenge for theoretical chemistry. Part of the difficulty in modeling strongly correlated systems arise from the fact that most of the approximation methods developed rely on a delocalized Hartree-Fock reference state, which becomes qualitatively incorrect as the strong interactions between electrons drives the system to a more localized structure. In this talk, I will discuss some of our group's recent work on developing new ab initio methods that are realized directly in a basis of tensor products of locally correlated many-body states. For systems that retain identifiable local character (excitonic excited states in molecular materials, low-energy levels in organometallic compounds, etc.), correlated tensor product states increase compactness of the wavefunction, allowing larger active spaces to be modelled. To illustrate the practical usefulness of theseapproaches, I will show recent numerical examples of biexcitonic excited states in singlet fission chromophores as well as low-energy states of organometallic complexes.