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Scalable and Accelerated Non-Orthogonal Configuration Interaction for Molecular Fragment Wavefunctions

ABSTRACT:
Methods based on delocalized molecular orbitals form the basis for most electronic structure studies in chemistry and biochemistry because of the computational advantages that the orthogonality of these orbitals provides. A renewed interest in the development and use of the computationally more demanding methods based on non-orthogonal orbitals is made possible by the availability of ever increasingly powerful computer systems. Such methods, including valence bond and non-orthogonal configuration interaction, provide more intuitive descriptions of molecular systems for specific classes of applications. An example is charge or energy transfer processes in molecular systems where a description in terms of individually optimized molecules or molecular fragments has significant advantages but results in non-orthogonal fragment wave functions.

GronOR is a new software development effort for non-orthogonal configuration interaction, as an international collaboration of the University of Groningen, Oak Ridge National Laboratory, and University Rovira i Virgili. GronOR is designed to construct anti-symmetrized spin-adapted product wave functions for large molecular systems from optimized multi-configuration fragment wave functions and use these as non-orthogonal basis states in subsequent configuration interaction calculations. The fragment wave functions used are complete active space self-consistent field (CASSCF) wave functions, but can, in principle, be generated by any multi-configuration electronic structure method. The computationally demanding evaluation of contributions to the Hamiltonian and overlap matrix elements from matrix elements between Slater determinants uses factorization of the second-order co-factor coefficient matrix.

Here we present the methodological development and implementation details of GronOR and demonstrate near-linear scalability and high accelerated performance on large, massively parallel, GPU-accelerated computer systems. Example calculations and a comparative benchmark analysis are given for different generations of GPU accelerators such as those available on Summit, one of the most powerful supercomputers in the US at the Oak Ridge Leadership Computing Facility, and on Juwels, one of the largest supercomputers in Europe at the Jülich Research Center.

BIO:
Dr. Straatsma is a Distinguished Research Scientist in the National Center for Computational Sciences at the Oak Ridge National Laboratory (ORNL) in Oak Ridge, TN, and Adjunct Professor of Chemistry at the University of Alabama, in Tuscaloosa, AL. He earned his Ph.D. in Mathematics and Natural Sciences from the University of Groningen. After postdoctoral and research assistant professor appointments at the University of Houston, he was a staff member and Laboratory Fellow at the Pacific Northwest National Laboratory, before joining ORNL. He leads the development of ARGOS, a molecular dynamics simulation application, and of GronOR, a highly scalable and accelerated application for non-orthogonal configuration interaction for molecular fragment wave functions. He is a core developer of the NWChem molecular science software suite.