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Development of Low-Order Scaling Multicomponent Methods

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Nuclear quantum effects (NQEs) are essential for accurately describing many chemical systems, particularly when light nuclei such as protons are involved. [1] However, their explicit quantum-mechanical treatment remains computationally demanding, especially for large and complex molecular systems.
To address this challenge, efficient low-scaling multicomponent quantum-chemical methods were developed within the Nuclear-Electronic Orbital (NEO) framework. [2] The developed methodology spans wave-function-based, density-based, and hybrid approaches, with a focus on improving their computational efficiency, scalability, and applicability to complex molecular systems. By combining integral-direct algorithms with density fitting and local approximations, the computational scaling and memory requirements were substantially reduced, enabling multicomponent calculations for systems with multiple quantum nuclei at previously inaccessible sizes. [3]
The resulting methods were applied to a broad range of systems, including protonated water clusters, DNA strands, hydrogen-bonded complexes, and molecular systems investigated experimentally in helium nanodroplets. They enable the investigation of proton affinities, structural changes induced by NQEs, vibrational hydrogen-bond shifts, and proton dynamics, and demonstrate cases where an explicit multicomponent treatment is essential to reproduce experimental observations. Together, these developments significantly expand the scope and applicability of multicomponent quantum chemistry and provide a foundation for the routine simulation of NQEs in complex molecular systems.

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[2] Pavošević, F., Culpitt, T., Hammes-Schiffer, S. Multicomponent Quantum Chemistry: Integrating Electronic and Nuclear Quantum Effects via the Nuclear-Electronic Orbital Method. Chem Rev 120 (9), 4222-4253 (2020) DOI: 10.1021/acs.chemrev.9b00798.
[3] Hasecke, L. Development of Low-Order Scaling Multicomponent Methods. University of Göttingen (2026) DOI: 10.53846/goediss-11783.