

Abstract Submitted
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Optimized orbitals with second order opposite-spin correlation

MARTIN HEAD-GORDON, University of California, Berkeley — Despite tremendous progress, the most ubiquitous electronic structure methods, based on density functional theory (DFT), that can be applied to molecules ranging well over 100 atoms, exhibit failures for molecules with strong correlations, some types of radicals, and systems where dispersion interactions are important. At the same time, the most accurate electronic structure methods, based on coupled cluster theory, remain too computationally demanding to enable the routine treatment of molecules containing more than about 20 atoms. I will discuss a new self-consistent approach that correctly and inexpensively recovers dispersion interactions, without either excessive spin-contamination for radicals (as plagues traditional unrestricted Hartree-Fock-based methods), or the difficulties of self-interaction that can affect DFT calculations of radicals. This approach yields optimized Breuckner-type orbitals. Its performance for relative energies, structures, and frequencies will be assessed, both for closed shell molecules, radicals, as well as some cases which exhibit pathological failures at both the DFT and MP2 levels of theory.

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