Functionals Based on the Exact-Exchange Energy Density: Beyond the Zero-Sum Game

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ABSTRACT

I will give an overview over some of the recent progress we have made in the field of local hybrid functionals [1] (LHs) and beyond, which opens new avenues. Position-dependent exact-exchange admixture of LHs like LH20t [2] allows a flexible treatment of properties depending on different spatial regions, e.g. in the core, valence, far away from the nuclei, or in different parts of a larger composite system. This has been explored, e.g., for NMR and EPR parameters, including the setup of new, extended benchmark sets for NMR shifts for main-group and transition-metal nuclei. [3] We recently extended the efficient implementation of LHs in Turbomole to range-separated LHs [4] and reported the ωLH22t RSLH. [4] It rivals the optimally-tuned RSH approach for quasiparticle energies in molecular electronics, without the need for system-dependent tuning. [5] The introduction of strong-correlation factors into LHs [6] and RSLHs (like ωLH23tdE or ωLH23tdB) [7] allows us to increasingly escape the usual zero-sum game between reducing fractional charge errors (self-interaction errors) and fractional spin errors (static correlation errors), providing powerful new DFT approaches. I will show real-life application examples including spin-symmetry breaking and hyperfine coupling in open-shell transition-metal complexes, [8] or NMR chemical shifts of main-group and transition-metal systems. [9] Many of the new methods have just become available with Turbomole 7.8.

References

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