Multiconfiguration Short-Range Density Functional Theory: Status and Perspectives

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Many interesting molecules exhibit strong correlation effects such as near-degeneracy static correlation and spin correlation. Reliable modeling of such molecules with Kohn-Sham DFT is often problematic. A primary objective for our work is to provide a reliable and affordable tool for such molecules, not the least for reliable description of transition-metal complexes as well as metallo-enzymes.

We advocate the MC-srDFT method, in which the long-range electron-electron repulsion is treated with MCSCF and the short-range part is treated with tailored density functionals. Compared to alternative methods as CASPT2 and MC-PDFT it is *fully* variational and therefore immediately applicable to electric, magnetic and optical response properties as well as geometry derivatives. Some examples are NMR shielding and spin-spin constants, frequency-dependent polarizabilities and hyper-polarizabilitites, as well as electronic excitation energies, two-photon absorption and transition moments.

Recently, several important developments and extensions have been made to this method in our groups (two examples [1-2]). It has been developed for open-shell systems and extended to molecular gradients and Hessians, making studies of chemical reactions possible for reactions involving both closed- and open-shell species, any spin state, and excited states. Currently we are implementing on-top pair-density.

In this talk I will present a status report for our MC-srDFT developments in Odense and discuss perspectives.

[1] E. D. Hedegård, J. Toulouse, and H. J. Aa. Jensen. “Multiconfigurational short-range density-functional theory for open-shell systems.” *J. Chem. Phys.* **148**, 214103 (2018); <http://doi.org/10.1063/1.5013306>

[2] F. K. Jørgensen, E. Kjellgren, H. J. Aa. Jensen, E. D. Hedegård. “Multiconfigurational short-range density functional theory for nuclear magnetic resonance shielding constants with gauge-including atomic orbitals.” *J. Chem. Phys*. **157**, 164106 (2022); <https://doi.org/10.1063/5.0106422>