MRSF-TDDFT:
A Good Way of Introducing Strong Correlation

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A new quantum theory, MRSF-TDDFT (Mixed-Reference Spin-Flip Time-Dependent Density Functional Theory) has been developed[*] for both ground and excited electronic states. With the help of a unique spinor-like transformation, a *hypothetical* single reference is constructed from the two M_S=+1 and -1 components of the RO-KS determinant, doubling its response space.

As a result, MRSF-TDDFT eliminates the problematic spin-contamination pitfalls of SF-TDDFT as well as the general topological problem of conical intersection by TDDFT. Unlike DFT, it allows to study open-shell ground singlet states such as diradicals. Furthermore, it produces the HOMO-to-LUMO *doubly* excited configuration, which is the main ingredient to properly account for the interplay of bright and dark excited states.

In short, MRSF formalism gives a balanced treatment of *dynamic and nondynamic* electron correlations, with the convenience of single determinant orbital optimization. Here, we highlight its advantages by presenting our recent results on excited state nonadiabatic dynamics, conical intersection, open shell singlet system, nonadiabatic coupling, spin-orbit coupling and X-ray absorption/ionization applications.

[*] (a) Lee, S., Filatov, M., Lee, S., & Choi, C. H. (**2018**). *J. Chem. Phys.*, 149(10), 104101. (b) Lee, S., Kim, E., Nakata, H., Lee, S. & Choi, C. H. (**2019**). *J Chem. Phys.*, 150(18), 184111. (c) Park, W., Shen, J., Lee, S., Piecuch, P., Filatov, M., Choi, C. H. (**2021**) *J. Phys. Chem. Lett.*, 12, 39, 9720–9729