

INSIGHTS FROM COMPUTATIONAL MODELING ON THE SINGLET-TRIPLET CONVERSION IN MR-TADF AND INVERT SINGLET-TRIPLET GAP MATERIALS

Yoann Olivier^{1*}

¹Laboratory for Computational Modeling of Functional Materials, Namur Institute of Structured Matter, Université de Namur, Rue de Bruxelles, B-5000 Namur, Belgium.

yoann.olivier@unamur.be

Keywords: OLEDs, TADF emitters, computational chemistry

Thermally Activated Delayed Fluorescence (TADF) process has appeared as the most popular design strategy towards reaching 100% internal quantum efficiency for Organic Light-Emitting Diodes (OLEDs). TADF consists in promoting upconversion of triplet excited states into emissive singlet ones through Reverse InterSystem Crossing (RISC), a process driven by spin-orbit coupling (SOC) and requiring a small singlet-triplet gap ΔE_{ST} . The advancement of the TADF field occurred essentially through materials design, the first strategy, as proposed by C. Adachi and co, consisting in connecting electron donating and accepting units to decrease the ΔE_{ST} . However, in doing so, the lower-lying singlet and triplet excited states bear a dominant charge transfer character that translates into a broad emission spectrum.

In this contribution, we will discuss based on computational considerations how doped triangle-shaped molecules can lead to (i) concomitant narrow emission, high quantum yield of emission and small ΔE_{ST} resulting in a whole new generation of TADF emitters, the multi-resonant TADF emitters^[1] and to (ii) a new family of compounds with an inverted singlet-triplet gap and potentially, a downwards energy RISC.^[2,3] To do so, we rely on high level quantum chemical calculations and show that an accurate description of electron correlation effects is key to correctly predict the excited states ordering as well as the optical properties of these compounds.^[4]

- [1] A. Pershin, D. Hall, V. Lemaire, J.C. Sancho-García, L. Muccioli, E. Zysman-Colman, D. Beljonne, and Y. Olivier, *Nat. Commun.* **2019**, 10, 597.
- [2] G. Ricci, E. San-Fabián, Y. Olivier, J. C. Sancho-García, *ChemPhysChem* **2021**, 22, 553.
- [3] J. Sanz-Rodrigo, G. Ricci, Y. Olivier, J. C. Sancho-García, *J. Phys. Chem. A* **2021**, 125, 513.
- [4] G. Ricci, J. C. Sancho-García, Y. Olivier, *J. Mater. Chem. C* **2022**, 10, 12680.