in silico design of organic molecules for triplet excited states manipulation

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Singlet-Triplet intersystem crossing (ISC) is a complex but common phenomenon in molecular spectroscopy. The combination of the improvement of spectroscopic techniques along with the molecular synthesis facilities allowed to design in the last years molecule families able to use this ISC for numerous applications such as in photodynamic therapy (based on singlet oxygen production) or light emitting devices (using the TADF phenomenon). In parallel to these experimental developments, the quantum chemistry revealed its capacity to understand the ISC phenomenon and beyond to design *in silico* new molecules able to use triplet excited states. These achievements are mainly obtained through TD-DFT, an approach able to provide a quantum description of chemically realistic systems.

In this talk, the effects of molecular architectures on the Spin-Orbit Coupling (SOC) and the ISC will be presented from the TD-DFT point of view and supported by experimental characterizations. We will first investigate how the control of the π-system distortion by molecular design allows to tune the SOC intensity and finally the ISC. The examples will be based on benzothioxanthene imide (BTXI) molecules, developed for OLEDs[1,2] and photodynamic therapy[3]. We will then move to dimers of BTXI. In such systems, the excitonic states obtained by the interaction of the monomers excited states allow a fine tuning of the SOC offering a new perspective on the manipulation of the triplet excited state populations (Fig. 1).[4] Beyond the molecular design aspect, these results also allow to understand the strength and limitations offered by TD-DFT to investigate triplet excited states.

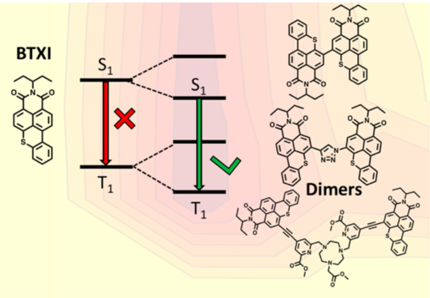

Figure 1: Schematic representation of the changes made by dimerization.

References

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