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Exploring Electronic Structure in Thermally Activated Delayed Fluorescence Emitters Using Wavefunction-based Descriptors

Thermally activated delayed fluorescence (TADF) systems, including multi-resonance (MR) and inverted singlet-triplet gap (INVEST) emitters, show great potential for applications such as organic light-emitting diodes due to their high emission efficiency. However, accurately predicting their electronic properties remains computationally challenging due to the delicate balance of electronic excited states involved, limiting efficient molecular discovery and design for industrial applications. This study investigates two important aspects of MR/INVEST emitters: their electronic structure and excited-state rate constants. First, we study the relationship between the electronic structure of MR/INVEST emitters and their energy gaps using wavefunction-based descriptors. These descriptors, originally proposed for ionic states, measure the distribution of charges in atomic centers based on the transition density matrix. Here, we directly analyze the transition density matrix connecting the singlet and triplet excited states involved in TADF. Our findings reveal a correlation between the wavefunction descriptor and the energy gap, with smaller (more negative) gaps corresponding to higher descriptor values. The descriptors also distinguish excited states involved in TADF within the triplet manifold, making it a potential cheap measure for identifying molecules with a small singlet-triplet energy gap. Then, we identified the potential deactivation pathways using vibronic calculations. We calculated excited-state decay rate constants, including fluorescence, direct and reverse intersystem crossing (RISC), where we assess the importance of Herzberg-Teller effects and broadening functions, among other parameters. Optimized structures and Hessians are obtained at (TDA)-DFT/CAM-B3LYP, and excitation energies are calculated using SCS-CC2 and ADC(2) to ensure accurate energy values for rate calculations and energy gaps. Overall, the calculated RISC rate constants align well with experimental values, typically within one order of magnitude, with the Herzberg-Teller correction playing a dominant role.

Primary author(s) : Mr. BADAWY, Youssef (KU Leuven); Prof. ESCUDERO, Daniel (KU Leuven)

Presenter(s) : Mr. BADAWY, Youssef (KU Leuven)