

## Unravelling Optical Properties Of Extended Core Conjugated Chromophores With Computational Strategies: From Single Molecule To Aggregates

Yasi Dai<sup>a</sup>, Sofia Canola<sup>a,b</sup>, Fabrizia Negri<sup>a,c</sup>

- a. Università di Bologna, Dipartimento di Chimica "Giacomo Ciamician", Via F. Selmi 2, Bologna, Italy
- b. Present address: Institute of Physics of the Czech Academy of Sciences, Cukrovarnická 10/112, CZ16200 Praha 6, Czech Republic
- c. INSTM, Udr Bologna, Via F. Selmi, 2, Bologna, Italy  
yasi.dai2@unibo.it

Carbon-based conjugated molecules with extended  $\pi$  cores can be considered as *building blocks* for carbon nanotubes and *models* for graphene nanoribbons and defected graphene. Among polycyclic aromatic hydrocarbons (PAHs), those possessing diradical character have undergone a strong resurgence of interest due to their potential applications in optoelectronic devices [1]. Beside small singlet-triplet gaps, one distinctive character of their optical properties is the presence of a low-lying double-exciton (DE) state dominated by the H,H $\rightarrow$ L,L configuration[2] which may influence photophysical properties such as two-photon absorption efficiency and quenching of the fluorescence. Thanks to their efficient  $\pi$ - $\pi$  interactions, aggregation in PAH derivatives affects remarkably the optical properties of their condensed phases [3] a fact that has a strong impact in real optoelectronic devices.

In recent years, we have been interested in modelling the optical properties of large conjugated systems, focusing not only on isolated molecules but also considering aggregation effects. The goal of this contribution is therefore twofold. On one side, we report our recent results on the investigation on the DE state of conjugated diradicals with cost-effective computational methods such as *TDUDFT*, *SF-TDDFT* [4] and *DFTMRCI* [5]. On the other hand, we present an investigation on the modulation of exciton states in aggregates of perylene di-imide, a well-known example of n-type organic semiconductor. To disentangle the crucial role played by charge transfer contributions to exciton states [6,7], we discuss a diabaticization procedure and a simple, dimer-based, model Hamiltonian able to reproduce the results of quantum-chemical calculations on full aggregates [8].

### References

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