Building the Bethe–Salpeter Hamiltonian in clusters from the monomers' contributions.

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ABSTRACT

The thorough understanding of excited state properties of molecular solids (e.g. crystal, films, interfaces) is essential to prompt the rational design of high-performance functional materials for applications in light harvesting, light emission, energy transfer. The excited states of supramolecular systems can be expressed in terms of the local intramolecular Frenkel Excitations (FE) and the non-local intermolecular Charge Transfer (CT) states (see Fig. 1). The computation of the excited states in these systems is a formidable task because of their large size and the difficulties arising from the need for a well-balanced description of FE and CT states. An original cost-effective method for the accurate description of excited states is introduced by using the Bethe–Salpeter equation (BSE) [1], which allows deriving an effective low-energy Hamiltonian on the basis of excitations and charges localized on single molecular fragments. The method employs a projective scheme that ensures a high-quality description of reference BSE eigenvalues and eigenvectors, fully exploiting the advantages of hybrid QM/MM embedding schemes [2] to account for the effect of the environment. In this presentation, I will describe the theoretical and practical details behind this method and discuss some applications to representative examples relevant to applications in organic opto-electronics.

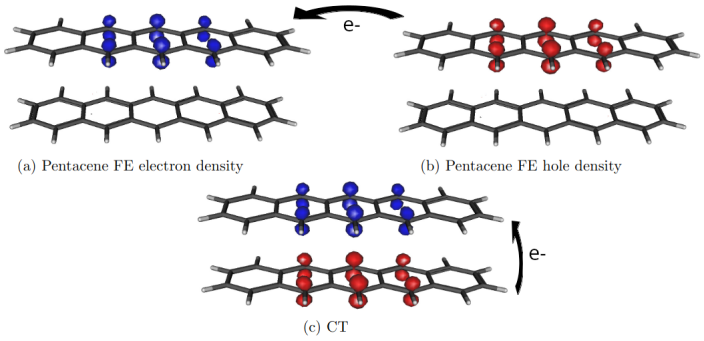

Figure 1: FE and CT electron-hole densities in Pentacene dimer.

References

[1] Blase, X.; Duchemin, I.; Jacquemin, D. The Bethe–Salpeter equation in chemistry: relations with TD-DFT, applications and challenges. *Chemical Society Reviews* **2018**, *47*, 1022.

[2] Li, J.; D'Avino, G.; Duchemin, I.; Beijonne,D.;Blase,X. Accurate description of charged excitations in molecular solids from embedded many-body perturbation theory. *Physical Review B* **2018**, *97*, 035108.