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PhD Project

Modelisation des états excitoniques au sein d'aggrégats moléculaires

Modeling of Excitonic States in Molecular Aggregates

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Molecular aggregates play a crucial role in the optical properties of several technologically relevant materials, including organic nanocrystals[1] and organic-inorganic hybrids[2-4]. For instance, the aggregation-induced enhanced fluorescence in organic nanocrystals has direct applications in biosensing for deep-imaging in diffuse tissues, which is used to investigate cancer development or brain diseases. Furthermore, the aggregation of organic dyes is known to significantly affect the optical properties of hybrid organic-inorganic materials, which have also attracted considerable attention for applications in biosensing[2] and photoenergy conversion[3]. The importance of molecular aggregation is due to the fact that photophysical properties of these materials are not only determined by the intrinsic electronic properties of the single molecular constituents but also by the excitonic coupling rising from molecular aggregation. Predicting and controlling the effects of aggregation in optical materials is thus of fundamental relevance for the design of materials with outstanding optical properties.

In this project, we aim at developing a computational tool that will be able to determine the photophysical properties of molecular aggregates in optical materials for direct comparison with experimental measurements. Theoretical investigations of molecular excitons are generally based on the point-dipole approximation, which fails at (small) distances where orbital-overlap between the molecular systems is important and electronic interactions cannot be neglected. In this project, increasing quality of molecular excitons characterization will be achieved by employing transition density matrix theory[5] and hybrid excitonic modeling. Moreover, state-of-the-art modeling techniques will be employed to extend the size of the quantum-mechanical treatment of the multi-chromophoric systems (supermolecular approach) and to include environmental effects present in condensed phases.[6] The expected outcome is novel tool and protocols to simulate linear absorption, two-photon absorption and emission spectra for comparison and interpretation of experimental data.

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