Unraveling white light emission and non-linear optical responses of chromophores using quantum chemistry

3-year PhD position
(Oct. 2020 – Sept. 2023)

Supervisors

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A full-time PhD position is available in the department of Inorganic Theoretical Chemistry (CTI) at the Institute of Chemical Sciences of Rennes (ISCR, University of Rennes 1) for a talented and ambitious student. The position is fully funded by the French Ministry of Higher Education, Research and Innovation.

Predicting and modeling the optical responses of chromophores such as those of interest for the next generation of white-light emitters or two-photon probes remains a challenge. If achievable at a low computational cost, it would save natural, human and financial resources. This thesis work proposes to tackle the problem by *combining state-of-the-art Time Dependent Density-Functional Theory (TD-DFT) calculations on newly designed synthetic targets with the development of a cost-effective protocol based on "Density Functional Tight-Binding" (DFTB).*

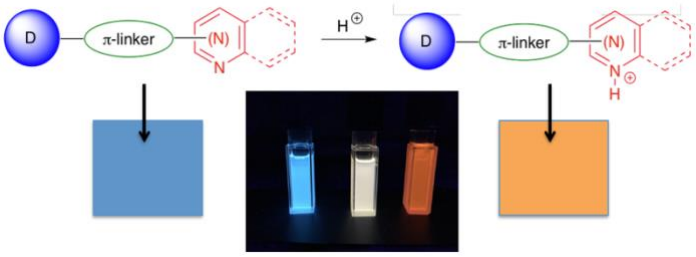

Figure: Change of color upon protonation to obtain white-light emission

On one hand, the project will be carried out in the context of long-term collaborations between experimentalists from our institute and abroad who develop organic molecules targeted for specific spectroscopic applications. Currently, we are actively seeking for novel classes of white organic light emitting diodes (WOLEDs). A new strategy consists in integrating a single emitting specie with two forms of complementary colors. White light emitting materials could be achieved based on an equilibrium between neutral and protonated forms (Figure) [1,2]. The next step is to increase the external quantum efficiency through triplet harvesting by thermally activated delayed fluorescence (TADF) [3]. Besides, we are also involved in the design of drug delivery systems demonstrating high uncaging efficiencies upon two-photon (TP) excitation using near-infrared light [4-6]. Both these developments require a fine tailoring of the chemical structure of the molecules, based upon a rationalization of the electronic structure and optical features with theoretical studies. *To this end, the PhD student will elaborate a state-of-the-art theoretical spectroscopy methodology, based on TD-DFT methods, able to predict qualitatively and quantitatively, the behavior of series of targeted molecules*.

On the other hand, such studies can involve considerable computational resources that may appear as the limiting factor at some point. In order to perform an efficient molecular design and allow considering ever more complex situations (larger compounds, explicit description of the environment), *the PhD candidate will also be in charge of developing a computational protocol based on the "Density Functional Tight-Binding" (DFTB) model*. Especially, the TD-DFTB formalism [7] would routinely permit a rapid description of the excited state properties for systems encompassing hundreds of atoms. This methodological work will be supported by another ongoing collaboration with the CMS group at the Bremen University. This will include assessment of the DFTB method' accuracy and its adaption to the properties of interest, starting with linear optical properties up to non-linear ones.

The CTI team

The PhD student will work in the Inorganic Theoretical Chemistry (CTI) team, in the Institute of Chemical Sciences of Rennes. The CTI team gathers several theoreticians (14 permanent staff members, 15 students) with complementary skills in theoretical chemistry but also physics, working with a broad set of quantum chemical tools, ranging from high precision *ab initio* wavefunction-based calculations to fast semi-empirical methods. The studied systems in CTI are diverse, including isolated species, bulk materials and surfaces, mainly of high experimental and societal interest. This has led to fruitful joint collaborations with experimentalists from ISCR as well as major national and international groups. The team is also strongly involved in the collective effort made by the French community of theoretical chemists at the national level, in the quest of bridging the gap between state-of the art quantum tools and real life applications. The CTI team thus provides a stimulating scientific environment, also offering regular team meetings, invited seminars as well as visitors internationally recognized. Local and national computing means are available for the purposes of the scientific projects.

Profile of the candidate

The PhD candidate should possess a general background in quantum chemistry. Prior experience with DFT/TD-DFT methods and/or theoretical spectroscopy is a plus. The work will be conducted with regular discussions and meetings with experimentalists and good command in English will be appreciated. Scientific curiosity and general knowledge in physical chemistry are also expected.

Application

The PhD project will start in October 2020. Applications are already open and candidates shall contact both supervisors by e-mail, with a CV and a motivation letter, including clear description of previous Master internship(s).

References:

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[3] R. Komatsu, T. Ohsawa, H. Sasabe, K. Nakao, Y. Hayasaka, J. Kido, *ACS Appl. Mater. Interfaces,* **2017**, *9*, 4742
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