“Two-color emission” fluorescence photoswitchable systems

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Abstract:
This PhD project focuses on the design and implementation of photoswitchable molecular assemblies to control Förster resonance energy transfer (FRET) over long distances. Playing on the specific organization of multiple fluorophores and photochromic units in 2D or 3D architectures, this research aims to develop a “two-color emission” fluorescence photoswitchable system. The study will explore cooperative and synergistic effects to improve FRET efficiency and control exciton transport. Understanding exciton diffusion anisotropy and optimizing photophysical mechanisms will be key objectives. The ultimate goal would be to integrate these materials towards molecular logic gate applications, paving the way for a fully optical transistor.

Details of proposed PhD position

Context: Förster resonance energy transfer (FRET) allows exciton diffusion between molecules with very weak coupling over characteristic distances of 1 to 10 nm. This process occurs between an energy donor and an acceptor, the incorporation and organization of multiple fluorophores facilitates exciton diffusion over extended ranges by exploiting homo-FRET and hetero-FRET phenomena (Fig. 1).[1]

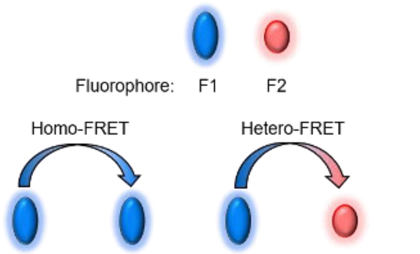

Figure 1. Scheme of homo and hetero-FRET

FRET is a fundamental mechanism in the development of photoswitchable luminescent devices, which are highly desirable for applications requiring high sensitivity and precise temporal and spatial control of optical states. At the molecular level, photoswitchable luminescent systems are designed by combining two key components: a luminescent material and a photochromic compound. Photochromic molecules isomerize reversibly between two states, both having different absorption spectra and geometry. When combined with a suitable fluorophore, given the appropriate spectral overlap and molecular alignment, fluorescence photoswitching from a dark state to a bright one can be achieved (Fig. 2).[2] Gathering these photoswitchable luminescent systems in nano-objects leads to intriguing fluorescence and photochromic responses due to multiple energy transfers.[3]. Up to now however, literature has mainly reported the case of systems switching between bright and dark states.

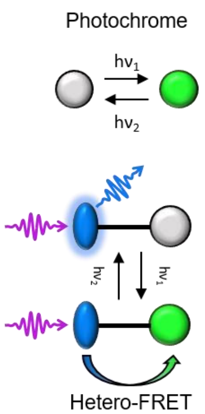

Figure 2.Scheme of photochromism and fluorescence photoswitching

Replacing the dark state with an emissive acceptor could extend exciton diffusion distances and enable their detection, this is the main motivation of the proposed PhD.

A crucial aspect of this research will be to understand exciton diffusion and determine whether anisotropic diffusion occurs. By extending the understanding of photophysical phenomena and optimizing FRET mechanisms, the project aims to achieve a "two-color emission" fluorescence photoswitchable system (Fig. 3) and apply this system to molecular logic gate applications, ultimately leading to the development of a fully optical transistor.

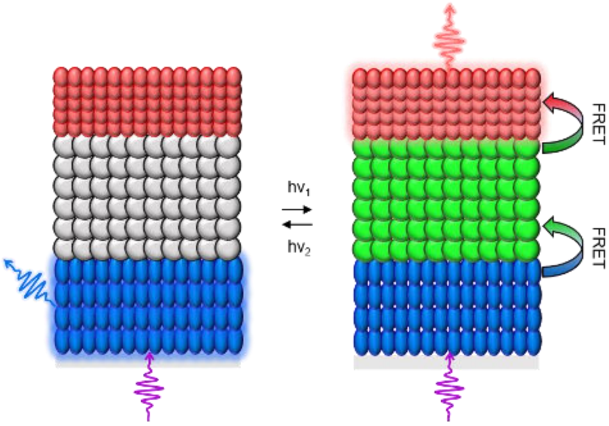

Figure 3. Scheme of the "two-color emission" fluorescence photoswitchable system.

Objective & methodology: The main objective is to control FRET through the design, implementation and characterization of complex functional molecular assemblies. The main tasks include

- Fabrication of specific photoswitchable 2D monolayers and 3D architectures with controlled organization of the different molecules.
- Characterization of the specific optical properties of these materials, with particular emphasis on exciton diffusion.
- Optimization of these molecular architectures, combining optical measurements and local probe microscopy to analyze fluorescence photoswitching in correlation with structural changes at the single molecule level.
- Demonstration of a "two-color emission" fluorescence photoswitchable system exploit cooperative and synergistic effects between fluorescent and photochromic systems as schematized in figure 3.

Thesis environment: **This thesis fully funded by CEA**.

The applicant should hold a Master degree and be highly motivated to work on an interdisciplinary subject involving physics and physical chemistry. The expected skills are: a strong background in optics or photophysics. Knowledge in physical chemistry would be appreciated.

The project will be held in collaboration with chemists, in France (Institut Lavoisier de Versailles, Institut parisien de chimie moléculaire) and abroad. A collaboration with a Japanese university is already planned. Scientific stay in Japan might be arranged during the thesis.

Please do not hesitate to contact us for further discussions together with a visit of the lab.

References:

[1] Wagner, R. W.; Lindsey, J. S. A. *J. Am. Chem. Soc.* **1994**, *116* (21), 9759–9760.[2] Irie, M.; Fukaminato, T.; Sasaki, T.; Tamai, N.; Kawai, T. *Nature* **2002**, *420* (6917), 759–760 [3] Fabre, N.; Fukaminato, T.; Ikariko, I.; Chocron, L.; Brosseau, A.; Métivier, R.. *Adv. Optical Mater.* **2024**, *12* (22), 2400452–2400458.