

Summary

Developing new organic materials is a global endeavour because it promises to bring key advantages, such as processability and scalability. The functional properties that can be pursued are widespread, ranging from structural bulk characteristics to the precise control of optical and electronic properties. In the following, the common ground of the different lines of research that will be described is organic chemistry, which provides us with an infinite playground of possibilities and makes it conceivable to contribute to several research domains. Beyond individual molecules that can be prepared with unique precision, supramolecular chemistry provides the next hierarchical step, either bridging the size gap or providing functionality at the molecular level.

This work is subdivided into three main topics: (i) Functional supramolecular polymers, in which we describe the role that shape and curvature play on the design and (self-)assembly of these materials; (ii) The use of molecular switches and motors for the modulation of either molecular shape or their intrinsic optical properties. We employ spirooxazines for the conception of switchable fluorescent probes enabling RESOLFT super-resolution microscopy protocols in cells, and diazoketone caged rhodamines for imaging nanostructures in organic solvents. On the other hand, overcrowded alkenes (i.e. molecular motors of the Feringa type) are investigated for the enhancement of their emissive properties as a result of reduced molecular motion controlled by light; (iii) We explore the structure-to-properties relationships in photo-generated triplet–radical systems, which can be seen as molecular qubit candidates. Here, we have studied various chromophores: perylene diimides (PDI), perylenes, and Boron Dipyrromethenes (BODIPY). Within each family, we have explored the effect of minor structural modifications and evidenced significant changes in the spin communication between the unpaired electrons in the excited state. Critically, we were able to optimize the spin coherence time *via* synthetic modifications (including isotopic substitution) and by controlling the viscosity of the medium.