

## Organic Nanoparticles Made from Polarizable Polar Dyes (PPD) : Dipolar interactions controlled self-aggregation, core-shell arrangement and their effect on surface properties and luminescence

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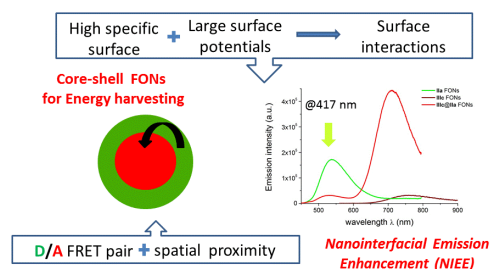
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In the last decades, luminescent nanoparticles have attracted growing attention in the field of bioimaging. In that context, fully organic nanoparticles offer interesting potentialities and among them self-assembled systems have received considerable attention as they offer potential for a range of applications. In that context, we have been interested in designing biocompatible organic nanoparticles combining tunable emission, huge brightness', excellent colloidal and photo-stability in bioenvironments, aiming at various imaging modalities (including *in vitro* and *ex vivo* single particle tracking [1] and *in vivo* two-photon (2P) imaging [2a-b]). To this aim, we have shown that subtle molecular engineering of specific dyes subunits - namely polarizable and (multi)polar dyes (PPDs) - leads the way to such fluorescent organic nanoparticles [1-2]. Indeed, the specifically designed PPDs spontaneously aggregate in water to yield molecular-based fluorescent organic nanoparticles (FONs) of controlled size (typically 10 to 100 nm in diameter) [1-2]. The self-aggregation process and self-orientation of the PPDs within FONs is controlled not only by **hydrophobic/hydrophilic interactions** but also by **strong dipole-dipole interactions**. Strikingly these **interactions influence the luminescence** and the 2P absorption properties of the resulting FONs [2a-b]. The self-orientation of the PPDs within FONs also defines the nature of the surface of those FONs and more specifically their interface with bulk water [2c], resulting in large surface potentials (typically in the -20 mV to -90 mV range [1-2]). The surface potentials and subsequent surface interactions can be exploited for the creation of **core-shell nanoparticles** from complementary PPDs [2c, 3]. These fully molecular-based core-shell nanoparticles (Fig.1) promote efficient **shell-to-core fluorescence resonant energy transfer and induce extraordinary fluorescence quantum yield and 2P absorption enhancement** [2b,3].

[1] a) E. Genin et al, Adv. Mater. **26**, 225 (2014); b) J. Daniel et al, J. Phys. D: Appl. Phys. **49**, 084002 (2016); c) M. Rosendale et al, Adv. Mater. in press (2021).

[2] a) V. Parthasarathy et al, Small **7**, 3219 (2011); b) C. Mastrodonato et al Molecules **21**, 1227 (2016); c) J. Daniel et al, ACS Photonics **2**, 1209 (2015).

[3] E. Campioli et al, Small **9**, 1982 (2013); E. Campioli et al, J. Mater. Chem. C **3**, 7483 (2015).



**Figure 1:** Design of molecular-based Core-Shell Fluorescent Nanoparticles made from PPDs for energy harvesting and Nanointerfacial Emission enhancement of luminescence and NLO properties