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T4. Spectroscopy

Energy and charge transfer pathways at the organic-lanthanide interface

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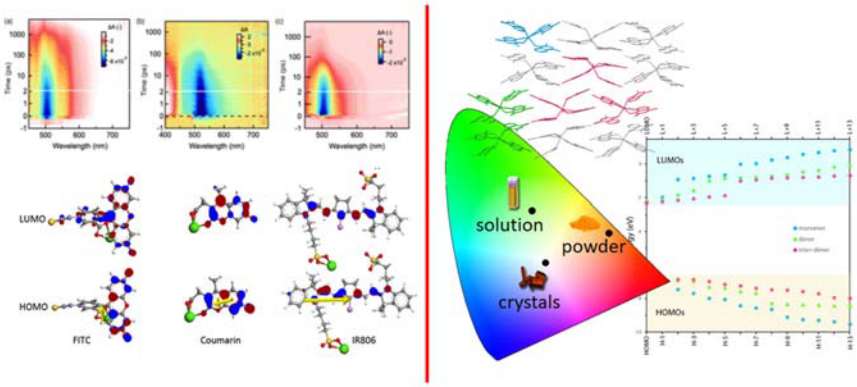
Controlling the multi-step emission photocycle in organically-sensitized lanthanide based systems is an exceptional opportunity for achieving efficiency enhancement and light color tunability. The chemical nature, shape, geometry, size and electronic density distribution of the organic sensitizer, not only have a striking influence on the overarching Förster's Resonance Energy Transfer pathways in the emission photocycle, but can bring novel functionalities through charge transfer routes.

In this contribution, we first show how ultra-fast transient absorption (TA) spectroscopy combined with time-resolved photoluminescence (PL) reveals the mechanisms and the short-lived intermediates at the organic-inorganic interface in dye-sensitized Ln^{3+}-doped fluoride nanoparticles. It is demonstrated that the sensitization efficiency from the dye to the doped nanoparticle is regulated by two types of regimes, "distance dominated" (ultra-fast route) and "resonance dominated" (slow route) depending on the geometry and localization of the transition dipole moment of the dye molecule.[1-2] Moreover, we discuss intra- and supra-molecular energy and charge transfer pathways leading to dual near-infrared and mechanochromic tunable visible orange-to-panchromatic white light emission in a dysprosium single molecular complex.[3]



Left. 2D TA maps showing the excited state ultrafast dynamics and DFT calculated frontier MOs and transition dipole moments in Ln^{3+} doped nanoparticles with three different dyes. Right. Supramolecular architecture, DFT calculated electronic levels and CIE color coding diagram for a Dy-quinolinolate single molecule mechanochromic emitter.

References

[1] F. Artizzu *et al.*, *Adv. Opt. Mater.*, **2021**, 2001678.
[2] R. Van Deun *et al.*, *Photonics Research*, **2021**, *9*, 2037.
[3] F. Artizzu *et al.*, *J. Mater. Chem. C*, **2021**, *9*, 15641.