Real-time tracking of the ultrafast chirality and energy transfer in a chiral OLED complex with circularly-polarized luminescence

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Circularly-polarized luminescence (CPL) has promising applications in the fields of optical data storage, biomedical diagnosis and for the creation of more efficient OLED displays. [1] In this respect, chiral lanthanide complexes have gained a lot of attention as CPL emitters due to their sharp and intense emission lines, long emission lifetimes and above all, due to their high degree of circular polarization. However, despite the impressive progress in the synthesis of metallo-organic CPL complexes, the underlying excited state dynamics of their chiral luminescent states have thus far remained inaccessible due to a lack of ultrafast spectroscopic techniques with sufficient chiral sensitivity.

To address this gap, we have developed a novel ultrafast circular dichroism instrument that combines ultra-sensitive broadband detection with sub-picosecond time resolution. [2] We now apply this technique to resolve the CPL mechanism of the prototypical $\mathrm{CsEu((+)\text{-}hfbc)_4}$ (hfbc = 3-heptafluoro-butylyrylcamphorate), which displays a record CPL emission with 85% of the photons being left-circularly polarized at 595 nm. [3] Although its luminescent transitions are metal-centered (MC), the chiral ligands play a crucial role in the CPL mechanism. First, they act as photosensitizers providing efficient energy transfer to the dipole-forbidden MC transition. Second, theoretical models predict that the high degree of luminescence dissymmetry arises from a transient coupling of ligand-centered electronic transition dipoles to the active MC transition of the Eu(III) ion, thereby creating the chiral luminescent excited state. [4,5]

Combining ultrafast circular dichroism with transient absorption and anisotropy measurements from the visible to the deep ultraviolet, we now present the first experimental investigation of the ultrafast energy and chirality transfer of a CPL complex, thereby providing a complete picture of the excited state CPL mechanism.

More generally, our results highlight that with ultrafast circular dichroism it is now possible to capture the evolution of chiral excited states in real-time, opening a new path to the direct investigation and optimization of enantioselective photochemical processes in their native solution phase.

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