

Twistable Molecules for Next Generation OLEDs

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Extended Abstract

Organic molecules present a multitude of new scientific challenges, as they are fundamentally different to existing classes of semiconductors. A key example is the role of quantum-mechanical spin. For instance, electrons and holes recombining in an OLED form Coulombically-bound excitonic states in a statistical mix of three spin-1 triplets to one spin-0 singlet. The excited state can only give out light if it can decay to the spin-0 ground state. Finding ways of harvesting light from spin-1 excitations has remained the fundamental problem in OLED technology for the last three decades, since the choice of strategy to achieve this necessarily impacts materials design, device architecture, and the processes limiting device lifetime.

Here we demonstrate a new approach to rapid triplet harvesting. We introduce a novel class of linear donor-bridge-acceptor light-emitting molecules which twist in their excited states, changing the coupling between electron and hole.[1] These enable doped polymer LEDs with near-100% internal quantum efficiency even at high brightness.[2] These materials achieve current efficiency, power efficiency and brightness comparable to or exceeding those of state-of-the-art vacuum-deposited OLEDs and quantum dot LEDs, despite being processed at low temperature entirely from solution.

Using time-resolved spectroscopy, we establish that luminescence via triplets occurs because molecular geometries exist at which the singlet-triplet energy gap (exchange energy) is close to zero. This allows rapid interconversion between spin states and, unlike other low exchange energy systems, substantial oscillator strength is sustained at the point where singlet and triplet cross.

We describe the experimental and theoretical evidence for the rotationally accessed spin-state mixing, and show how the resulting emission depends strongly in the interplay between rotational energetics, temperature, oscillator strength and the nanomorphology of the emissive layer. This gives us new tools to control emission colour and rate by changing the nano-environment of the emitting molecules. Doing so, we tune emission from green to sky-blue, and achieve external quantum efficiency at 1000 cdm^{-2} of nearly 30%.

References

- [1] A. S. Romanov *et al.*, “Copper and Gold Cyclic (Alkyl)(amino)carbene Complexes with Sub-Microsecond Photoemissions: Structure and Substituent Effects on Redox and Luminescent Properties,” *Chem. - A Eur. J.*, vol. 23, no. 19, pp. 4625-4637, 2017.
- [2] D. Di *et al.*, “High-performance light-emitting diodes based on carbene-metal-amides,” *Science*, vol. 356, no. 6334, pp. 159-163, 2017.