

Beyond the Trade-Off: Molecular Design of Dual-Functional 2PA-TADF Materials

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Abstract:

Thermally activated delayed fluorescence (TADF) has emerged as a key photophysical mechanism for achieving highly efficient organic light-emitting diodes (OLEDs) by harvesting both singlet and triplet excitons without the use of rare metals. In parallel, organic molecules exhibiting strong two-photon absorption (2PA) have attracted increasing attention for nonlinear optical applications, particularly near-infrared (NIR) bioimaging, where deep tissue penetration, low photodamage, and high spatial resolution are required. The integration of these two properties is highly attractive for realizing high-contrast, noninvasive imaging technologies. However, despite their individual importance, the simultaneous realization of efficient TADF and strong 2PA within a single organic molecule remains a major challenge due to fundamentally conflicting molecular design requirements.

Efficient TADF typically relies on twisted donor–acceptor (D–A) architectures that minimize HOMO–LUMO overlap, generating charge-transfer (CT) excited states with small singlet–triplet energy gaps (ΔE_{ST}) and enabling reverse intersystem crossing (RISC). In contrast, strong 2PA generally favors planar molecular structures with extended π -conjugation and large orbital overlap. As a result, conventional TADF emitters often exhibit limited 2PA cross-sections, whereas highly planar chromophores suitable for nonlinear optics tend to suppress TADF activity. Overcoming this trade-off represents a central challenge in the development of multifunctional 2PA–TADF materials. In this seminar, I will present a rational molecular design strategy that reconciles these opposing requirements by exploiting state-dependent electronic structures within a single chromophore. Experimental and theoretical studies demonstrate that our 2PA–TADF materials exhibit high two-photon absorption cross-sections exceeding 100 Göppert–Mayer (GM) in the NIR region, while maintaining small ΔE_{ST} values and accelerated RISC processes. When incorporated into OLED devices, these materials deliver high external quantum efficiencies (EQEs). These results show that precise electronic fine-tuning, rather than strict structural planarization or twisting, enables multifunctionality beyond conventional design paradigms, opening new opportunities in OLEDs and NIR bioimaging applications.

Reference:

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