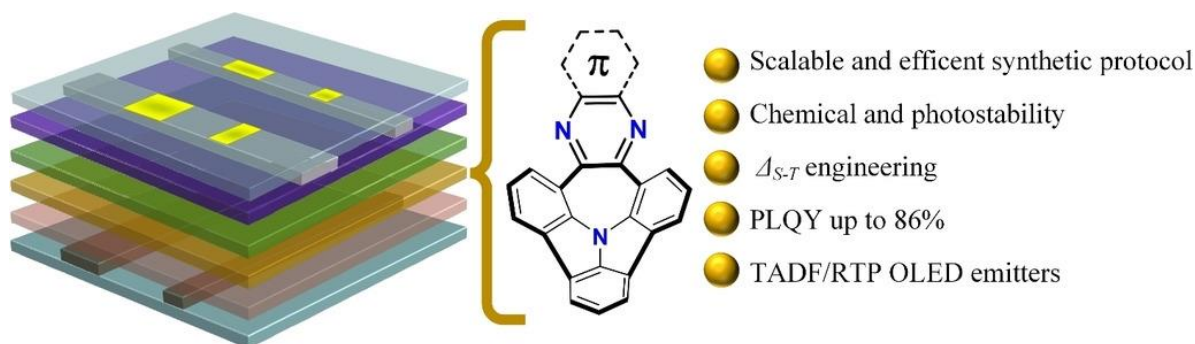


N-doped concave PAHs with fused donor-acceptor structure for high-performance OLEDs and tunable emission mechanisms

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The chemistry of polycyclic aromatic hydrocarbons (PAHs) plays a fundamental role in the development of optoelectronic devices including organic light-emitting diodes (OLEDs) or organic solar cells.¹ Nitrogen dopant in a polycyclic system, surrounded by 5- or 7-membered rings, results in a negative curvature of the structure and affects the energy of molecular orbitals.² Both factors influence the photophysical properties of PAHs.³ However, the use of N-doped bowl-shaped PAHs in optoelectronics is usually confined by low photoluminescence quantum efficiencies (PLQYs).



To address this issue, we report the synthesis and characterization of a series of curved and fully conjugated nitrogen-doped PAHs. The curvature increases the spatial separation between the highest occupied and lowest unoccupied molecular orbitals, leading to low singlet–triplet gaps, which are essential for high reverse intersystem crossing rates. Consequently, through structural modifications to the electron-accepting moiety, we are able to switch the mechanism of luminescence between thermally activated delayed fluorescence (TADF) and room-temperature phosphorescence (RTP), and to tune the overall PLQY in the range from 9 % to 86 %. As a proof of concept, we constructed solid-state organic light-emitting diode (OLED) devices, which has not been explored to date in the context of concave N-doped systems being TADF/RTP emitters. The best-performing dye, possessing a peripheral trifluoromethyl group adjacent to the phenazine acceptor, exhibits yellow to orange emission with a maximum external quantum efficiency (EQE) of 12 %, which is the highest EQE in a curved D-A embedded N-PAH to date.

References

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