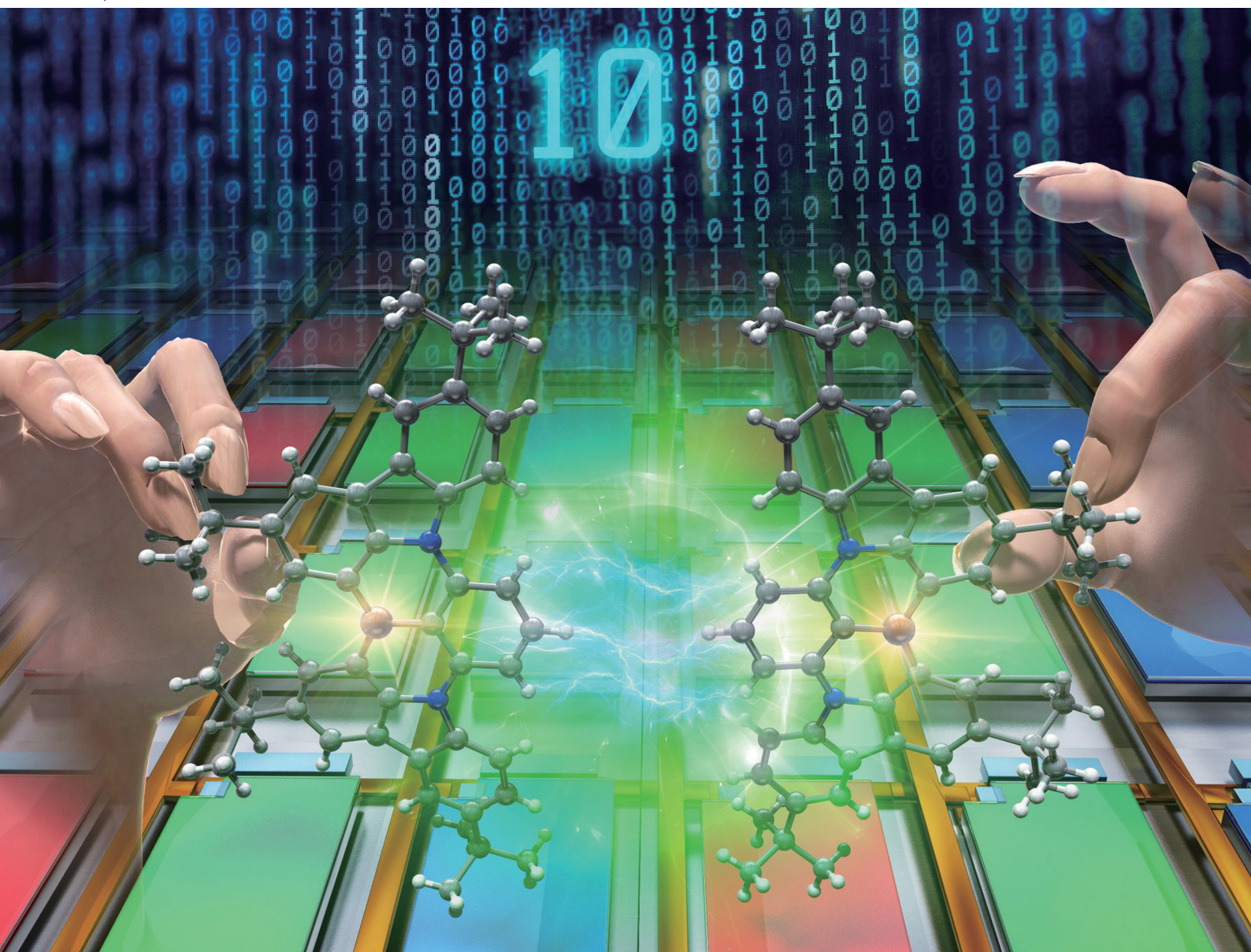


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Facile dimerization strategy for producing narrowband green multi-resonance delayed fluorescence emitters†

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Establishing a simple molecular design strategy for enabling redshifted emissions while maintaining high color purity in multi-resonance thermally activated delayed fluorescence (MR-TADF) remains a crucial yet challenging task. Herein, we introduce a new design concept based on a dimerization strategy for constructing pure green MR-TADF emitters. Two isomeric MR dimers, namely *p*-CzB and *m*-CzB, were developed by tethering two MR fragments through different linking positions. The interconnection mode between the two MR fragments in these dimeric MR-TADF systems plays a vital role in regulating photophysical properties as well as exciton dynamics. Comprehensive photophysical and computational studies revealed that *m*-CzB exhibits superior green MR-TADF characteristics compared to *p*-CzB. A *m*-CzB-based organic light-emitting diode (OLED) delivered pure green electroluminescence with CIE coordinates of (0.20, 0.70), a maximum external quantum efficiency of 23.5%, and alleviated efficiency roll-off.

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Introduction

Boron- and nitrogen-embedded polycyclic aromatic hydrocarbons (B,N-PAHs) have recently attracted burgeoning interest,^{1–9} which is ascribable to their promising use in optoelectronic devices as typified by organic light-emitting diodes (OLEDs). The opposite resonance characteristics of mutually *ortho*-disposed B and N atoms in B,N-PAHs are capable of inducing the multiple-resonance (MR) effect, leading to the localization and separation of the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) on different constituent atoms.^{5,6} This MR feature endows B, N-PAHs with a small singlet–triplet energy gap (ΔE_{ST}) and thermally activated delayed fluorescence (TADF) properties as a consequence.^{4–9} The biggest advantage of such an MR-TADF emitter is its narrowband emission capability, with an extremely small full width at half maximum (FWHM < 40 nm) enabled by suppressing structural relaxation and vibronic coupling in excited states. Combining the capabilities of full exciton use and narrowband emission leads to ideal emitters

for OLED applications, with superior electroluminescence (EL) efficiencies and color purities.

Three ultrapure red, green, and blue (RGB) primary emitters are essential for realizing wide-color-gamut OLEDs for use in ultrahigh-definition displays. The advent of sophisticated MR-TADF emitters, as represented by *ν*-DABNA, has led to the realization of high-efficiency pure blue EL.^{6,10–15} However, owing to atomically separated HOMOs and LUMOs, these MR-TADF emitters typically possess feeble intramolecular long-range charge-transfer (CT) characteristics; hence, large bathochromic shifts from the blue emission-band region are difficult to achieve. Therefore, new viable design strategies that expand the color gamut of MR-TADF emitters beyond the blue region are in strong demand. In this regard, we focused on **BBCz-SB**^{8,16} (Fig. 1) as one of the simplest B,N-based MR frameworks. Peripherally decorating **BBCz-SB** with either auxiliary donor or acceptor substituents on HOMO- or LUMO-dominant carbons has been reported to stabilize the respective energy levels, resulting in bathochromically shifted emissions.^{17–23} Our group reported the first deep-red MR-TADF emitter featuring a large fused polycyclic π -system based on *para*-B- π -B and *para*-N- π -N conjugation.⁸ High-efficiency green MR-TADF emitters have recently been developed by extending π -systems *via* additional ring fusion.^{9,24–27}

Herein, we unveiled a simple but effective dimerization strategy for producing pure green MR-TADF emitters. Two isomeric MR dimers, namely *p*-CzB and *m*-CzB, were designed

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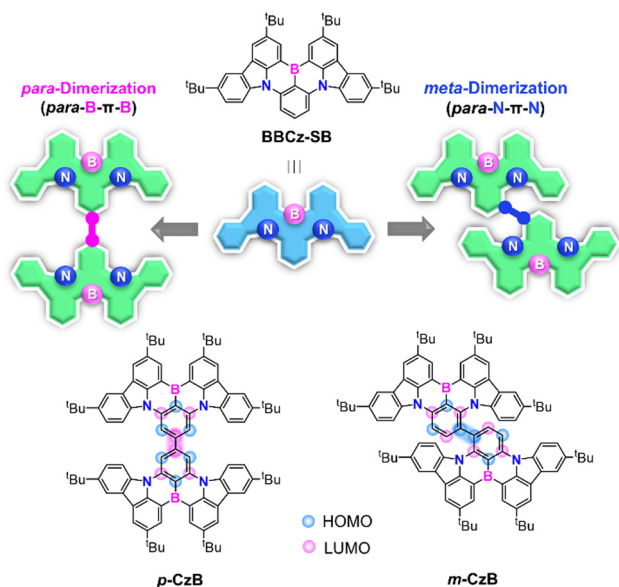
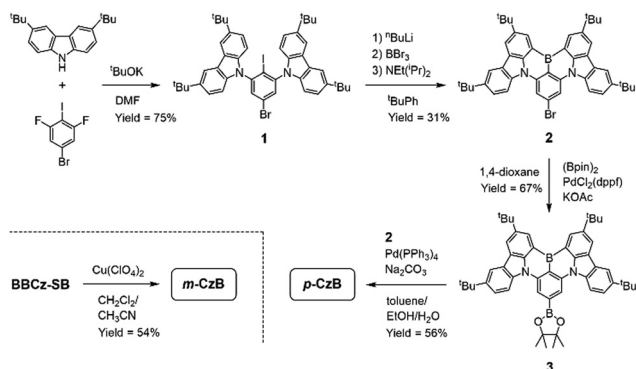


Fig. 1 Design of green MR-TADF emitters, *p*-CzB and *m*-CzB, based on the dimerization strategy.

by tethering two **BBCz-SB** fragments through different linking positions (Fig. 1). This dimerization strategy enables π -systems to be extended while maintaining intrinsic MR characteristics, thereby facilitating conspicuous bathochromic emission-band shifts without compromising narrowband spectral features. The dimerization strategy has previously been proposed for the design of TADF emitters;^{28,29} however, narrowband emissions with an adequately small FWHM have not been achieved owing to their strong CT character. Indeed, *p*-CzB and *m*-CzB exhibited intense green photoluminescence (PL) emissions that peak at 505 and 515 nm in solution, with narrow FWHMs of 34 and 35 nm, respectively. Furthermore, an OLED based on *m*-CzB as the MR-TADF emitter displayed pure green EL with a high maximum external quantum efficiency (EQE_{max}) of 23.5% and high color purity.

Results and discussion

Scheme 1 outlines the route used to synthesize the dimeric MR-TADF emitters; detailed synthesis procedures and



Scheme 1 Synthetic Routes to *p*-CzB and *m*-CzB.

characterization data are provided in the ESI.[†] The *para*-linked isomer (*p*-CzB) was finally prepared in 56% yield by Suzuki–Miyaura cross-coupling between precursors 2 and 3. Meanwhile, the *meta*-linked isomer (*m*-CzB) was synthesized in 54% yield by the oxidative coupling of **BBCz-SB** using $\text{Cu}(\text{ClO}_4)_2 \cdot 6\text{H}_2\text{O}$ as the oxidant.³⁰ Since the HOMO of **BBCz-SB** is largely distributed on the *para*- and *ortho*-carbons of the central benzene ring with respect to the *N*-carbazolyl substituents,⁸ oxidative coupling proceeded preferentially at this position to afford *m*-CzB in high yield. *m*-CzB has a distorted helicene-like structure with a *para*-N- π -N conjugated linkage. In addition, both dimeric *p*-CzB and *m*-CzB were found to be highly thermally stable with 5%-weight-loss decomposition temperatures (T_d) of 522 and 511 °C, respectively (ESI[†]), which are much higher than that of monomeric **BBCz-SB** ($T_d = 411$ °C).⁸ The final products were purified by vacuum sublimation for subsequent use.

The photophysical properties of *p*-CzB and *m*-CzB were investigated as dilute toluene solutions and doped thin films, the results of which are shown in Fig. 2, with relevant photophysical data summarized in Table 1. Both *p*-CzB and *m*-CzB exhibit vivid green PL in solution, with emission peaks (λ_{PL}) at 504 and 515 nm, respectively, which are redshifted by 15–26 nm compared to that of **BBCz-SB** ($\lambda_{\text{PL}} = 489$ nm). Despite each dimeric structure being linked through a single bond, *p*-CzB and *m*-CzB exhibited small Stokes shifts of 28 and 27 nm and emission FWHMs of 34 and 35 nm, respectively, suggestive of miniscule geometrical changes in their excited states. *m*-CzB has a higher molar absorptivity (ϵ) than *p*-CzB, corresponding to a stronger MR-induced HOMO \rightarrow LUMO transition. This trend eventually resulted in *m*-CzB exhibiting a higher PL quantum yield ($\Phi_{\text{PL}} = 90\%$) than *p*-CzB ($\Phi_{\text{PL}} = 85\%$) in solution. Solid thin films of *p*-CzB and *m*-CzB embedded in mCBP-CN³¹ (3,3'-di(carbazol-9-yl)-5-cyano-1,1'-biphenyl), a bipolar host, showed narrowband green PL emissions with $\lambda_{\text{PL}} = 513$ and 517 nm, and $\Phi_{\text{PL}} = 80\%$ and 85%, respectively (Fig. 2c, Table 1, and ESI[†]). The S_1 and T_1 excitation energies (E_S/E_T) of *p*-CzB and *m*-CzB were determined to be 2.43/2.29 and 2.40/2.31 eV, respectively, based on the peak wavelengths of their fluorescence and phosphorescence spectra (ESI[†]). Accordingly, *m*-CzB exhibited a smaller ΔE_{ST} than *p*-CzB (0.09 vs. 0.14 eV). Moreover, *p*-CzB underwent serious aggregation-caused emission quenching (ACQ) in the doped films with increasing the dopant concentration, whereas sterically distorted *m*-CzB exhibited slightly reduced ACQ effect, resulting in higher Φ_{PL} values (ESI[†]).

The transient PL decay profiles of the doped films of *p*-CzB and *m*-CzB show distinct two-component emissions (Fig. 2d). The prompt and delayed fluorescence lifetimes (τ_p/τ_d) were determined to be 9.6 ns/32 μs for *p*-CzB and 5.6 ns/15 μs for *m*-CzB. Benefiting from their prominent MR electronic features, both materials exhibited radiative decay rates (k_r) that far exceed 10^7 s⁻¹. Moreover, *m*-CzB (with the smaller ΔE_{ST}) exhibited a shorter τ_d , resulting in a k_{RISC} of 2.3×10^5 s⁻¹ (Table 1), which is three-times higher than that of *p*-CzB ($k_{\text{RISC}} = 4.7 \times 10^4$ s⁻¹). Notably, the k_{RISC} value of *m*-CzB is a

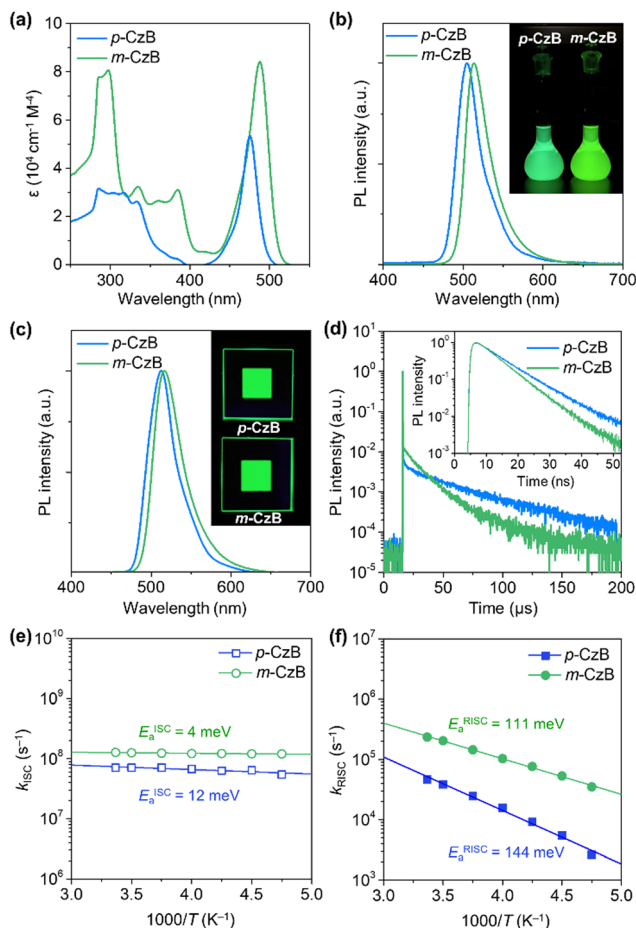


Fig. 2 (a) UV-vis absorption and (b) PL spectra of *p*-CzB and *m*-CzB in deoxygenated toluene (10^{-5} M) with a photographic image showing green emissions when illuminated by UV light (365 nm). (c) Steady-state PL spectra with corresponding emission images and (d) transient PL decay profiles acquired at 300 K for the doped films of *p*-CzB and *m*-CzB in mCBP-CN host matrices at a doping concentration of 1 wt%. The inset in (d) shows transient PL in the nanosecond (0–50 ns) region. (e and f) Arrhenius plots of k_{ISC} and k_{RISC} measured with the 1 wt%-emitter: mCBP-CN doped films, in which the solid lines represent the least-squares fittings for determining $E_{\text{a}}^{\text{ISC}}$ and $E_{\text{a}}^{\text{RISC}}$.

standout among reported green MR-TADF emitters (ESI[†]). To further understand exciton thermodynamic behavior, we examined the temperature dependences of k_{ISC} and k_{RISC} for the

doped films and determined the activation energies for ISC ($E_{\text{a}}^{\text{ISC}}$) and RISC ($E_{\text{a}}^{\text{RISC}}$) by fitting with the Arrhenius equation (Fig. 2e and f). While k_{ISC} showed negligibly small temperature dependences, k_{RISC} demonstrated clear positive temperature dependences, to give $E_{\text{a}}^{\text{RISC}}$ values of 144 and 111 meV for *p*-CzB and *m*-CzB, respectively, which are in reasonable agreement with the foregoing ΔE_{ST} values. These results support the notion that the smaller ΔE_{ST} of *m*-CzB accounts for its faster RISC.

To gain deeper insight into the nature of the excited singlet and triplet states, *p*-CzB and *m*-CzB were subjected to natural transition orbital (NTO) analyses using time-dependent density functional theory (TD-DFT) at the B3LYP/DZP level (Fig. 3). *p*-CzB has quasi-degenerate highest occupied NTOs that reflect its symmetrical dimeric structure, where hole wave functions reside on different B/N-based MR cores and give rise to doubly degenerate singlet (S_1 and S_2) and triplet (T_1 and T_2) excited states (Fig. 3a). In this case, the electron wave functions are located across the two MR cores through *para*-B- π -B conjugation. Meanwhile, *m*-CzB (with *para*-N- π -N conjugation) has degenerate lowest unoccupied NTOs; consequently, the electron wave functions are distributed in each of the two MR cores, while hole wave functions are extended to the dimeric MR structure (Fig. 3b). It is worth noting that the linking position significantly affects the resulting dimeric π -conjugated system and excited-state electronic properties.

We next calculated the spin-orbit coupling (SOC) matrix elements ($\langle S_m | \hat{H}_{\text{SOC}} | T_n \rangle$), $m = 1, 2$ and $n = 1-4$) for *p*-CzB and *m*-CzB (Fig. 3). As predicted by the El-Sayed rule,³² the SOC values for T_1 - S_1 and T_2 - S_2 couples with the same orbital occupations are rather small (0.13 and 0.12 cm^{-1} for *p*-CzB and 0.04 and 0.03 cm^{-1} for *m*-CzB) because of negligible changes in orbital angular momentum. In contrast, T_2 - S_1 and T_1 - S_2 couples exhibit distinctly larger SOC values that are ascribable to larger changes in orbital angular momentum (0.42 and 0.40 cm^{-1} for *p*-CzB and 0.36 and 0.54 cm^{-1} for *m*-CzB). We therefore envisage that such quasi-degenerate low-energy excited states induced by these dimeric MR structures contribute to SOC enhancement and offer additional T-S spin-flip channels for RISC.³³ Indeed, despite very similar ΔE_{ST} values, dimeric *p*-CzB and *m*-CzB exhibit RISC rates that are accelerated by factors of more than three and eight, respectively, compared to monomeric BBCz-SB ($k_{\text{RISC}} = 1.4 \times 10^4 \text{ s}^{-1}$ in toluene).⁸

Table 1 Photophysical data

| compound | state ^a | λ_{PL}^b (nm) | FWHM ^c (nm/eV) | Φ_{PL}^d (%) | Φ_{p}^e (%) | Φ_{d}^e (%) | τ_{p}^f (ns) | τ_{d}^f (μs) | k_{r}^g (10^7 s^{-1}) | k_{RISC}^h (10^4 s^{-1}) | E_{S}^i (eV) | E_{T}^i (eV) | ΔE_{ST}^j (eV) |
|---------------|--------------------|------------------------------|---------------------------|--------------------------|-------------------------|-------------------------|--------------------------|---------------------------------------|--|---|-----------------------|-----------------------|-------------------------------|
| <i>p</i> -CzB | Sol | 505 | 34/0.16 | 85 | 32 | 53 | 9.3 | 58 | 3.3 | 4.2 | 2.46 | 2.29 | 0.17 |
| | Film | 513 | 41/0.20 | 80 | 35 | 45 | 9.6 | 41 | 3.7 | 4.6 | 2.43 | 2.29 | 0.14 |
| <i>m</i> -CzB | Sol | 515 | 35/0.16 | 90 | 23 | 67 | 4.9 | 32 | 4.7 | 12 | 2.42 | 2.29 | 0.13 |
| | Film | 517 | 40/0.19 | 85 | 24 | 61 | 6.0 | 15 | 4.1 | 23 | 2.40 | 2.31 | 0.09 |

^a Sol = deoxygenated toluene solution (10^{-5} M) at 300 K; film = 1 wt%-doped film in a mCBP-CN host matrix at 300 K under N_2 . ^b PL emission maximum. ^c Full width at half maximum of the PL spectrum given in wavelength and energy. ^d Absolute PL quantum yield evaluated using an integrating sphere under N_2 . ^e Fractional quantum yields for prompt fluorescence (Φ_{p}) and delayed fluorescence (Φ_{d}); $\Phi_{\text{p}} + \Phi_{\text{d}} = \Phi_{\text{PL}}$. ^f Emission lifetimes for prompt fluorescence (τ_{p}) and delayed fluorescence (τ_{d}). ^g Rate constant for fluorescence radiative decay ($S_1 \rightarrow S_0$); $k_{\text{r}} = \Phi_{\text{p}}/\tau_{\text{p}}$. ^h RISC rate constant ($T_1 \rightarrow S_1$); $k_{\text{RISC}} = \Phi_{\text{d}}/(k_{\text{ISC}} \cdot \tau_{\text{p}} \cdot \tau_{\text{d}} \cdot \Phi_{\text{p}})$. ⁱ Lowest excited singlet (E_{S}) and triplet (E_{T}) energies determined from fluorescence and low-temperature phosphorescence spectral peaks for the doped films. ^j Singlet-triplet energy gap: $\Delta E_{\text{ST}} = E_{\text{S}} - E_{\text{T}}$.

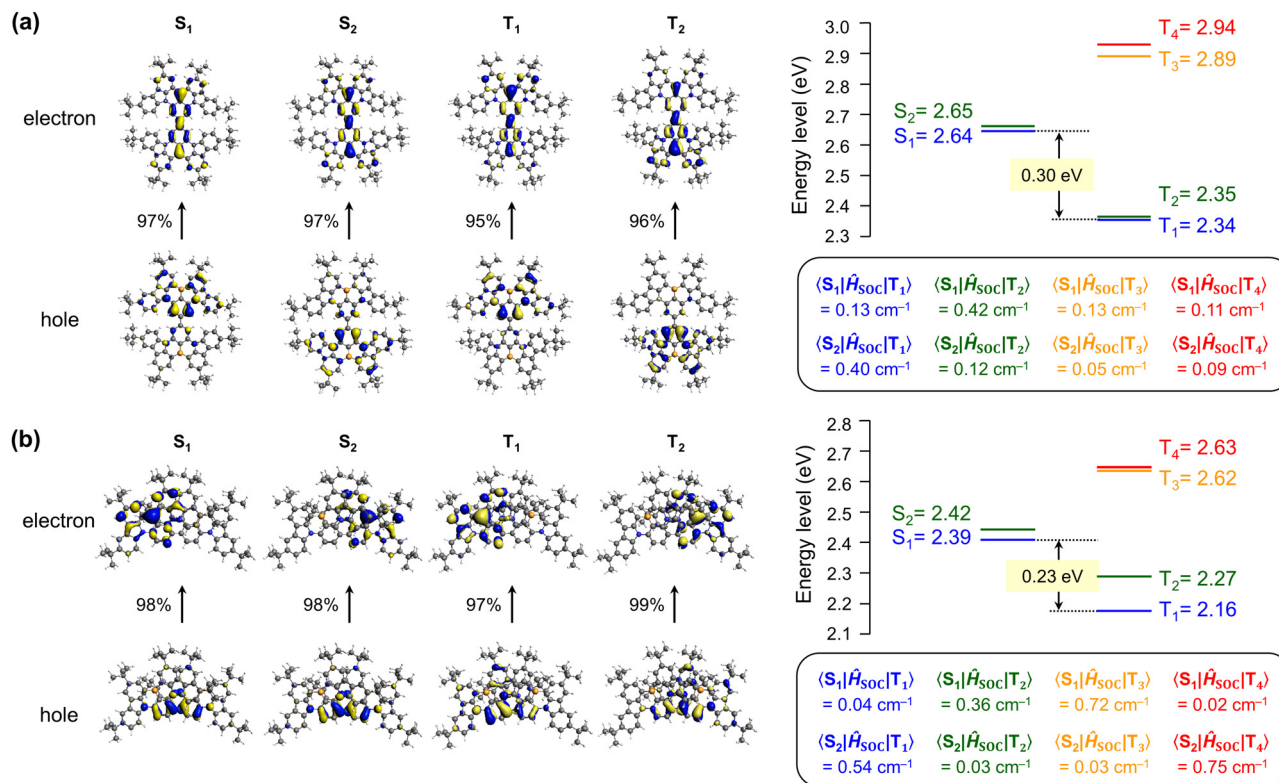


Fig. 3 Natural transition orbitals (NTOs) for the excited singlet (S_m) and triplet (T_n) states and simulated energy-level diagrams for (a) *p*-CzB and (b) *m*-CzB. NTOs, excitation energies for S_m and T_n , and associated spin-orbit coupling (SOC) matrix elements between the T_n and S_m states were calculated at the B3LYP/DZP level.

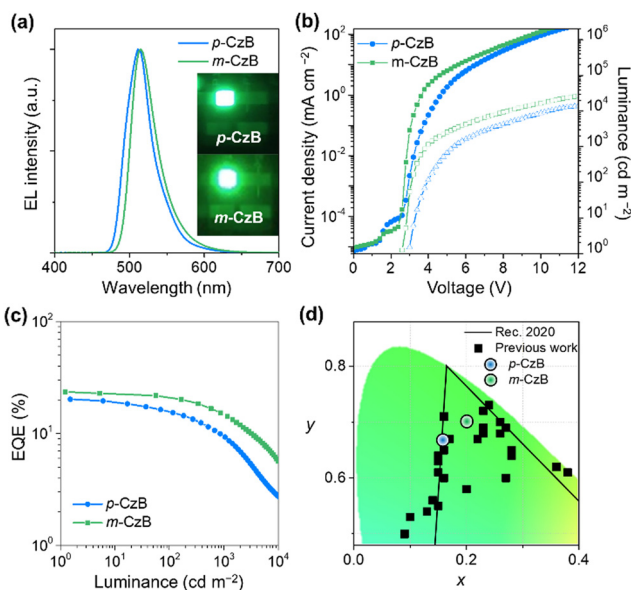


Fig. 4 EL characteristics of OLEDs that use *p*-CzB and *m*-CzB as MR-TADF emitters: (a) EL spectra and green EL images acquired at 10 mA cm⁻², (b) current density–voltage–luminance (J – V – L) characteristics, (c) EQE– L plots. (d) Comparing CIE coordinates for current and previously reported green OLEDs based on MR-TADF emitters (see ESI† for plotted data sets and source references).

Hence, the present dimerization strategy not only modulates the emission color but also favourably impacts excitonic spin conversion crucial for TADF.

To evaluate the actual EL performance of *p*-CzB and *m*-CzB, we tested TADF-OLEDs fabricated using the following device structure: indium-tin-oxide (ITO)/HAT-CN (10 nm)/TAPC (50 nm)/mMCP (5 nm)/1 wt% emitter:mCBP-CN (30 nm)/PPF (5 nm)/B3PyPB (50 nm)/Liq (1 nm)/Al (100 nm);¹³ the chemical structures of the materials are provided in the ESI† Fig. 4 shows EL spectra, current density–voltage–luminance (J – V – L) characteristics, and EQE– L plots for the fabricated TADF-OLEDs, and relevant EL data are summarized in Table 2. TADF-OLEDs that incorporate *p*-CzB and *m*-CzB display narrowband green EL with emission peaks (λ_{EL}) at 511 and 515 nm and FWHMs of 41 and 39 nm, which correspond to CIE chromaticity coordinates of (0.16, 0.66) and (0.20, 0.70), respectively. Notably, the green EL color of the *m*-CzB-based device fulfils the requirements of the National Television System Committee (NTSC) for pure green (0.21, 0.71) and approaches the coordinates of Rec.2020 pure green (0.17, 0.79)³⁴ (Fig. 4d). Moreover, these OLEDs delivered high maximum EQEs of 20.2% and 23.5%, respectively (Fig. 4c and Table 2). It is worth noting that the *m*-CzB-based device showed a more alleviated efficiency roll-off and retained high EQEs of 21.0% and 15.0% at luminances of 100 and 1000 cd m⁻². The relatively high k_{RISC} of 2.3×10^5 s⁻¹ for *m*-CzB accounts for the alleviated efficiency roll-off compared to that of *p*-CzB (4.6×10^4 s⁻¹) and contributes to suppressing

Table 2 OLED Performance

| Emitter | <i>p</i> -CzB | <i>m</i> -CzB |
|--|---------------|---------------|
| V_{on}^a (V) | 3.2 | 2.8 |
| λ_{EL}^b (nm) | 511 | 515 |
| FWHM ^c (nm/eV) | 41/0.20 | 39/0.19 |
| CIE ^d (x, y) | (0.16, 0.66) | (0.20, 0.70) |
| EQE _{max} ^e (%) | 20.2 | 23.5 |
| EQE _{100/1000} ^f (%) | 15.4/9.3 | 21.0/15.0 |
| CE ^g (cd A ⁻¹) | 67.9 | 85.1 |
| PE ^h (lm W ⁻¹) | 69.9 | 94.7 |

^a Turn-on voltage at a luminance above 1 cd m⁻². ^b EL emission maximum at 10 mA cm⁻². ^c Full width at half maximum of the EL spectrum given in wavelength and energy. ^d Commission Internationale de l'Éclairage (CIE) chromaticity coordinates recorded at 10 mA cm⁻². ^e Maximum external EL quantum efficiency. ^f External EL quantum efficiencies at luminance of 100 and 1000 cd m⁻². ^g Maximum current efficiency. ^h Maximum power efficiency.

detrimental exciton quenching, such as triplet–triplet annihilation (TTA) in operation at high luminance. These different EL characteristics are also reflected in the operational stabilities of the TADF-OLEDs. The *m*-CzB-based device exhibited an operational lifetime with a 50% luminance reduction (LT₅₀) of 25.5 h at an initial luminance (L_0) of 100 cd m⁻², which is approximately four-times longer than that of the *p*-CzB-based device (LT₅₀ = 6.4 h) (ESI[†]). These device stabilities still leave much room for improvement; however, isomeric *p*-CzB and *m*-CzB reveal that a subtle difference in linking position can greatly impact the efficiency roll-off behavior and operational stability in OLEDs.

Conclusions

In summary, we introduced a facile dimerization strategy for producing narrowband green MR-TADF emitters. Based on this strategy, we developed *para*- and *meta*-linked isomeric MR dimers, *p*-CzB and *m*-CzB. The position connecting the two MR cores serves as a conjugation valve that regulates the photophysical properties of the emitter. Both *p*-CzB and *m*-CzB exhibited distinct MR-TADF characteristics, concurrently delivering pure green emissions and narrow spectral FWHMs. Notably, the color purity and k_{RISC} associated with the *m*-CzB emission are standouts among reported green MR-TADF emitters. Consequently, the *m*-CzB-based green OLED achieved a considerably high EQE_{max} of 23.5% as well as alleviated efficiency roll-off, outperforming the *p*-CzB-based device. We expect that these results will expedite new material designs that feature the dimerization strategy for constructing wide-color-gamut MR-TADF systems.

Author contributions

M. Y., R. K. K., and T. Y. conceptualized the project. M. Y. and S. S. synthesized materials and performed quantum chemical calculations. M. Y. performed photophysical analysis and device evaluations. M. Y., R. K. K., and T. Y. co-wrote the manuscript. T. Y. supervised the entire research project.

Conflicts of interest

There are no conflicts to declare.

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