

Title: Organic spontaneous emission approaching the monochromatic limit

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Abstract: Spontaneous emission is inherently associated with spectral broadening mechanisms, resulting in finite bandwidth in the emitted light. Narrowing this linewidth toward the monochromatic limit has long been a central pursuit in photonics, as it determines the ultimate color purity of non-stimulated light sources. Organic luminescent materials offer facile wavelength tunability but typically exhibit broad emission bands (>40 nm). The emergence of multiple-resonance emitters has provided a promising route to overcome this limitation, yet most reported systems remain within 20–30 nm. Here we presented a molecular design strategy that amplified the multiple-resonance effect via molecular repetition, yielding fluorescence with linewidths of 6.9 nm in toluene, 5.5 nm in 3-methylpentane, and 9.1 nm in the film, placing this molecular framework among the narrowest-band organic luminophores reported.

Main Text:

Light emission is a fundamental process that defines how energy is converted into electromagnetic radiation across natural and artificial systems (1). Two distinct mechanisms—spontaneous and stimulated emission—govern light generation in light-emitting diodes (LEDs) and lasers, respectively (2). Stimulated emission provides monochromatic and coherent light but requires strong excitation energy to achieve population inversion and optical feedback. In contrast, spontaneous emission is a primary mechanism occurring with much lower energy input, enabling LEDs to offer low power consumption, compact size, and low manufacturing cost. Inorganic and organic LEDs (OLEDs) are well-established in commercial displays (3), and OLEDs, in particular, are envisioned for advanced lighting sources in flexible and wearable photonic devices. Although narrowband emission would greatly expand their application utility, spontaneous emission remains intrinsically spectrally broad. From an application perspective, narrow emission bandwidths are critically important for achieving highly saturated colors in displays, especially in the blue and green regions, where color purity strongly influences efficiency and color gamut coverage. Achieving fluorescence with spectral sharpness approaching that of stimulated emission is a long-standing challenge at the interface of materials chemistry and optical physics.

The emission bandwidth in LEDs is primarily governed by luminescent materials. Commercial inorganic emitters such as InGaN and AlGaInP cover the visible light with full-width at half-maximum (FWHM) values of 15–40 nm depending on wavelength (4,5). Notably, the bandwidth broadens in the blue-to-green region (450–550 nm) and quantum efficiency drops significantly in the green emission range, known as the “green gap” problem (6–8). Quantum dots (QDs) offer tunable emission wavelength through size control, although their FWHM remains around ~20 nm (9–11). Metal halide perovskite emitters also exhibit a similar FWHM range (12,13). Sodium-vapor lamps and rare-earth ions like Eu^{3+} exhibit intrinsically narrow emissions (~10 nm) (14–19), yet their wavelengths are fixed by localized atomic transitions and are difficult to tune, and these transitions have inherently low oscillator strengths (20). In contrast, organic luminophores including metal complexes (21) offer color tunability and high photoluminescence quantum yields (PLQYs) across the full color range, but their emission bands are generally wide (>40 nm) due to strong vibronic coupling. Consequently, achieving pure blue and green fluorescence with extremely small FWHM values remains an outstanding challenge in materials design (22), despite its immediate practical importance for next-generation display technologies.

Multiple-resonance (MR) emitters have recently emerged as a key class of materials capable of suppressing vibronic coupling and exchange energy through alternating electron-donating nitrogen and electron-accepting boron atoms within rigid π -conjugated frameworks (23,24). The opposing mesomeric effects of these atoms localize the frontier molecular orbitals (MOs) on distinct atomic sites, giving rise to short-range charge transfer (CT) and minimal structural relaxation upon excitation. This electronic structure yields high PLQY, narrow emission bands, and a small singlet–triplet (S_1 – T_1) energy gap (ΔE_{ST}), often accompanied by thermally activated delayed fluorescence (TADF) for efficient triplet harvesting in OLEDs. A representative MR-TADF emitter, DABNA, has achieved a FWHM of ~30 nm, and its derivatives, such as ν -DABNA and V-DABNA featuring multiple DABNA subunits (Fig. 1A) (25,26), have achieved even narrower FWHM values of 15–20 nm. However, further narrowing has been constrained by the inherent trade-off between orbital delocalization and vibronic suppression.

Here, we introduce a molecular design strategy that expands the MR framework through a modular repetition architecture, effectively amplifying the MR effect by spatially extending the CT distribution (Fig. 1B). This π -extension preserves the local CT character with short-range CT

distances in each MR unit, allowing the system to inherit the electronic rigidity of the smaller cores, and enhance the overall CT amplitude. Using this approach, we synthesized a series of emitters exhibiting fluorescence spanning from sky-blue to green with exceptionally narrow emission bands. The sky-blue emitter displays an FWHM of 6.9 nm (36 meV), accompanied by an extremely small ΔE_{ST} of ≈ 4 meV and ultrafast TADF ($\tau_{TADF} = 0.46 \mu s$), and the green analogues retain sharp spectra with similarly high PLQY. These results define a regime of spectral precision previously considered unattainable in organic molecules and establish a molecular framework for suppressing vibronic coupling and achieving remarkably high color purity across the visible range.

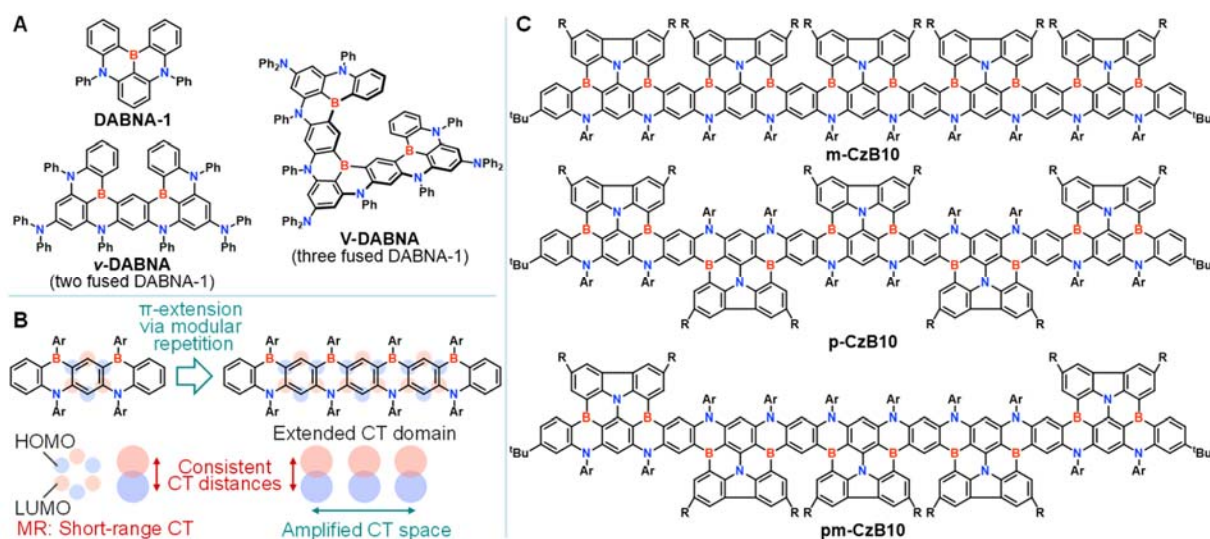


Fig. 1. Chemical structures of π -extended MR compounds and a series of CzB10 derivatives. (A) Molecular structures of DABNA-1, v-DABNA, and V-DABNA. (B) Schematic illustration of the short-range CT framework based on the modular repetition architecture. (C) Chemical structures of m-CzB10-Mes, p-CzB10-Mes, and pm-CzB10-Mes, each incorporating 10 boron atoms, where R = 2-ethylhexyl and Ar = mesityl.

Molecular Design and Synthesis

The molecular structures investigated here (Fig. 1C) are based on the repetition of the fundamental CzB2 unit, which contains two boron atoms and exhibits cyan emission at 490 nm with an FWHM of approximately 30 nm (27). We previously developed a series of CzB n compounds ($n = 4, 6$, and 8), that showed progressive spectrum narrowing with increasing molecular size (FWHM of 14 nm for CzB4, 12 nm for CzB6, and 12 nm for CzB8 in 0.1 wt%-doped polystyrene films), while maintaining similar emission maxima (28). Although the trend in linewidth reduction appears to approach saturation, other properties continue to improve with molecular extension ($\tau_{TADF} = 5.0 \mu s$ for CzB4, 2.9 μs for CzB6, and 1.4 μs for CzB8), indicating progressively stronger MR behavior. To further explore the limit of π -expansion, we synthesized m-CzB10-Mes, which features an extended molecular framework comprising ten boron atoms. In earlier CzB n compounds, the Ar group was a 3,5-dimethylphenyl group. For the larger CzB10

framework, we selected a mesityl (Mes) group as a sterically protecting symmetric substituent to ensure solubility and structural regularity in the expanded molecule.

Experimentally, because CzB4–8 exhibit minimal emission-wavelength shifts, CzB10 was expected to emit near 490 nm in the sky-blue region. Theoretical analysis confirmed the preservation of the highest occupied and the lowest unoccupied molecular orbital (HOMO and LUMO) and excited-state energies, despite the delocalization of MOs over the entire conjugated framework (figs. S1, S2, and table S1). This result contrasts with π -extended systems, which show red-shifted emission by long-range electronic delocalization (29). To expand the emission range beyond blue, we further adopted an alternative connection mode of the CzB2 subunit, incorporating two boron and nitrogen atoms in a para-relationship (30,31). This design led to p-CzB10-Mes and pm-CzB10-Mes, both expected to emit in the green region (figs. S3, S4, and table S2).

The target compounds were synthesized via a one-shot borylation approach (32), introducing ten boron atoms in a single step. Most of the synthetic routes (supplementary materials section 2-1) involve Buchwald-Hartwig coupling, which afforded the precursor amines in good yields. The borylation reaction for m-CzB10-Mes using boron triiodide (BI₃) and 2,6-di-*tert*-butylpyridine (DTBPy) proceeded quantitatively at 150 °C, yielding the product with 100% conversion as determined by ¹H-NMR and a 92% isolated yield. This percentage corresponds to over 99% yield per boron atom introduced with three electrophilic C–B bond formations. Such efficiency surpasses that of the CzB n series, is attributed to the complete suppression of side borylation at the Ar groups by replacing the xylyl group with a mesityl group. At lower temperatures, incompletely borylated byproducts (e.g., CzB8 and CzB9) were detected in increased proportions but were difficult to isolate. In contrast, p-CzB10-Mes was successfully obtained in 93% yield even at 30 °C, reflecting the higher reactivity of the para-derivatives, which can be rationalized by the HOMO energies of the intermediates (fig. S5). Overall, these results demonstrate a remarkably facile and extremely efficient borylation process.

Photophysical Properties

The ultraviolet-visible (UV-vis) and photoluminescence (PL) spectra of dilute toluene solutions (10⁻⁶ M) of CzB10 derivatives are shown in Fig. 2. Although a concentration of 10⁻⁵ M is typically employed, the weight percentage at this concentration corresponds to a standard experimental level given the large molecular weights of the compounds. Indeed, the PL spectra at 10⁻⁵ M exhibit a pronounced shoulder peak, indicating significant aggregation (fig. S6). Therefore, reliable data can be obtained at concentrations lower than 10⁻⁶ M. Among the derivatives, m-CzB10-Mes exhibited highly efficient emission with a PLQY of 99% and an exceptionally narrow FWHM of 7.5 nm (39 meV) (Fig. 2A). A more distinctive feature is its notably small full-width at tenth maximum (FWTM) of 17.5 nm, indicating a significantly weak 0-1 transition, in clear contrast to the 48 nm of FWTM observed for ν -DABNA. At a concentration of 10⁻⁷ M, the PL spectrum became even sharper, with an FWHM of 6.9 nm (36 meV) and a FWTM of 15.9 nm, suggesting that aggregation was not completely suppressed even under highly dilute conditions. Meanwhile, this concentration was not suitable for evaluating PLQY and other parameters because of insufficient absorbance. Aggregation was more pronounced in meta-type structures, indicating that the perpendicular orientation of the Mes group relative to the π -plane helps suppress aggregation, whereas the carbazole moiety can engage in π - π interactions. Although concentration-dependent PL behavior was previously overlooked in the CzB n series, aggregation was also

observed at higher concentrations (fig. S7). Consequently, dilute toluene solutions revealed a clear trend of decreasing FWHM values for m-CzB n derivatives (13.2, 11.0, 8.9, and 7.5 nm for $n = 4, 6, 8$ and 10, respectively; corresponding to 15–20% decreases at each step) as well as Stokes shift values (3.8, 2.3, 1.1, and 0.4 nm for $n = 4, 6, 8$ and 10, respectively). This feature suggests that the MR effect can be enhanced by increasing molecular size while maintaining a short-range CT distance (fig. S8). These results are fully supported by theoretical investigations, which show that the Huang-Rhys factors and reorganization energies for the S_1 – S_0 transitions are markedly reduced from m-CzB4 to m-CzB10, resulting in almost complete suppression of vibronic coupling (Fig. 3). The TADF properties of m-CzB10-Mes were also improved, resulting in a remarkably short delayed lifetime of 464 ns (Fig. 2B), which is much shorter than those of conventional donor–acceptor-type TADF materials (33,34). Phosphorescence spectra were not observed in toluene except for p-CzB10-Mes (fig. S9), likely due to the high fluorescence rate constant (k_F) leading to predominant decay from the S_1 state. Alternatively, phosphorescence spectra were measured in 3-methylpentane (3-MP) (Fig. 2C), where weak phosphorescence signals were observed for all compounds. The ΔE_{ST} of m-CzB10-Mes was calculated to be only 4 meV, consistent with its rapid TADF process. Theoretical calculations also support the experimental results, showing a large spin-orbit coupling (SOC) (fig. S10) (35,36). Notably, the FWHM in 3-MP was only 5.5 nm (30 meV), demonstrating the immense potential of organic luminophores for future applications.

As expected, p-CzB10-Mes and pm-CzB10-Mes exhibited green emission at 561 nm and 543 nm (Fig. 2, D and G), respectively, with the latter being particularly close to the ideal green region. Although their FWHM values (15.8 nm and 17.5 nm, respectively) are slightly broader than that of m-CzB10-Mes, as suggested by theoretical investigations (Fig. 3), the corresponding energy width of 70 meV is among the best values ever reported for green emitters. Notably, green emitters with FWHM <20 nm are typically based on DABNA and ν -DABNA cores, which generally emit below 530 nm (37–40). In contrast, efficient emitters with emission wavelengths >530 nm are mainly based on the fused DABNA structure and tend to exhibit broader spectra (41–44). Despite the relatively small oscillator strength (f) for the S_0 – S_1 transition in pm-CzB10-Mes (table S3 and fig. S11), the PLQY reached 100%, which is attributed to the unexpectedly strong suppression of non-radiative decay (k_{IC}). Indeed, the prompt fluorescence decay is relatively slow (fig. S12), and the peak intensity at the lowest energy absorption band is not particularly strong. However, this characteristic may be advantageous due to reduced self-absorption. These features originate from the nature of the excited states in pm-CzB10-Mes, where S_1 , S_2 , T_1 and T_2 dominantly localized on the para-subunit orbitals, and S_3 and T_3 involve transitions with greater contribution from the meta-subunit. This distribution clearly reflects the para-meta fusion effect in the modular repetition structure.

The central benzene ring of the para-subunit contains two nitrogen atoms and two boron atoms in a para-relationships. This configuration raises the HOMO energy by forming antibonding orbitals between the benzene ring and the nitrogen lone pair electrons, and lowers the LUMO energy through bonding interactions between the benzene ring and the vacant p-orbitals of boron. As a result, the HOMO-LUMO gap is reduced, leading to green emission. This tendency is also observed in the other isomeric structures, although they are expected to exhibit inferior spectral characteristics (figs. S13 and S14). Despite this energy-level control, the HOMO-LUMO separation via the MR effect is also diminished, resulting in a long delayed emission lifetime of 115 μ s for p-CzB10-Mes and no observable delayed emission for pm-CzB10-Mes (Fig. 2, E and H). The ΔE_{ST} values are relatively large (150 meV and 200 meV, respectively), though still smaller than expected (Fig. 2, F and I) (45). The absence of TADF behavior in pm-CzB10-Mes is inevitable due to complete decay from the S_1 state, possibly caused by a large S_1 – T_n gap with weak

SOC, which hinders intersystem crossing (ISC) and triplet formation (figs. S15 and S16). Although TADF is absent, pm-CzB10-Mes can still be considered an outstanding fluorescent material. Overall, as summarized in Fig. 2J and table S4, the CzB10 series has achieved several significant milestones, including a dramatic advancement that yields the narrowest emission spectra to date and the development of highly efficient green emitters.

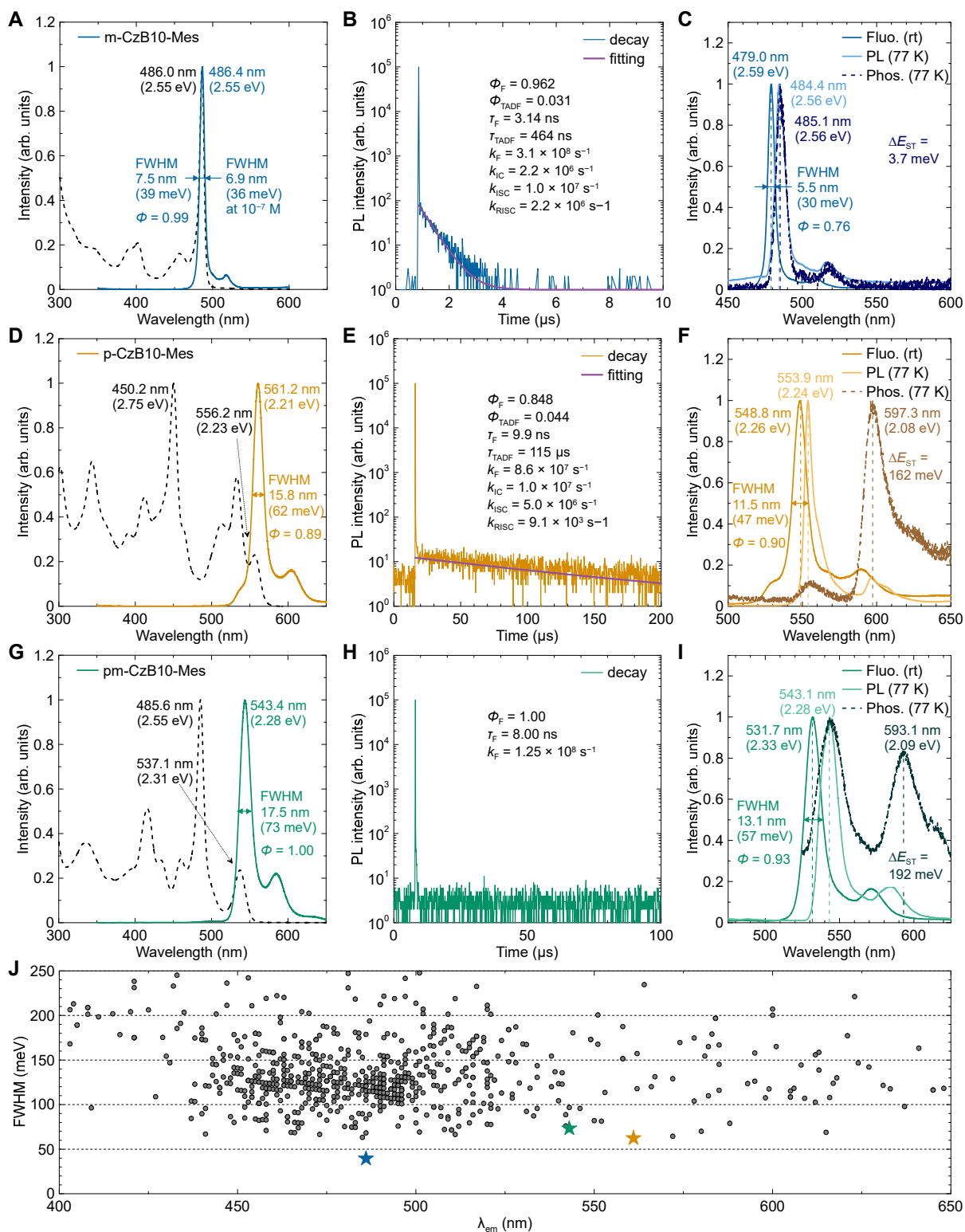


Fig. 2. Photophysical properties of CzB10 derivatives. (A, D, and G) Normalized absorption and fluorescence spectra in toluene at the concentration of 10^{-6} M for (A) m-CzB10-Mes, (D) p-CzB10-Mes, and (G) pm-CzB10-Mes with absorption/emission maxima (nm, eV) and FWHM

(nm, meV). (B, E, and H) Transient decay spectrum at rt and its relevant parameters for (B) m-CzB10-Mes, (E) p-CzB10-Mes, and (H) pm-CzB10-Mes. Φ_F and Φ_{TADF} indicate quantum yields of the fluorescent and TADF components, respectively; τ_F and τ_{TADF} indicate the lifetimes of the fluorescent and TADF components, respectively. (C, F, and I) Steady-state PL (fluorescence) spectra at rt and 77 K without delay time, and phosphorescence spectra at 77 K with a delay time of 25 ms in 3-methylpentane at the concentration of 10^{-6} M for (C) m-CzB10-Mes, (F) p-CzB10-Mes, and (I) pm-CzB10-Mes with emission maxima (nm, eV) and FWHM (nm, meV). (J) Comparison of FWHM values (meV), estimated from wavelength (nm) (fig. S17) values under the assumption of symmetric spectra, measured in toluene for approximately 750 reported compounds based on boron–nitrogen-type MR systems.

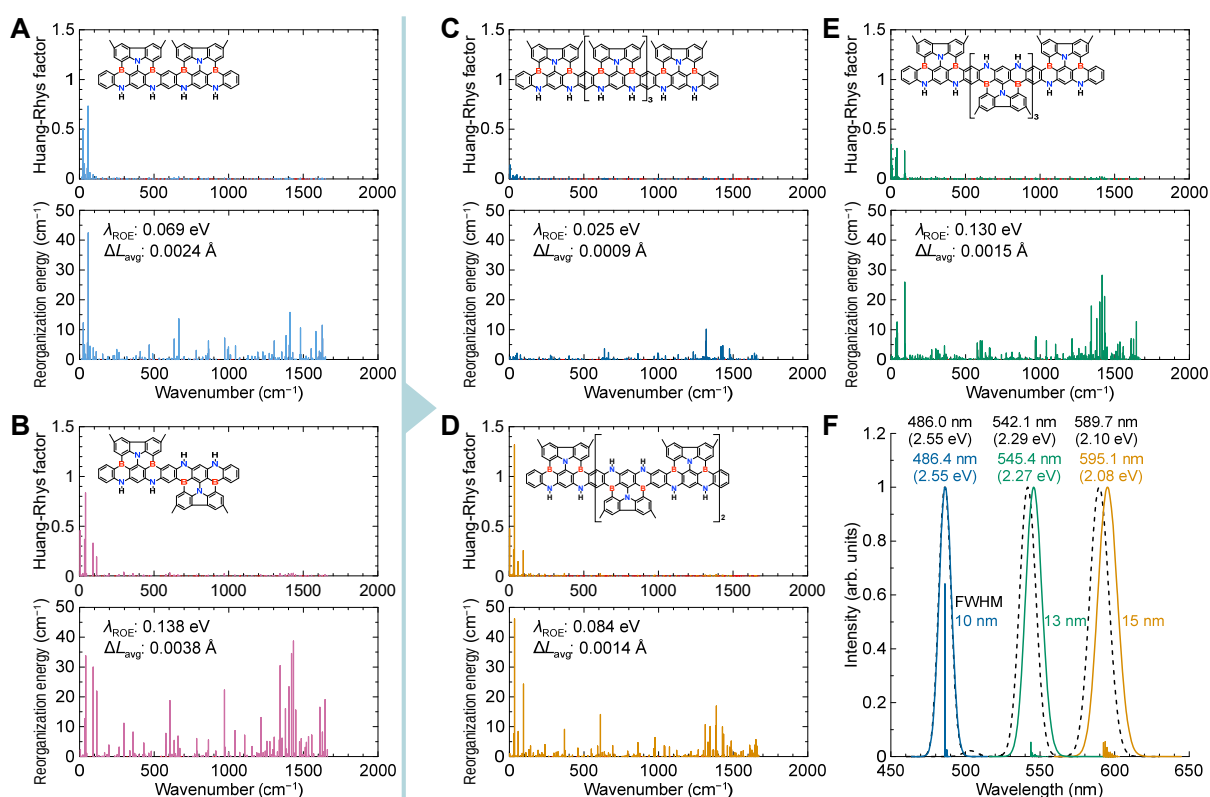


Fig. 3. Theoretical study of the excited states. (A, B, C, D, and E) Huang-Rhys factors and reorganization energies of each vibrational mode contributing to the S_1 – S_0 transition for the model compounds of (A) m-CzB4, (B) p-CzB4, (C) m-CzB10, (D) p-CzB10, and (E) pm-CzB10 calculated with MOMAP software. The λ_{ROE} (reorganization energy) and ΔL_{avg} (the structural deviation) between S_0 and S_1 structures were calculated with Gaussian software. (F) Simulated absorption (dashed lines) and emission (solid lines) spectra by Franck–Condon analysis for the model compounds of m-CzB10 (left), p-CzB10 (right), and pm-CzB10 (center), where the FWHM was set to 400 cm^{-1} .

OLED Characteristics

Finally, we explored the potential of using m-CzB10-Mes and pm-CzB10-Mes in solution-processed OLEDs (SOLEDs). Solution processing is an actively explored fabrication route for OLEDs and remains attractive for low-cost and large-area device manufacturing from a technological perspective. Because of their large molecular architectures, the present emitters are inherently more compatible with solution processing than with vacuum-deposited OLED fabrication. However, practical implementation of SOLED displays remains challenging, and their efficiencies are generally lower than those of OLEDs fabricated using thermal evaporation because of the difficulty in using the multilayer structure without dissolving the underlying layer (46). Therefore, SOLEDs still require technological improvement in many aspects. In addition, aggregation issues, which were observed even in dilute solutions of m-CzB10-Mes, are recognized as a general challenge inherent to the fundamental core structures of MR compounds, and thus, various individual approaches have been proposed to address this issue (47,48). In particular, solution processing may exacerbate aggregation, as the ink must be relatively concentrated to form films of sufficient thickness. At present, however, CzB10 derivatives have not yet incorporated specific strategies to suppress aggregation. Therefore, future development of materials integrating such approaches is expected to provide improved performance.

Doped films of m-CzB10-Mes in a polymer host (Fig. 4A) still exhibited narrow emission with an FWHM of 9.1 nm (47 meV) (Fig. 4B). A slight broadening of the PL spectrum is primarily attributed to the higher polarity of the host material. Aggregation likely occurred as well, as PLQY slightly decreased with increasing doping concentration. Upon optimization of the doping concentration, simple OLEDs employing an EML consisting of x-wt% m-CzB10-Mes in a polymer host revealed that the highest external quantum efficiency (EQE, η_{ext}) was obtained at a concentration of 1.0–1.5 wt% (fig. S18). This result can be attributed to exciton loss due to insufficient energy transfer from the host material at low concentrations and aggregation-induced quenching at higher concentrations. Based on these findings, TADF-sensitized fluorescence (TSF, hyperfluorescence) (49,50) devices incorporating a blue TADF sensitizer (4tCzBN-PhCN) (51) as an assistant dopant (AD) for 1 wt% m-CzB10-Mes as the terminal emitter were fabricated. The PL spectra with AD are slightly broadened, and the PLQY values were improved (Fig. 4C). Consequently, a reasonably high EQE of 18.0% was obtained, which is a significant improvement compared to 10.8% for the device without the sensitizer (Fig. 4D–4G). An exceptionally narrow FWHM of approximately 11 nm was also achieved, although the EL spectrum broadened slightly with increasing AD concentration relative to the ~10 nm obtained without sensitizer. These results indicate that achieving highly efficient energy transfer becomes even more crucial for extremely narrowband emitters such as m-CzB10-Mes. The fast k_{RISC} and k_{f} of m-CzB10-Mes help suppress efficiency roll-off at high current densities (52), maintaining a relatively high EQE values of 13.7% and 8.1% at 1000 cd m⁻² for devices with and without sensitizer, respectively. The operational lifetime of the TSF device was assessed at 4 mA cm⁻² with an initial luminance L_0 above 1000 cd m⁻², yielding an LT₅₀ of 28 h. Considering that an optimized vacuum-deposited TSF device using an MR emitter based on the CzB2 structure shows an LT₅₀ of 75 h (53), the present system demonstrates high operational stability. Based on the acceleration factor (Fig. S20), the LT₅₀ value at 100 cd m⁻² is estimated to approximately 1000 h, indicating considerable practical potential. Overall, the EL spectrum achieved by m-CzB10-Mes may be the narrowest reported to date among solution-processed devices (Fig. 4H), demonstrating the high potential of the CzB10 framework in OLEDs. Devices incorporating pm-CzB10-Mes employed phosphor-sensitized fluorescence (PSF) to facilitate efficient triplet exciton utilization (fig. S19) (54). The addition of pm-CzB10-Mes markedly reduced the FWHM of the phosphor emission spectrum. The device with a 1 wt%-

pm-CzB10-Mes-doped emissive layer maintained an EQE above 20% at 1000 cd m⁻², and the FWHM could be reduced by half. This result represents one of the highest performances in green SOLEDs. However, further increases in doping concentration led to a decrease in EQE, likely due to direct charge recombination in pm-CzB10-Mes or Dexter energy transfer to its triplet excited state. Meanwhile, phosphor emission was still observed at a doping concentration of 4 wt%, indicating that energy transfer was not highly efficient.

Thus, CzB10 derivatives share a common future challenge: improving energy transfer efficiency from the host material. Although suppression of aggregation is of primary importance, simple steric protection strategies may hinder energy transfer, necessitating diversified molecular designs such as increasing the dihedral angle of the π -plane (55). Ultimately, because aggregation and energy transfer are subject to extrinsic influences, with media polarity being one of the key external factors, there remains room for improvement depending on the device architecture materials. For example, less polar host materials may further improve the FWHM values and such a system can still be used for optical pumping by vacuum-deposited OLEDs or LEDs (56). Overall, the high potential for narrowband emission of CzB10 derivatives has been convincingly demonstrated in device applications.

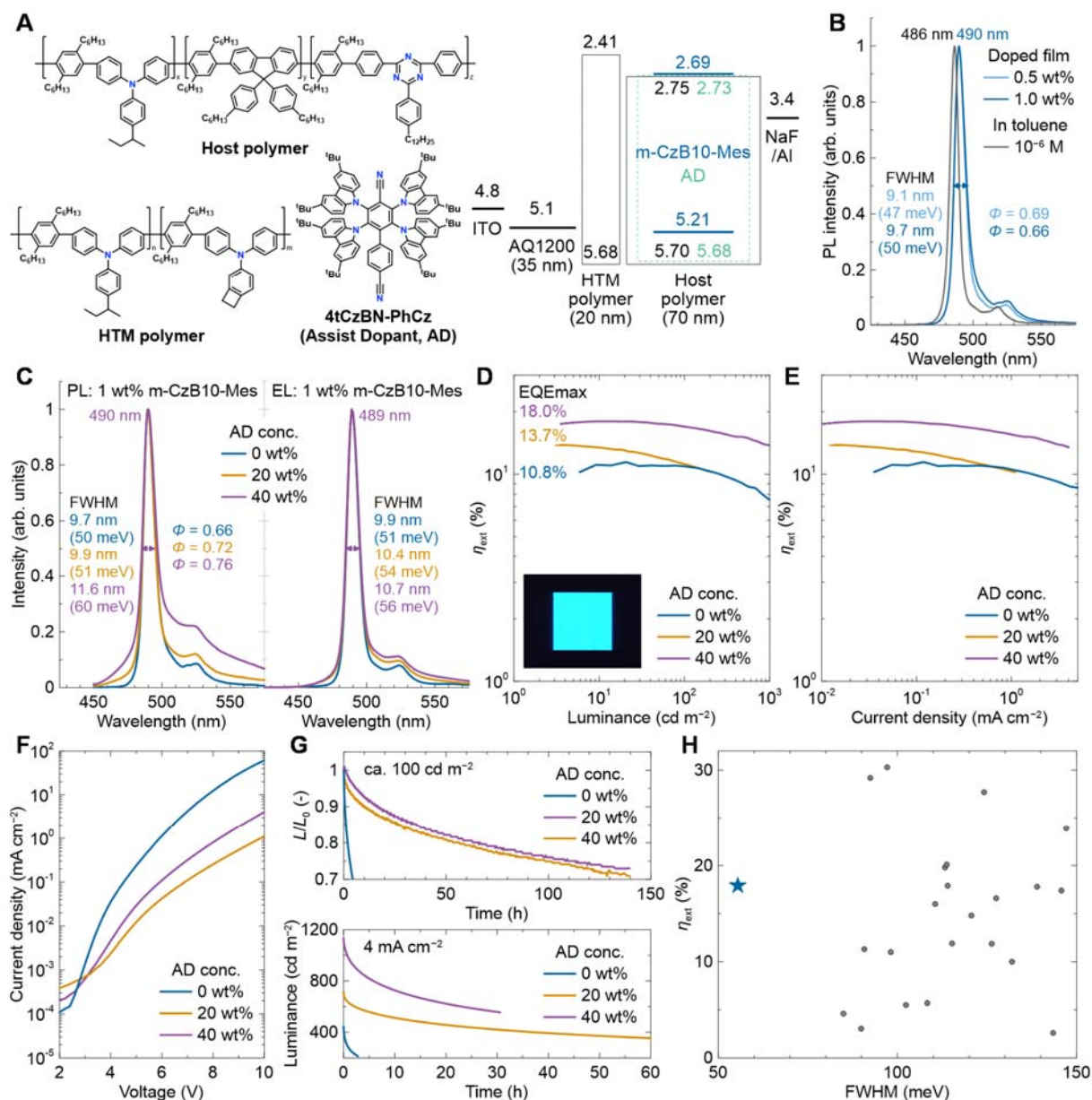


Fig. 4. Electroluminescence properties of TADF-sensitized fluorescence OLEDs using m-CzB10-Mes. (A) Chemical structures for hole-transporting material (HTM), host material (supplementary materials section 2-2), and TADF assistant dopant (AD), device structure and energy levels in eV for each component. (B) Normalized PL spectra of m-CzB10-Mes in the corresponding host excited at 420 nm. (C) Normalized PL and EL spectra for the films composed of 1 wt% m-CzB10-Mes and y wt% AD in the host polymer. (D) EQE-luminance (EQE-L) characteristics. (E) EQE-Current density (EQE-J) characteristics. (F) Current density-voltage (J-V) characteristics. (G) Luminance changes versus operational time at an initial luminance of ca. 100 cd m⁻² and 4 mA cm⁻². (H) Comparison of FWHM versus maximum EQE for reported solution-processed TADF, perovskite, and QD LEDs (57,58).

Conclusions

In summary, we have demonstrated ultranarrow emission bandwidths in organic luminescent materials, overturning the conventional belief that organic luminophores inherently exhibit broad emission spectra. To date, over 1,000 compounds based on the MR framework have been developed; however, to the best of our knowledge, none have achieved an FWHM below 10 nm in standard toluene solution. The performance of m-CzB10-Mes is clearly exceptional (Fig. 2J), where the FWHM is approximately two-thirds that of the previous best value. Furthermore, we identified clear trends in the CzB n series, including a decrease in FWHM and an increase in TADF characteristics, indicating a rational design strategy behind the enhancement of MR effects. This performance was attributed to π -extension through repetition of the fundamental MR core unit, which boosted MR effects by maintaining a short-range CT distance. Our work opens a new avenue for the development of highly efficient MR materials. The unprecedented performance—surpassing that of many systems including inorganic, organometallic, and organic-inorganic hybrid materials—demonstrates the future promise of organic devices, which inherently possess advantages such as low-cost fabrication and superior processability. Although several challenges remain for OLED applications based on this material system, we anticipate that our findings will inspire further advances in organic luminescent materials and contribute to the realization of next-generation photonics and electronics.

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Supplementary Materials

Materials and Methods

Supplementary Text

Figs. S1 to S63

Tables S1 to S17

Data S1 to S2

FIGURE LEGENDS

Fig. 1. Chemical structures of π -extended MR compounds and a series of CzB10 derivatives.

(A) Molecular structures of DABNA-1, ν -DABNA, and V-DABNA. (B) Schematic illustration of the short-range CT framework based on the modular repetition architecture. (C) Chemical structures of m-CzB10-Mes, p-CzB10-Mes, and pm-CzB10-Mes, each incorporating 10 boron atoms, where R = 2-ethylhexyl and Ar = mesityl.

Fig. 2. Photophysical properties of CzB10 derivatives. (A, D, and G) Normalized absorption and fluorescence spectra in toluene at the concentration of 10^{-6} M for (A) m-CzB10-Mes, (D) p-CzB10-Mes, and (G) pm-CzB10-Mes with absorption/emission maxima (nm, eV) and FWHM (nm, meV). (B, E, and H) Transient decay spectrum at rt and its relevant parameters for (B) m-CzB10-Mes, (E) p-CzB10-Mes, and (H) pm-CzB10-Mes. Φ_F and Φ_{TADF} indicate quantum yields of the fluorescent and TADF components, respectively; τ_F and τ_{TADF} indicate the lifetimes of the fluorescent and TADF components, respectively. (C, F, and I) Steady-state PL (fluorescence) spectra at rt and 77 K without delay time, and phosphorescence spectra at 77 K with a delay time of 25 ms in 3-methylpentane at the concentration of 10^{-6} M for (C) m-CzB10-Mes, (F) p-CzB10-Mes, and (I) pm-CzB10-Mes with emission maxima (nm, eV) and FWHM (nm, meV). (J) Comparison of FWHM values (meV), estimated from wavelength (nm) (fig. S17) values under the assumption of symmetric spectra, measured in toluene for approximately 750 reported compounds based on boron–nitrogen-type MR systems.

Fig. 3. Theoretical study of the excited states. (A, B, C, D, and E) Huang-Rhys factors and reorganization energies of each vibrational mode contributing to the S_1 – S_0 transition for the model compounds of (A) m-CzB4, (B) p-CzB4, (C) m-CzB10, (D) p-CzB10, and (E) pm-CzB10 calculated with MOMAP software. The λ_{ROE} (reorganization energy) and ΔL_{avg} (the structural deviation) between S_0 and S_1 structures were calculated with Gaussian software. (F) Simulated absorption (dashed lines) and emission (solid lines) spectra by Franck–Condon analysis for the model compounds of m-CzB10 (left), p-CzB10 (right), and pm-CzB10 (center), where the FWHM was set to 400 cm^{-1} .

Fig. 4. Electroluminescence properties of TADF-sensitized fluorescence OLEDs using m-CzB10-Mes. (A) Chemical structures for hole-transporting material (HTM), host material (supplementary materials section 2-2), and TADF assistant dopant (AD), device structure and energy levels in eV for each component. (B) Normalized PL spectra of m-CzB10-Mes in the corresponding host excited at 420 nm. (C) Normalized PL and EL spectra for the films composed of 1 wt% m-CzB10-Mes and y wt% AD in the host polymer. (D) EQE–luminance

(EQE– L) characteristics. **(E)** EQE–Current density (EQE– J) characteristics. **(F)** Current density–voltage (J – V) characteristics. **(G)** Luminance changes versus operational time at an initial luminance of ca. 100 cd m^{-2} and 4 mA cm^{-2} . **(H)** Comparison of FWHM versus maximum EQE for reported solution-processed TADF, perovskite, and QD LEDs (55,56).

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