



Trifluoromethyl enable high-performance single-emitter white organic light-emitting devices based on quinazoline acceptor

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ABSTRACT

Single-emitter white organic light-emitting diode (WOLED) based on small organic molecule exhibits great potential in simplifying fabrication process of WOLEDs. However, the design and synthesis of molecule for highly efficient single-emitter WOLED still remains a challenge. Herein, two asymmetric donor-acceptor-acceptor' (D-A-A') type molecule (PTZ-PQ-F and PTZ-PQ-CF₃) are developed by employing trifluoromethyl (CF₃) or fluorine atom as secondary acceptor, which can exhibit white lighting with dual emission bands consisting of blue traditional fluorescence from *quasi*-axial (ax) conformer and orange thermally activated delayed fluorescence (TADF) from *quasi*-equatorial (eq) conformer. The introduction of CF₃ into PTZ-PQ-CF₃ greatly enhanced the photoluminescence quantum yield (PLQY) by suppressing the nonradiative deactivation. Owing to electron-inductive-effect of CF₃, the "eq" conformer of PTZ-PQ-CF₃ exhibits a much smaller ΔE_{ST} of 0.01 eV to realize more efficient reverse intersystem crossing (RISC) process, and then enhance the exciton utilization (nearly 100%) of the whole dual emission system. Consequently, single-emitter WOLEDs based on PTZ-PQ-CF₃ show nearly standard white emission with EQE of 13.0% and CIE of (0.35, 0.36) in mCP host and show warm white emission with high EQE of 25.5% and CIE of (0.40, 0.47) in 35 DczPPy host, which are the best performance among reported single-emitter WOLEDs.

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White organic light-emitting diode (WOLED) has received significant development in the past few years owing to its great potential in the next-generation solid-state lighting such as eye protection, power conservation, intrinsic lightweight, and flexibility [1–6]. In generally, white lighting emission is composed of two-color emitters (blue and yellow) or three-color emitters (blue, green, and red), and therefore WOLED is usually fabricated based on multiple emission layers with different color emitters or single emission layer including mixed fluorophores or phosphors emitters [7–11]. Despite the success of WOLEDs based on multi-emitters, the relatively complicated production process of devices leads to unsatisfactory color stability and reproducibility, moreover, increased cost [12–15]. Recently, many reported single small organic molecules can exhibit white emission with dual or more emission bands and then several WOLEDs based on single-emitter were successfully fabricated, which provided potential to

solve the above issue [16–22]. However, the external quantum efficiency (EQE) of them are relatively unsatisfactory resulted from the restricted exciton utilization in these multi emission systems. Among them, small organic molecule based on phenothiazine (PTZ) unit is most commonly employed for single-emitter WOLED due to its dual emission bands caused by conformational isomerism, in which the blue traditional fluorescence comes from *quasi*-axial (ax) conformer and orange thermally activated delayed fluorescence (TADF) comes from *quasi*-equatorial (eq) conformer [19–21]. Since the traditional fluorescence of "ax" conformer can only utilize singlet excitons and its internal quantum efficiency (IQE) is limited to be only 25%, an alternative method to improve efficiency of single-emitter WOLED is to develop highly efficient TADF of "eq" conformer which can also utilize triplet excitons *via* reverse intersystem crossing (RISC) to realize nearly 100% IQE due to small energy gap (ΔE_{ST}) between the lowest singlet excited state (S_1) and the lowest triplet excited state (T_1) [23]. In addition, in such dual emission system, the energy will transfer from host to "ax" conformer and "eq" conformer simultaneously, while part of energy on "ax" conformer will transfer to "eq" conformer and then radiate

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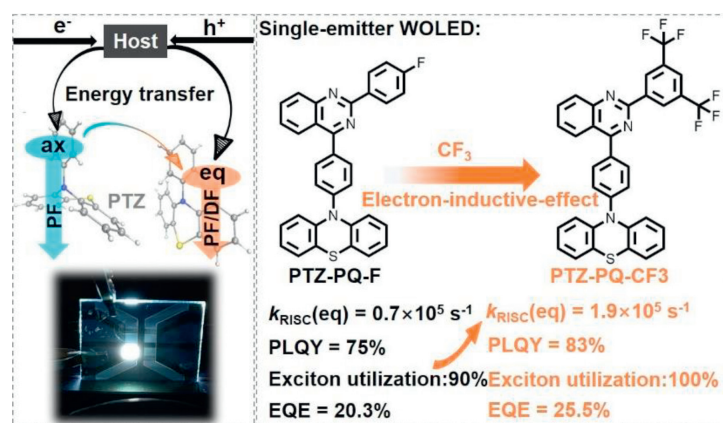


Fig. 1. Single-emitter WOLED and the performances of this work.

by TADF channel. Obviously, “eq” conformer plays a key intermediary role in this process and therefore highly efficient TADF of “eq” conformer is necessary to ensure high exciton utilization in such dual emission system.

Trifluoromethyl (CF₃) is a strong electron withdrawing group resulted from electron-inductive-effect [24]. The large steric hindrance of CF₃ will be able to reduce intermolecular interaction and then result in high photoluminescence quantum yield (PLQY) [25,26]. The F atoms in CF₃ with large electronegativity can form hydrogen bonds between adjacent molecules, and therefore enhance the rigidity and thus suppress the nonradiative deactivation [27]. In addition, owing to inductive effect, the CF₃ can be used to adjust electrochemical property, excited-state property and luminescence property of TADF emitter [28–30]. Recently, many TADF emitters with CF₃ substituents were successfully synthesized and exhibited excellent performance [24–33]. For example, in 2021, Duan *et al.* introduced CF₃ group into TADF emitter as secondary acceptor aiming to optimize molecular electronic and photophysical properties and then improved performance of OLEDs with EQE over 30% [31]. In 2023, they first developed multiresonance (MR) acceptor with CF₃ substituents to modify the distribution of frontier molecular orbitals (FMO) and optimize excited-state property. Finally, the OLED exhibited high EQE over 40% [32]. These excellent performances indicate the effectiveness of the CF₃ substituent strategy in TADF emitters.

Herein, as shown in Fig. 1, CF₃ group was elaborately used to be secondary acceptor connected to quinazoline (PQ), in which PTZ was selected as electron donor to construct asymmetric donor-acceptor-acceptor’ (D-A-A’) structure with dual emission bands, namely PTZ-PQ-CF₃. The molecular PTZ-PQ-F with fluorine (F) atom as secondary acceptor was also synthesized to illustrate the effectiveness of the CF₃ group. The OLEDs exhibit white emission with dual emission bands from “ax” conformer and “eq” conformer at low doping concentration, while single orange emission band from “eq” conformer was obtained at high doping concentration, confirming the energy transfer process from “ax” conformer to “eq” conformer. The C–H...F hydrogen bonds from CF₃ groups in PTZ-PQ-CF₃ greatly enhance the rigidity to suppress the nonradiative deactivation and then provide higher PLQY. Owing to strong electron-inductive-effect, the CF₃ groups in PTZ-PQ-CF₃ successfully modulate the excited state of “eq” conformer and then minimize its ΔE_{ST} as only 0.01 eV which is much smaller than that of PTZ-PQ-F (0.13 eV), leading to larger k_{RISC} ($1.9 \times 10^5 \text{ s}^{-1}$) of PTZ-PQ-CF₃ than $0.7 \times 10^5 \text{ s}^{-1}$ of PTZ-PQ-F. As the station of energy transfer, the improved performance of “eq” conformer will therefore greatly enhance exciton utilization of the whole dual emission system. As expected, the exciton utilization of single-emitter WOLED increased from 90% (PTZ-PQ-F) to 100% (PTZ-PQ-CF₃). Compared

with PTZ-PQ-F, the single-emitter WOLED based on PTZ-PQ-CF₃ displays higher EQE of 25.5% with smaller roll-off.

As shown in Scheme S1 (Supporting information), PTZ-PQ-F and PTZ-PQ-CF₃ were simply synthesized starting from PTZ-pH-Bpin by two-step Suzuki-Miyaura reaction with high yield over 70%. The more detailed information was provided in Supporting information. Their highest occupied molecular orbital (HOMO) energy levels were calculated from the onset of the oxidation potential measured by cyclic voltammetry (CV) as –5.16 eV and –5.15 eV. Meanwhile, the lowest unoccupied molecular orbital (LUMO) were obtained as –2.12 eV and –2.17 eV from the optical bandgaps. The LUMO of PTZ-PQ-CF₃ is slightly deeper than PTZ-PQ-F because of strong electron-inductive-effect of CF₃.

The single crystal of PTZ-PQ-CF₃ was obtained to explore the influence of CF₃ group on stacking mode (Fig. 2). It is noted that only “eq” conformation is observed in single crystal which is different with our previous observation, presumably resulted from the influence of secondary acceptor [21]. The large dihedral angle of 89.31° between phenyl bridge and PTZ will promote separation of the frontier orbital, leading to a small ΔE_{ST} . The large steric hindrance of CF₃ groups greatly expand the distance between two acceptors of adjacent molecules and then weaken the accumulation of triplet excitons [25,26]. Moreover, the multiple intramolecular and intermolecular C–H...F hydrogen bonds from CF₃ groups will greatly enhance the rigidity and thus suppress the nonradiative deactivation by restricting the molecular vibrations. Unfortunately, we failed to obtain the single crystal of PTZ-PQ-F after many attempts with different method. We estimate the PTZ-PQ-CF₃ has more C–H...F hydrogen bonds than that of PTZ-PQ-F due to more

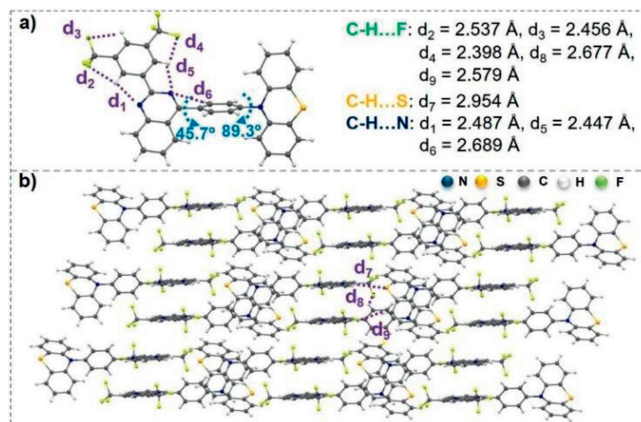


Fig. 2. (a) Single-crystal structure and (b) packing mode of PTZ-PQ-CF₃.

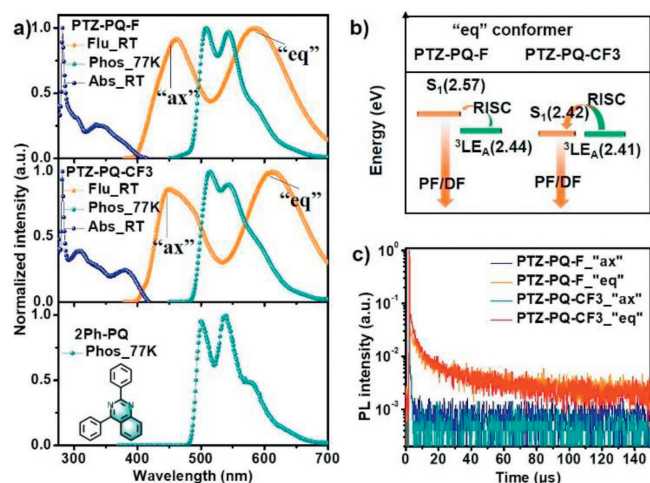


Fig. 3. (a) UV-vis absorption, fluorescence spectra (measured at 300 K) and phosphorescence spectra (measured at 77 K) in toluene (10^{-5} mol/L). (b) Energy levels diagram of "eq" conformers. (c) Transient photoluminescence decay curves in doping film of 35DCzPPy.

fluorine atoms from CF₃, therefore a higher PLQY in PTZ-PQ-CF₃ expectedly.

As shown in Fig. 3a, the ultraviolet-visible (UV-vis) absorption, photoluminescence (PL), and phosphorescence (pH) spectra were measured in toluene solution to study the photophysical properties. For UV-vis spectra, the broad absorption peaks (λ_{abs}) at around 350–450 nm belong to intramolecular charge transfer from PTZ to PQ. Meanwhile, the PL spectra exhibits obvious two different emission bands. According to previous works, the "ax" conformer exhibits a higher energy emission without TADF character and the "eq" conformer exhibits a lower energy emission with TADF character [19–21]. Therefore, the blue emission bands 463 nm of PTZ-PQ-F and 481 nm of PTZ-PQ-CF₃ belong to traditional fluorescence "ax" conformers and orange emission bands 588 nm of PTZ-PQ-F and 617 nm of PTZ-PQ-CF₃ belong to TADF "eq" conformers [21]. The PTZ-PQ-CF₃ shows obvious red-shift emission compared to PTZ-PQ-F due to stronger secondary acceptor of CF₃ group. For pH spectra, PTZ-PQ-F, PTZ-PQ-CF₃, and 2Ph-PQ show similar characteristic vibrational structure, indicating the lowest triplet states of them are local excited states from PQ acceptor (³LE_A), which will be able to enhance the spin orbital coupling (SOC) between the singlet and triplet states [34–37]. From the corresponding onsets of PL and pH spectra, the S₁ and T₁ of "ax" conformers are estimated to be 2.98 eV/2.44 eV for PTZ-PQ-F and 2.90 eV/2.41 eV for PTZ-PQ-CF₃ and then the ΔE_{ST} of them are calculated to be 0.54 eV and 0.49 eV, respectively, which are too large for RISC process so that the "ax" conformers may belong to traditional fluorescence. In terms of "eq" conformers (Fig. 3b), due to the strong electron-inductive-effect of CF₃ group, the S₁ of PTZ-PQ-CF₃ (2.42 eV) is much lower than 2.57 eV of PTZ-PQ-F, leading to much smaller ΔE_{ST} of PTZ-PQ-CF₃ (0.01 eV) than 0.13 eV of PTZ-PQ-F and then the faster RISC process of PTZ-PQ-CF₃ will be expected.

The photophysical properties in doped films were also measured. As shown in Fig. S3 (Supporting information), at 1 wt% doping concentration, both PTZ-PQ-F and PTZ-PQ-CF₃ exhibit white emission with dual emission bands from "ax" conformer and "eq" conformer. The PLQYs of them are measured to be 75% and 83%, respectively. When the doping concentration is 15 wt%, they exhibit only one emission band from "eq" conformer and then the PLQYs are 74% and 79%. The higher PLQY of PTZ-PQ-CF₃ confirms the effectiveness of C–H...F hydrogen bonds from CF₃ groups. As shown in Fig. 3c, the transient PL spectra at high energy emission

peaks corresponding to "ax" conformers exhibit only prompt fluorescence component of 21 ns for PTZ-PQ-F and 18 ns for PTZ-PQ-CF₃, confirming the traditional fluorescence character. Meanwhile, the transient PL spectra at low energy emission peaks corresponding to "eq" conformers exhibit obviously second-order exponential decay, in which the prompt fluorescence lifetimes are 2.1 ns for PTZ-PQ-F and 2.8 ns for PTZ-PQ-CF₃ and the delayed fluorescence lifetimes are 54.5 μs for PTZ-PQ-F and 40.3 μs for PTZ-PQ-CF₃, confirming the TADF character. Therefore, the k_{RISC} of "eq" conformers are calculated to be $0.7 \times 10^5 \text{ s}^{-1}$ and $1.9 \times 10^5 \text{ s}^{-1}$ for PTZ-PQ-F and PTZ-PQ-CF₃, respectively, in which the k_{RISC} of PTZ-PQ-CF₃ is much larger than PTZ-PQ-F contributed to its smaller ΔE_{ST} and enhanced SOC. In addition, the k_{RISC} of non-fluorinated substituted 2PQ-PTZ is obvious slower, indicating the effectiveness of the secondary acceptor in enhancing RISC (Fig. S2 in Supporting information). As expected, the enhanced k_{RISC} of "eq" conformer in PTZ-PQ-CF₃ will be able to increase exciton utilization of the whole dual emission system and therefore a higher EQE in single-emitter WOLED.

To investigate the performances of PTZ-PQ-F and PTZ-PQ-CF₃ in OLEDs, multilayer devices were fabricated with a simple architecture of ITO/TAPC (50 nm)/TCTA (5 nm)/x wt% emitter in host (30 nm)/TmPyPB (30 nm)/LiQ (1 nm)/Al (100 nm) (Figs. 4a and b). Among them, 1,1-bis[(di-4-tolylamino)phenyl]cyclohexane (TAPC) was used as hole-transporting layer (HTL) while 3,3'-[5'-(3-pyridinyl)phenyl] [1,1':3',1',-terphenyl]-3,3',-diyl]bipyridine (TmPyPB) was used as electron-transporting layer (HTL). Tris(4-carbazolyl-9-ylphenyl)amine (TCTA) worked as electron-blocking layer. 1,3-Di(9H-carbazol-9-yl)benzene (mCP) or 3,5-bis(3-(9H-carbazol-9-yl)phenyl)pyridine (35DCzPPy) are separately used as the host, in which the bipolar host 35DCzPPy has more balanced charge transporting character and displays lower triplet energy with T₁ of 2.71 eV than T₁ of 2.91 eV in mCP. The performances of OLEDs are shown in Table 1.

As shown in Fig. 4c, both PTZ-PQ-F and PTZ-PQ-CF₃ exhibit white emission with two obvious emission bands, confirming that the "ax" and "eq" conformers can also coexist and emit individ-

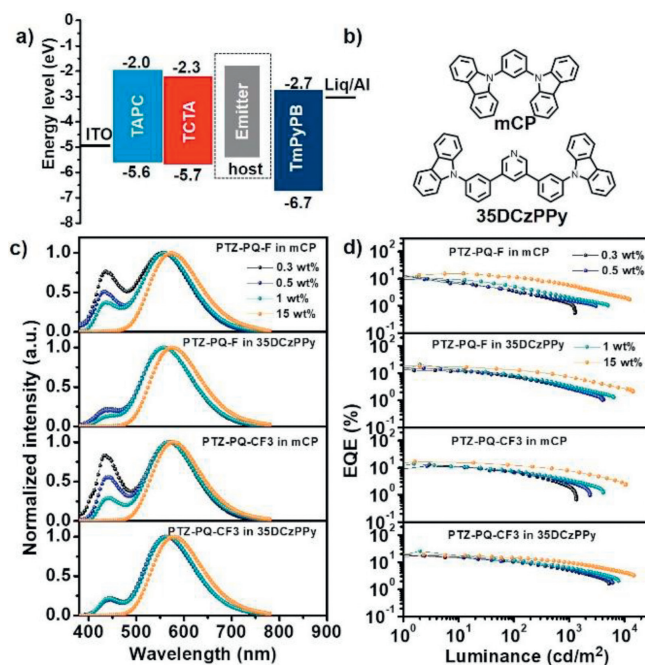


Fig. 4. (a) Energy level diagrams of the materials used in OLEDs. (b) Molecular structure. (c) Electroluminescence spectra at 1000 cd/m². (d) EQE-luminance curves of OLEDs.

Table 1
Summary of OLED device performance of PTZ-PQ-F and PTZ-PQ-CF3.

Emitter:host	x (wt%)	λ_{EL} (nm) ^a	PE (lm/W) ^b	EQE (%) ^c	Roll-off (%) ^d	Luminance (cd/m ²) ^e	CIE ^f
PTZ-PQ-F:mCP	0.3	436, 560	20.0	9.3	88	1280	(0.32,0.37)
	15	572	33.4	15.4	62	20740	(0.46,0.51)
PTZ-PQ-F:35DCzPPy	1	450, 560	51.8	20.3	83	6402	(0.39,0.49)
	15	576	43.3	18.1	61	14313	(0.47,0.50)
PTZ-PQ-CF3:mCP	0.3	436, 568	25.4	13.0	85	1357	(0.35,0.36)
	15	580	31.0	16.8	52	10557	(0.47,0.50)
PTZ-PQ-CF3:35DCzPPy	1	444, 564	62.1	25.5	75	7817	(0.40,0.47)
	15	580	34.7	19.2	49	14860	(0.47,0.50)

^a Electroluminescence peak wavelength at 1000 cd/m².^b Maximum power efficiency.^c Maximum external quantum efficiency.^d EQE roll-off from the maximum value compared to that at 1000 cd/m².^e Maximum luminance.^f CIE at 1000 cd/m².

ual emission from each other under electroexcitation in OLED. The long wavelength emission peaks show red-shift with the doping concentration increased which is attributed to polarization phenomena, confirming the strong ICT character of “eq” conformer. Since the limited exciton utilization of “ax” conformer, part of energy on “ax” conformer will transfer to “eq” conformer and then radiate by TADF channel, leading to increased proportion of orange emission band from “eq” conformer in EL spectra. This process will be enhanced with the increased doping concentration and therefore pure orange OLEDs are obtained at high doping concentration of 15 wt%. As the station of energy transfer, “eq” conformer plays a key intermediary role to ensure high exciton utilization in this process. Consequently, owing to higher k_{RISC} of “eq” conformer in PTZ-PQ-CF3, both single-emitter WOLEDs and pure orange OLEDs based on PTZ-PQ-CF3 exhibit higher EQE and smaller roll-off than that of PTZ-PQ-F. As shown in Fig. 4d and Fig. S6 (Supporting information), for mCP host, at 0.3 wt% doping concentration, the single-emitter WOLED based on PTZ-PQ-CF3 is turned on at 3.4 eV and exhibit higher current efficiency (CE), power efficiency (PE), and EQE of 30.0 cd/A, 25.4 lm/W, and 13.0% with nearly standard CIE of (0.35, 0.36). For 35DCzPPy host, due to the balanced bipolar carrier transport capability and more efficient Dexter energy transfer (DET) process from host to “eq” conformers, the single-emitter WOLED exhibit higher EQE and more warm white emission than that in mCP host. At 1 wt% doping concentration, the single-emitter WOLED based on PTZ-PQ-CF3 shows much higher CE, PE, and EQE of 69.2 cd/A, 62.1 lm/W, and 25.5% than that of PTZ-PQ-F (CE, PE, and EQE of 57.7 cd/A, 51.8 lm/W, and 20.3%, respectively). According to Eq. S13 (Supporting information) [38,39], the exciton utilization of single-emitter WOLED based on PTZ-PQ-F is 90% while the PTZ-PQ-CF3 is up to nearly 100%. As a result, the more excellent performance and higher exciton utilization of single-emitter WLEDs based on PTZ-PQ-CF3 confirm the effectiveness of the secondary acceptor strategy based on CF₃ group.

In conclusion, two novel asymmetric emitters PTZ-PQ-CF and PTZ-PQ-CF3 were successfully designed and synthesized by employing secondary acceptor. They can emit white lighting with dual emission bands from “ax” conformer and “eq” conformer, and then the experimental results prove that “ax” conformer belongs to traditional fluorescence and “eq” conformer belongs to TADF. The CF₃ groups in PTZ-PQ-CF3 provide steric hindrance and C–H...F hydrogen bonds to suppress the nonradiative deactivation and then improve PLQY. Owing to inductive effect, the CF₃ groups decrease S₁ energy of “eq” conformer in PTZ-PQ-CF3 and then successfully minimize its ΔE_{ST} , which therefore greatly enhance k_{RISC} . As a result, the exciton utilization of the whole dual emission system increases to nearly 100% in PTZ-PQ-CF3. Furthermore, highly efficient single-emitter WOLED with CIE of (0.35,0.36) and EQE of 13.0% and warm single-emitter WOLED with CIE of (0.40,0.47) with EQE of

25.5% were obtained. These excellent performances confirm the effectiveness of our molecular design strategy for the single-emitter WOLED, which will provide a promising method for the development of high-performance WOLED.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.ccllet.2023.109118.

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