Rational design of perfectly oriented thermally activated delayed fluorescence emitter for efficient red electroluminescence

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ABSTRACT How to control the dipole orientation of organic emitters is a challenge in the field of organic light-emitting diodes (OLEDs). Herein, a linear thermally activated delayed fluorescence (TADF) molecule, PhNAI-PMSBA, bearing a 1,8-naphthalimide-acridine framework was designed by a double-site long-axis extension strategy to actively control the dipole orientation. The horizontal ratio of emitting dipole orientation of PhNAI-PMSBA reaches 95%, substantially higher than that of isotropic emitters (67%). This unique feature is associated with the intrinsically horizontal molecular orientation of PhNAI-PMSBA and the good agreement between its transition dipole moment direction and molecular long axis. The PhNAI-PMSBA-based OLED achieves an ultrahigh optical outcoupling efficiency of 43.2% and thus affords one of the highest red electroluminescence with an external quantum efficiency of 22.3% and the Commission International de l'Eclairage 1931 coordinates at around (0.60, 0.40).

Keywords: organic light-emitting diodes, thermally activated delayed fluorescence, dipole orientation, outcoupling efficiency

INTRODUCTION

Controlling and tuning the molecular orientation of organic functional materials is an attractive issue in the field of organic light-emitting diodes (OLEDs), because the molecular orientation greatly affects the optical and electrical properties, such as refractive index, carrier transportation and light extraction [1–4]. Especially for

organic emitters used in OLEDs, the horizontal molecular orientation usually benefits horizontal emitting dipole orientation (EDO). Accordingly, the theoretical limit of optical outcoupling efficiency (Φ_{out}) for OLEDs can be boosted from nearly 30% for isotropic emitters to ~45% for fully horizontal emitters [5–8]. Therefore, the control of molecular orientation has been regarded as a promising method in developing excellent organic emitters towards ultrahigh-efficiency OLEDs. The molecular orientation of organic emitters is generally influenced by various factors, including molecular structure, substrate temperature and host-guest interactions [3,9–11]. Up to now, a number of empirical trends are demonstrated in the emitter design to achieve preferentially horizontal molecular orientation and EDO. Moreover, the underlying mechanism about the effect of molecular structure on dipole orientation is still unclear [12–14]. The control of the dipole orientation of organic emitters is challenging.

In the past decade, thermally activated delayed fluorescence (TADF) emitters are particularly attractive in OLEDs because of their merits in harvesting triplet excitons without using noble metals [15–19]. To further boost device performance of TADF OLEDs, many efforts have been made to control the EDO of TADF emitters [20–22]. In virtue of variable molecular structures, TADF emitters' molecular conformation can be easily altered to linear or planar shape, leading to favorable anisotropic

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orientation [23–26]. To design excellent TADF emitters, the rigid molecular structure to suppress non-radiative processes and the suitable separation of frontier molecular orbital (FMO) distribution to guarantee effective reverse intersystem crossing (RISC) process should also be taken into consideration when modulating molecular shape [27–31]. In this context, some TADF emitters have been developed to exhibit preferentially horizontal dipole orientation [22,32,33]. For example, Komino *et al.* [34] reported a fully horizontal oriented TADF emitter at the optimized deposition temperature and in host matrixes. Despite these efforts, the TADF emitters with a near-perfect horizontal dipole ratio ($\Theta_{//}$) were rare.

Therefore, further work is highly required to construct TADF emitters with high $\Theta_{//}$ and gain a deeper understanding of the underlying mechanism behind the preferentially horizontal dipole orientation. In this study, a linear TADF emitter, 2-(4'-(*tert*-butyl)-[1,1'-biphenyl]-4-yl)-6-(10'-(4'-(*tert*-butyl)-[1,1'-biphenyl]-4-yl)-2,7-dimethyl-10*H*,10'*H*-9,9'-spirobi[acridi]-10-yl)-1*H*-benzo[*de*]isoquinoline-1,3(2*H*)-dione (PhNAI-PMSBA), was constructed using a double-site long-axis extension strategy (Fig. 1). Within this strategy, based on a rigid 1,8-naphthalimide-acridine (NAI-AC) hybrid framework, two *tert*-butylbiphenyl units were selectively introduced into the 1,8-naphthalimide (NAI) and acridine (AC) moieties to elongate the molecular long axis. The methyl groups in AC units not only enhanced the intramolecular charge transfer (ICT) transition between the NAI and AC units, but also afforded the suitable separation of FMO distribution of PhNAI-PMSBA. A reference emitter, 2-(4'-(*tert*-butyl)-[1,1'-biphenyl]-4-yl)-6-(2,7-dimethyl-10*H*-spiro[acridine-9,9'-fluoren]-10-yl)-1*H*-benzo[*de*]isoquinoline-1,3(2*H*)-dione (PhNAI-MFAC), was synthesized by incorporating only one *tert*-butylbiphenyl unit into the NAI moiety for comparison. As expected, both emitters exhibited red emissions with distinct TADF nature in both solution and doped films. Moreover, PhNAI-PMSBA had a linear molecular shape in a 35.7 Å length verified by the single-crystal structure. Inspiringly, PhNAI-PMSBA exhibited a high $\Theta_{//}$ of 95% in the host matrix, which is among the best value for horizontally oriented emitters. The molecular dynamics (MD) simulations and optical studies revealed that the underlying

mechanism of such high $\Theta_{//}$ was related to the intrinsically horizontal molecular orientation of PhNAI-PMSBA in the host matrix and the good consistence between the transition dipole moment (TDM) direction and the molecular long axis of PhNAI-PMSBA. Benefiting from the high $\Theta_{//}$, the resulting OLED based on PhNAI-PMSBA achieved an ultrahigh Φ_{out} of 43.2%, far exceeding the theoretical limit of nearly 30% Φ_{out} for isotropic emitters. Finally, a high external quantum efficiency (η_{ext}) of over 22% was demonstrated for the PhNAI-PMSBA-based OLED, accompanied with a red emission and favorable Commission International de l'Eclairage 1931 (CIE1931) coordinates located at around (0.60, 0.40).

EXPERIMENTAL SECTION

Synthesis

The synthetic routes of the intermediates of 2-bromo-*N*-(4-methoxybenzyl)-4-methyl-*N*-(*p*-tolyl)aniline (PMBDPA) and 6-bromo-2-(4'-(*tert*-butyl)-[1,1'-biphenyl]-4-yl)-1*H*-benzo[*de*]isoquinoline-1,3(2*H*)-dione (BrPhNAI) were provided in supplementary information. All reagents were used as received from commercial sources unless otherwise stated. Standard procedures were used to dry the solvents.

Synthesis of 10-(4-Methoxybenzyl)-2,7-dimethyl-10*H*,10'*H*-9,9'-spirobi[acridine] (A). *n*-BuLi (2.3 mol L⁻¹, 1.74 mL, 4.0 mmol) in hexane was dropwise added to a solution of PMBDPA (1.58 g, 4.0 mmol) in tetrahydrofuran (THF) (15 mL) at -78°C under argon atmosphere. After stirring at -78°C for 2 h, 10-((2-methoxyethoxy)methyl)acridin-9(10*H*)-one (1.25 g, 4.4 mmol) in THF (10 mL) was dropwise added. The mixture was stirred at -78°C for another 2 h, gradually warmed to room temperature, and then acidified with diluted HCl (0.5 mol L⁻¹, 10.0 mL) overnight. After stirring for one day, the reaction mixture was treated with aqueous K₂CO₃, extracted with dichloromethane, washed with brine, and dried over anhydrous Na₂SO₄. After removal of the solvent, the crude product was purified by column chromatography on silica gel with petroleum ether/dichloromethane (2:1 by vol.) as the eluent. Finally, the product was obtained as a white powder, 1.49 g, yield: 75%. Melting point (Mp): 244–247°C. ¹H NMR (400 MHz, CD₂Cl₂) δ (ppm): 7.24 (d, *J* = 8 Hz, 2H), 7.04 (t, *J* = 8 Hz, 2H), 6.93–6.91 (m, 4H), 6.81–6.76 (m, 4H), 6.72–6.66 (m, 6H), 6.38 (br, 0.9H), 5.16 (s, 2H), 3.79 (s, 3H), 2.03 (s, 6H). ¹³C NMR (100 MHz, CD₂Cl₂) δ (ppm): 160.7, 138.1, 137.6, 134.8, 134.5, 134.0, 132.7, 131.4, 131.1,

Figure 1 Molecular structures of PhNAI-MFAC and PhNAI-PMSBA.

129.8, 129.1, 128.9, 122.5, 116.1, 115.3, 114.5, 57.2, 52.2, 48.9, 22.1. High resolution mass spectrum (HRMS, ESI): m/z $[M + H]^+$ calcd. for $C_{35}H_{31}N_2O^+$: 495.2431; found: 495.2427.

Synthesis of 10'-(4'-(*tert*-butyl)-[1,1'-biphenyl]-4-yl)-10-(4-methoxybenzyl)-2,7-dimethyl-10*H*,10'*H*-9,9'-spirobi[acridine] (**B**). 4-Bromo-4'-(*tert*-butyl)-1,1'-biphenyl (864 mg, 3.0 mmol), compound **A** (1.24 g, 2.5 mmol), sodium *tert*-butoxide (288 mg, 3.0 mmol), palladium(II) acetate (20 mg, 0.09 mmol) and tri-*tert*-butylphosphonium tetrafluoroborate (78 mg, 0.27 mmol) were dissolved in dry toluene (12 mL) under argon atmosphere, and then stirred at 110°C for 12 h. After cooling to room temperature, the reaction mixture was extracted with dichloromethane, washed with brine, and dried over anhydrous Na_2SO_4 . After removal of the solvent, the residue was purified by column chromatography on silica gel using petroleum ether/dichloromethane (2:1 by vol.) as the eluent. Finally, the product was obtained as a white powder, 1.18 g, yield: 67%. Mp: 283–285°C. 1H NMR (400 MHz, $CDCl_3$) δ (ppm): 7.91 (d, $J = 8$ Hz, 2H), 7.69 (d, $J = 8$ Hz, 2H), 7.54 (dd, $J_1 = 16$ Hz, $J_2 = 8$ Hz, 4H), 7.29 (d, $J = 8$ Hz, 2H), 7.05 (d, $J = 8$ Hz, 2H), 6.95–6.90 (m, 6H), 6.82 (d, $J = 8$ Hz, 2H), 6.71 (dd, $J_1 = 16$ Hz, $J_2 = 8$ Hz, 4H), 6.38 (d, $J = 8$ Hz, 2H), 5.20 (s, 2H), 3.83 (s, 3H), 2.11 (s, 6H), 1.42 (s, 9H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm): 158.6, 150.8, 141.1, 140.1, 138.7, 137.3, 136.2, 133.1, 132.9, 132.5, 131.7, 131.6, 129.6, 129.4, 129.3, 128.0, 127.2, 126.9, 126.5, 125.9, 120.8, 114.4, 113.9, 112.5, 100.0, 55.4, 50.7, 47.1, 34.7, 31.4, 20.7. HRMS (ESI): m/z $[M + H]^+$ calcd. for $C_{51}H_{47}N_2O^+$: 703.3683; found: 703.3680.

Synthesis of 10'-(4'-(*tert*-butyl)-[1,1'-biphenyl]-4-yl)-2,7-dimethyl-10*H*,10'*H*-9,9'-spirobi[acridine] (PMSBA). Compound **B** (1.05 g, 1.5 mmol), Pd on carbon (10%) and formic acid (0.5 mL) in a mixed solvent of CH_2Cl_2 (6 mL) and CH_3OH (3 mL) were degassed with hydrogen for five times. After stirring at room temperature for 12 h, the reaction mixture was treated with CH_2Cl_2 , and then filtrated through kieselguhr. The combined organic phase was dried over Na_2SO_4 . After removal of the solvent, the residue was purified by column chromatography on silica gel using petroleum ether/dichloromethane (1:1 by vol.) as the eluent to afford a white powder, 716 mg, yield: 82%. Mp: 264–268°C. 1H NMR (400 MHz, $CDCl_3$) δ (ppm): 7.92 (d, $J = 8$ Hz, 2H), 7.69 (d, $J = 8$ Hz, 2H), 7.57–7.51 (m, 4H), 7.04 (d, $J = 8$ Hz, 2H), 6.93–6.85 (m, 6H), 6.73–6.65 (m, 4H), 6.38 (d, $J = 8$ Hz, 2H), 6.12 (br, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ (ppm): 150.9, 141.1, 140.1, 138.7, 137.3, 132.5, 131.6, 129.6, 127.9, 126.9, 126.4, 126.0, 120.7, 113.9, 113.1, 34.7, 31.4, 21.0. HRMS (ESI): m/z $[M$

+ $H]^+$ calcd. for $C_{43}H_{39}N_2^+$: 583.3108; found: 583.3106.

Synthesis of 2-(4'-(*tert*-butyl)-[1,1'-biphenyl]-4-yl)-6-(2,7-dimethyl-10*H*-spiro[acridine-9,9'-fluorene]-10-yl)-1*H*-benzo[*de*]isoquinoline-1,3(2*H*)-dione (PhNAI-MFAC). BrPhNAI (1.06 g, 2.2 mmol), 2,7-dimethyl-10*H*-spiro[acridine-9,9'-fluorene] (MFAC) (718 mg, 2.0 mmol), sodium *tert*-butoxide (211 mg, 2.2 mmol), palladium(II) acetate (14 mg, 0.06 mmol) and tri-*tert*-butylphosphonium tetrafluoroborate (52 mg, 0.18 mmol) were dissolved in dry toluene (10 mL) under argon atmosphere and then stirred at 110°C for 24 h. After cooling to room temperature, the reaction mixture was extracted with dichloromethane, washed with brine, and dried over anhydrous Na_2SO_4 . After removal of the solvent, the residue was purified by column chromatography on silica gel using petroleum ether/dichloromethane (1:2 by vol.) as the eluent. Finally, the product was obtained as a red powder, 1.22 g, yield: 80%. Mp: 396–397°C. 1H NMR (400 MHz, CD_2Cl_2) δ (ppm): 8.92 (d, $J = 8$ Hz, 1H), 8.71 (d, $J = 8$ Hz, 1H), 8.22 (d, $J = 8$ Hz, 1H), 8.02 (d, $J = 8$ Hz, 1H), 7.92 (d, $J = 8$ Hz, 1H), 7.87 (d, $J = 8$ Hz, 1H), 7.83–7.79 (m, 3H), 7.66 (d, $J = 8$ Hz, 2H), 7.55 (d, $J = 8$ Hz, 2H), 7.52–7.48 (m, 2H), 7.46–7.38 (m, 5H), 7.28 (t, $J = 8$ Hz, 1H), 6.65 (d, $J = 8$ Hz, 2H), 6.23 (s, 2H), 6.00 (d, $J = 8$ Hz, 2H), 1.92 (s, 6H), 1.39 (s, 9H). ^{13}C NMR (100 MHz, CD_2Cl_2) δ (ppm): 164.1, 163.8, 157.8, 156.3, 150.9, 144.6, 141.5, 140.4, 138.8, 138.4, 137.3, 134.6, 133.0, 132.1, 131.0, 130.8, 130.4, 130.3, 129.1, 128.5, 128.4, 128.3, 128.3, 128.2, 127.9, 127.8, 127.7, 126.9, 126.1, 125.9, 125.2, 124.2, 124.1, 123.3, 120.2, 120.2, 114.6, 56.7, 34.5, 31.1, 20.0. HRMS (ESI): m/z $[M + H]^+$ calcd. for $C_{55}H_{43}N_2O_2^+$: 763.3319; found: 763.3306.

Synthesis of PhNAI-PMSBA. A similar procedure to the preparation of PhNAI-MFAC was used but employing PMSBA instead of MFAC. A red powder was obtained with the yield of 84%. Mp: 445–446°C. 1H NMR (400 MHz, CD_2Cl_2) δ (ppm): 8.91 (d, $J = 8$ Hz, 1H), 8.72 (d, $J = 8$ Hz, 1H), 8.28 (d, $J = 8$ Hz, 1H), 7.98 (t, $J = 8$ Hz, 3H), 7.83–7.78 (m, 3H), 7.71 (d, $J = 8$ Hz, 2H), 7.67 (d, $J = 8$ Hz, 2H), 7.58–7.54 (m, 6H), 7.45 (d, $J = 8$ Hz, 2H), 7.23 (t, $J = 8$ Hz, 2H), 7.06 (s, 2H), 7.00–6.86 (m, 3H), 6.80 (t, $J = 8$ Hz, 1H), 6.66 (d, $J = 8$ Hz, 2H), 6.47 (q, $J_1 = 16$ Hz, $J_2 = 8$ Hz, 2H), 5.96 (d, $J = 8$ Hz, 2H), 2.10 (s, 6H), 1.41 (s, 18H). ^{13}C NMR (100 MHz, CD_2Cl_2) δ (ppm): 166.0, 165.7, 152.9, 152.8, 146.5, 143.5, 143.2, 141.8, 141.3, 140.0, 139.3, 139.1, 138.1, 136.6, 134.9, 134.8, 134.2, 134.1, 133.6, 133.5, 133.4, 133.0, 132.9, 132.7, 132.5, 132.2, 131.5, 131.1, 130.2, 129.8, 129.8, 128.8, 128.7, 128.7, 128.6, 127.9, 127.8, 126.0, 125.2, 122.7, 122.6, 116.4, 116.3, 115.8, 48.9, 36.5, 36.4, 33.0, 22.2. HRMS (ESI): m/z $[M + H]^+$ calcd.

for $C_{71}H_{60}N_3O_2^+$: 986.4680; found: 986.4689.

RESULTS AND DISCUSSION

DFT calculations

Before synthesis, density functional theory (DFT) and time-dependent DFT calculations were performed on PhNAI-MFAC and PhNAI-PMSBA to predict their geometric structures and electronic characteristics. The optimized ground state structure of PhNAI-PMSBA was highly twisted, coupled with a dihedral angle of 85.0° between the acceptor and donor units. This facilitates good separation between the highest occupied molecular orbit (HOMO) and the lowest unoccupied molecular orbit (LUMO) distributions (Figs S1, S2) and thus results in a small energy gap (ΔE_{ST}) of 0.04 eV between the lowest singlet state (S_1) and the lowest triplet state (T_1), potentially activating the RISC process (Fig. 2a and Table S1). The introduction of methyl groups in the AC unit of PhNAI-PMSBA afforded some HOMO-LUMO overlap, leading to a oscillator strength (f) of 0.0043. This reveals the decent emissive ability of PhNAI-PMSBA. The simulation of electrostatic surface on both molecules revealed that PhNAI-PMSBA had a significantly longer molecular length (38.2 Å) along the x axis than PhNAI-MFAC (28.8 Å). Meanwhile, the aspect ratio of molecular length to width for PhNAI-PMSBA (2.77) was much larger compared with 1.99 for PhNAI-MFAC (Fig. 2b and Table S2) [32]. The elongated molecular long axis of PhNAI-PMSBA may lead to a better horizontal molecular orientation.

Synthesis and characterization

As depicted in Scheme 1, A was firstly prepared by lithium-halogen reaction, nucleophilic substitution reaction, Friedel-Crafts reaction and acid-catalyzed cleavage of the 2-methoxyethoxymethyl group. Afterward, the linear donor unit, PMSBA, was synthesized by Pd-catalyzed C-N coupling reaction, followed by Pd-catalyzed hydrogenation reaction. Finally, the target molecule of PhNAI-PMSBA was obtained by Pd-catalyzed C-N coupling reaction of the PMSBA donor with the *tert*-butylbiphenyl-substituted NAI acceptor. By integrating a control donor moiety, MFAC, the reference molecule, PhNAI-MFAC, was also synthesized for comparison. X-ray crystallographic analysis was performed on single crystals of the compounds PhNAI-MFAC and PhNAI-PMSBA (Fig. 3 and Table S3). Both molecules exhibited highly twisted conformation, accompanied with nearly orthogonal dihedral angles between the donor and acceptor moieties (82.3° for PhNAI-MFAC and 86.7° for PhNAI-PMSBA). PhNAI-PMSBA preferred a linear molecular shape in 35.7 Å length, much longer than the length of 24.9 Å for PhNAI-MFAC. These results are in good agreement with the DFT simulation. In the molecular packing diagram, PhNAI-PMSBA exhibited an orderly anti-parallel arrangement. No π - π stacking was observed in the crystal of PhNAI-PMSBA except some short contacts such as C-H- π and C=O-H. This suggests that the intermolecular interaction of PhNAI-PMSBA is relatively weak. Besides, PhNAI-PMSBA was thermally stable to $> 450^\circ\text{C}$ (Fig. S3), rendering it well suited for a vacuum deposition process. In cyclic voltammetry mea-

Figure 2 (a) Calculated energy levels and (b) calculated electrostatic surface potentials at $\rho = 0.001 \text{ e bohr}^{-3}$ with optimized ground state structures.

Scheme 1 Syntheses of PhNAI-MFAC and PhNAI-PMSBA.

Figure 3 Oak ridge thermal ellipsoid plot (ORTEP) diagrams of (a) PhNAI-MFAC and (b) PhNAI-PMSBA without showing hydrogen atoms. Molecular packing diagram of (c) PhNAI-MFAC and (d) PhNAI-PMSBA in a single crystal.

measurements, PhNAI-PMSBA exhibited the double-electron oxidation process, which could be assigned to the different acridine units in the PMSBA donor (Fig. S4). Moreover, both emitters displayed one-electron reduction

behavior arising from the acceptor unit. The HOMO/LUMO levels were obtained as $-5.31/-3.21$ eV and $-5.34/-3.30$ eV for PhNAI-MFAC and PhNAI-PMSBA, respectively.

Photophysical properties

Ultraviolet-visible (UV-vis) absorption spectra of PhNAI-MFAC and PhNAI-PMSBA in toluene showed local excited (LE) transition peaks below 380 nm and ICT absorption bands around 500 nm (Fig. 4a). Similar to PhNAI-MFAC, PhNAI-PMSBA in toluene solution showed a structureless and red emission peaking at 628 nm. In addition, the optical properties of both emitters in the 9-(3-(9*H*-carbazol-9-yl)phenyl)-9*H*-carbazole-3-carbonitrile (mCPCN) host were also studied (Fig. 4b). Both emitters in the mCPCN host displayed similar ICT-featured emission profiles with main peaks at 603 and 606 nm for PhNAI-MFAC and PhNAI-PMSBA, respectively. Furthermore, PhNAI-PMSBA had similar S_1/T_1 levels with respect to PhNAI-MFAC (Table 1). This result strongly suggests the double-site long-axis extension approach rarely affects the excited states of emitters. Accordingly, both emitters in the mCPCN host possessed small ΔE_{ST} values of < 0.1 eV. As expected, the transient photoluminescence (PL) spectra of both emitters showed second-order exponential decays both in toluene solution and the mCPCN host (Fig. 4c and Figs S5, S6). The intensities of delayed fluorescence component increased as

the temperature rose from 200 to 300 K, demonstrating their distinct TADF nature (Fig. S7). The delayed fluorescence lifetime (τ_d) of PhNAI-PMSBA was fitted as short as 2.7 μ s (Table 1 and Table S4), indicative of its effective RISC process [35,36]. Moreover, PhNAI-PMSBA exhibited PL quantum yield (Φ_{PL}) and radiative rate constant ($k_{r,s}$) comparable to PhNAI-MFAC in the mCPCN host (Φ_{PL} = 55% and 61%, and $k_{r,s}$ = 0.90×10^7 and 1.74×10^7 s⁻¹, respectively).

Dipole orientation

To explore the influence of molecular length on the molecular orientation and dipole alignment, the $S_1 \rightarrow S_0$ transition dipole moments (TDMs) of both molecules were firstly simulated using the nuclear ensemble approach [37]. The TDM vectors of both molecules mainly aligned to the molecular long axis (x axis) (Fig. 5a, b), accompanied with large x components and small y and z components. In comparison with PhNAI-MFAC, the TDM vectors of PhNAI-PMSBA had larger x component and smaller y and z components. This result strongly indicates that PhNAI-PMSBA exhibited better agreement between the TDM direction and the molecular long axis.

Figure 4 (a) UV-vis absorption spectra and normalized PL spectra of PhNAI-MFAC and PhNAI-PMSBA in toluene solutions (1×10^{-5} mol L⁻¹) at 300 K (2nd harmonic wavelength of excitation has been deleted as break in PL spectra). (b) Normalized fluorescence (300 K) and phosphorescence (77 K) spectra of 1.5 wt% PhNAI-MFAC and PhNAI-PMSBA doped into the mCPCN host (2nd harmonic wavelength of excitation has been deleted as break in PL spectra). (c) Transient PL spectra of 1.5 wt% PhNAI-MFAC and PhNAI-PMSBA doped into the mCPCN host (inset: prompt components for these transient PL spectra).

Table 1 Summary of the optical properties of PhNAI-MFAC and PhNAI-PMSBA

Compound	λ_{abs} (nm) ^a	λ_{em} (nm) ^a	λ_{em} (nm) ^b	S_1/T_1 (eV) ^c	ΔE_{ST} (eV) ^d	τ_p (ns) ^e	τ_d (μ s) ^e	Φ_{PL} (%) ^f	Φ_{PF} (%) ^f	Φ_{DF} (%) ^f	$\Theta_{//}$ (%) ^g
PhNAI-MFAC	280/308/ 335/499	625	603	2.33/2.27	0.06	15.0	2.9	61	26	35	88
PhNAI-PMSBA	282/335/ 499	628	606	2.32/2.27	0.05	15.9	2.7	55	14	41	95

a) Measured in toluene (1×10^{-5} mol L⁻¹) at room temperature; b) measured in the mCPCN host; c) obtained from the onsets of fluorescence spectra and phosphorescence spectra of both emitters in the mCPCN host; d) calculated from S_1 and T_1 ; e) lifetimes for the prompt and delayed fluorescence components of both emitters in the mCPCN host; f) PL quantum yield, quantum yield for the prompt (Φ_{PF}) and delayed (Φ_{DF}) fluorescence of both emitters in the mCPCN host at room temperature; g) horizontal dipole ratios of both emitters in the mCPCN host. Note: doping concentrations were 1.5 wt% for PhNAI-MFAC and PhNAI-PMSBA.

Figure 5 Simulated transition dipole moment (as indicated by arrow) of (a) PhNAI-MFAC and (b) PhNAI-PMSBA with optimized S_1 structure. Ordinary (red lines) and extraordinary (dark lines) refractive indices (n_o , n_e) and extinction coefficients (k_o , k_e) of (c) PhNAI-MFAC and (d) PhNAI-PMSBA in neat films. Angular dependent p -polarized PL intensity of (e) PhNAI-MFAC and (f) PhNAI-PMSBA in the mCPCN host. Molecular stacking diagram and angular probability distributions of (g) PhNAI-MFAC and (h) PhNAI-PMSBA simulated by MD calculations (red molecules represent emitters and others represent mCPCN host).

Simultaneously, variable angle spectroscopic ellipsometry study also revealed that both emitters exhibited larger ordinary (horizontal) refractive indices (n_o) and extinction coefficients (k_o) than extraordinary (vertical) parameters (n_e , k_e) (Fig. 5c, d), indicating that both emitters showed intrinsically anisotropic properties in neat film [4]. Furthermore, PhNAI-PMSBA delivered larger differences between ordinary and extraordinary parameters compared with PhNAI-MFAC. Such results clearly indicate that PhNAI-PMSBA had a stronger tendency to exhibit horizontal alignment. To further determine this point, we measured angular dependent p -polarized PL intensity of both emitters in the mCPCN host (Fig. 5e, f). The $\Theta_{//}$ of PhNAI-PMSBA was extracted as high as 95%. This value is among the best results reported so far for horizontally oriented emitters. The higher $\Theta_{//}$ of PhNAI-PMSBA over PhNAI-MFAC ($\Theta_{//} = 88\%$) testified the validity of the double-site long-axis extension strategy in realizing horizontal dipole orientation. To get deeper

insights, MD simulations were carried out to mimic the vacuum deposition process of both emitters doped into the mCPCN host [38]. PhNAI-MFAC and PhNAI-PMSBA were prone to form no aggregates when doping into the mCPCN host molecules (Fig. 5g, h). Although the angles (θ) between molecular long axis and the normal direction of the substrate surface varied from 30° to 90° for PhNAI-MFAC and PhNAI-PMSBA, θ with the values higher than 55° ($\theta \approx 55^\circ$ corresponding to random orientation) contributed much more possibility. This suggests that both emitter molecules are likely to align along their molecular long axis on a substrate surface. Accordingly, the ensemble average θ of both emitters were simulated as $70.0^\circ \pm 2.6^\circ$ and $77.9^\circ \pm 2.1^\circ$ for PhNAI-MFAC and PhNAI-PMSBA, respectively. The larger ensemble average θ of PhNAI-PMSBA reveals its better horizontal molecular orientation. During the mimicked vacuum deposition, the average drift angles relative to the ultimate states for both emitters were

gradually reduced as the emitters covered by the following molecules, indicating the gradually stabilized molecular orientation (Fig. S8a). The fast reduction of average drift angle at the initial stage suggests both emitters quickly approached the final molecular orientation at the film surface. Consequently, the designed emitters exhibited rapidly decreased fluctuation at the initial stage and then equilibrated in the bulk (Fig. S8b). In comparison with PhNAI-MFAC, PhNAI-PMSBA revealed the smaller fluctuation range of θ during the deposition process. This suggests that PhNAI-PMSBA showed a better intrinsic tendency to keep its molecular orientation horizontal. To sum up, the double-site long-axis extension strategy can actively enhance horizontal molecular orientation and thus induce the preferred dipole alignment of emitters, which may benefit the light extraction of OLEDs.

Electroluminescent properties

To evaluate the effect of orientation of emitters on device performance, multi-layer OLEDs with a typical configuration of indium tin oxide (ITO)/MoO₃ (2 nm)/di-[4-(*N,N*-ditolyl-amino)-phenyl]-cyclohexane (TAPC) (60 nm)/*N,N*-dicarbazolyl-3,5-benzene (mCP) (10 nm)/emissive layer (EML) (20 nm)/tris-[3-(3-pyridyl)mesityl] borane (3TPYMB) (80 nm)/LiF (1 nm)/Al (Fig. 6a, b) were fabricated. PhNAI-MFAC or PhNAI-PMSBA (1.5 wt%) was doped into the mCPCN host as the EML. Similar to the PL in the mCPCN host, both devices ex-

hibited broad and red electroluminescence (EL) with main peaks at 610 and 615 nm for PhNAI-MFAC and PhNAI-PMSBA, respectively (Fig. 6c), corresponding to favorable CIE 1931 coordinates of (0.59, 0.41) and (0.60, 0.40). Moreover, both devices displayed similar current density-voltage-luminance (*J-V-L*) characteristics with the same turn-on voltages of 3.1 V (Fig. 6d). The PhNAI-PMSBA-based device achieved outstanding EL performance with maximum current efficiency (η_c) of 34.3 cd A⁻¹, a maximum power efficiency (η_p) of 35.9 lm W⁻¹ and a maximum η_{ext} of 22.3% (Fig. 6e and Table 2). These values are among the best EL performance for red TADF OLEDs [39–41]. It is also noted the resulting OLEDs suffer from severe efficiency roll-off at high luminance, which can be alleviated by optimizing doping concentration of both emitters in the host matrix (Fig. S9 and Table S5). Although the Φ_{PL} followed the trend in PhNAI-PMSBA < PhNAI-MFAC, the PhNAI-PMSBA-based device still delivered an external quantum efficiency comparable to the control device based on PhNAI-MFAC (η_{ext} = 22.5%). This may be associated with different $\Theta_{//}$ values of both emitters. According to the classical oscillating dipole model and the obtained $\Theta_{//}$ values, the Φ_{out} of the PhNAI-PMSBA-based device was simulated to be 43.2%, superior to the Φ_{out} (40.3%) of the PhNAI-MFAC-based control device. Furthermore, the 43.2% Φ_{out} is close to the theoretical limit of 45.2% for fully horizontal oriented emitters and is significantly higher than 30.6% for isotropic emitters simulated in the

Figure 6 (a) Energy level diagram and (b) molecular structures of materials used in the devices. (c) EL spectra and (d) current density-voltage-luminance characteristics for the devices. (e) External quantum efficiency, power efficiency and current efficiency *versus* luminance curves for the devices.

Table 2 The key EL data of the resulting OLEDs

Emitter	V_{on} (V) ^a	EL_{peak} (nm) ^b	η_c (cd A ⁻¹) ^c	η_p (lm W ⁻¹) ^d	η_{ext} (%) ^e	Φ_{out} (%) ^f	$\Phi_{\text{out}} \times \Phi_{\text{PL}}$ (%) ^g	CIE (x, y) ^h
PhNAI-MFAC	3.1	610	36.4, 24.5, 12.4	38.1, 16.4, 5.0	22.5, 15.1, 7.6	40.3	24.7	0.59, 0.41
PhNAI-PMSBA	3.1	615	34.3, 22.0, 8.7	35.9, 14.4, 3.3	22.3, 14.3, 5.7	43.2	23.8	0.60, 0.40

a) The turn-on voltage recorded at a brightness of 1 cd m⁻²; b) emission peak of EL spectra; maximum value, values at 100 and 1000 cd m⁻² of c) current efficiency, d) power efficiency and e) external quantum efficiency; f) optical out-coupling efficiency; g) the upper limit of external quantum efficiency; h) CIE 1931 coordinates.

same device architecture (Fig. S10). In this sense, the ultrahigh Φ_{out} of 43.2% and the inspiring η_{ext} of over 22% for the PhNAI-PMSBA-based device demonstrate the importance of controlling dipole orientation in upgrading device performance.

CONCLUSIONS

In summary, a red TADF emitter, PhNAI-PMSBA, with a linear molecular shape was developed by a double-site long-axis extension strategy. In comparison with the single-site extended reference molecule, the molecular shape of PhNAI-PMSBA was effectively elongated, resulting in a better horizontal molecular orientation and a higher horizontal dipole ratio of 95%. To the best of our knowledge, the horizontal dipole ratio of 95% is among the best values for horizontally oriented emitters. Molecular dynamics simulations and optical studies demonstrated the high horizontal dipole orientation of PhNAI-PMSBA was attributed to the synergistic regulation in its molecular shape and transition dipole moment direction. Optical simulation delivered an ultrahigh outcoupling efficiency of 43.2% for the PhNAI-PMSBA-based device, far exceeding 30.6% for isotropic emitters. As a result, using PhNAI-PMSBA resulted in an external quantum efficiency of over 22% and a red EL emission with CIE 1931 coordinates at (0.60, 0.40). This work presents an effective approach to control organic emitter's dipole orientation, and further efforts in developing multicolor organic emitters with fully horizontal orientation is ongoing.

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Author contributions Zeng W, Gong S and Yang C conceived the original idea for investigation. Zeng X, Huang YH, Gong S, Wu CC and Yang C wrote the manuscript. Zeng X, Yin X, Xiao X, Zhang Y, and Dong XQ synthesized the compounds. Zeng X and Huang YH measured photophysical, thermal and electrochemical properties of the compounds. Huang YH, Lee WK, Lu CH, and Lee CC fabricated and characterized the devices. Gong S, Wu CC and Yang C supervised this research. Zhong C performed the quantum chemical calculations and molecular dynamics simulation. All authors discussed the progress of research and reviewed the manuscript.

Conflict of interest The authors declare no conflict of interest.

Supplementary information Experimental details and supporting data are available in the online version of the paper.

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具有完美水平取向的红光型热活化延迟荧光材料

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摘要 如何实现发光材料跃迁偶极矩取向的调控一直是有机发光二极管(OLEDs)领域的巨大挑战之一。本文中, 我们基于双位点长轴延伸策略, 设计合成了一种线型的热活化延迟荧光(TADF)分子, PhNAI-PMSBA, 实现了对跃迁偶极矩取向的选择性调控。由于PhNAI-PMSBA具有显著的水平分子取向, 且其分子长轴与跃迁偶极矩取向高度统一, PhNAI-PMSBA在掺杂发光薄膜中获得的跃迁偶极矩水平取向度高达95%, 远高于各向同性发光分子67%的跃迁偶极矩水平取向度。因此, 基于PhNAI-PMSBA的红光OLED的光耦合输出效率可达43.2%, 最大外量子效率达到22.3%, 其CIE1931色坐标位于(0.60, 0.40)。