Supporting Information

Precise manipulation of carrier recombination zone: a universal novel device structure for highly efficient monochrome and white phosphorescent organic light-emitting diodes with extremely small efficiency roll-off

Yanqin Miao\textsuperscript{a,b,\textordfeminine}, Kexiang Wang\textsuperscript{a}, Long Gao\textsuperscript{a}, Bo Zhao\textsuperscript{a}, Hua Wang\textsuperscript{a}, Furong Zhu\textsuperscript{b,\textordfeminine}, Bingshe Xu\textsuperscript{a}, Dongge Ma\textsuperscript{c}

\textsuperscript{a}Key Laboratory of Interface Science and Engineering in Advanced Materials of Ministry of Education, Research Center of Advanced Materials Science and Technology, Taiyuan University of Technology, Taiyuan, 030024, P. R. China

\textsuperscript{b}Department of Physics, Institute of Advanced Materials, and Institute of Research and Continuing Education (Shenzhen), Hong Kong Baptist University, Kowloon Tong, Hong Kong, P. R. China

\textsuperscript{c}Institute of Polymer Optoelectronic Materials and Devices, State Key Laboratory of Luminescent Materials and Devices, South China University of Technology, Guangzhou, 510640, P. R. China

\textsuperscript{\textordfeminine}Corresponding e-mails: miaoyanqin@tyut.edu.cn (Yanqin Miao); frzhu@hkbu.edu.hk (Furong Zhu)
Section S1

ITO glass substrates were scrubbed and sonicated consecutively with detergent water, deionized water, and acetone, dried in drying cabinet, and then exposed to a UV ozone environment for 10 min. After these processes, the cleaned ITO glass substrates were loaded in a vacuum chamber, a base pressure of ≤ 5×10⁻⁴ Pa, for film deposition using thermal evaporation technology. The deposition rate and film thickness were monitored controlled by the calibrated crystal quartz sensors, e.g., the deposition rates of organic materials, MoO₃, LiF, and cathode Al were controlled at about 1 Å/s, 0.3 Å/s, 0.1 Å/s, and 3–6 Å/s, respectively. Organic films for PL measurements were fabricated with the same method as device fabrication. The EL spectra and CIE coordinates of all OLEDs were measured by a computer controlled PR-655 spectra scan spectrometer. The J-V-L characteristics, CE, and PE were recorded by a computer-controlled Keithley 2400 source integrated with a BM-70A luminance meter. The EQE was calculated from the J−V−L curve and spectra data.
**Fig. S1** The chemical structures of all organic functional materials and emitting materials involved in the monochrome and white PHOLEDs.

**Fig. S2** The normalized PL spectra of 40 nm-thick CBP, TCTA, Bepp₂, and TmPyPB films, and the normalized absorption spectra of Flrpic, Ir(ppy)₃, Ir(BT)₂(acac), and Ir(pq)₂(acac) (in tetrahydrofuran solution with a concentration of 10⁻⁵ M).
TCTA host-based probe devices:

**Device D2-1**: ITO/ MoO$_3$(3 nm)/ TAPC(40 nm)/ DCJTB(0.5 nm)/ TCTA (20 nm)/ TmPyPb(50 nm)/ LiF(1 nm)/ Al(100 nm)

**Device D2-2**: ITO/ MoO$_3$(3 nm)/ TAPC(40 nm)/ TCTA(5 nm)/ DCJTB(0.5 nm)/ TCTA (15 nm)/ TmPyPb(50 nm)/ LiF(1 nm)/ Al(100 nm)

**Device D2-3**: ITO/ MoO$_3$(3 nm)/ TAPC(40 nm)/ TCTA(15 nm)/ DCJTB(0.5 nm)/ TCTA (5 nm)/ TmPyPb(50 nm)/ LiF(1 nm)/ Al(100 nm)

**Device D2-4**: ITO/ MoO$_3$(3 nm)/ TAPC(40 nm)/ TCTA(20 nm)/ DCJTB(0.5 nm)/ TmPyPb(50 nm)/ LiF(1 nm)/ Al(100 nm)

**Fig. S3** The normalized EL spectra of the TCTA host-based probe devices D2-1 to D2-4 at different voltages of 4V, 5V, 6V, and 7V.
Bepp$_2$ host-based probe devices:

**Device D3-1**: ITO/ MoO$_3$(3 nm)/ TAPC(40 nm)/ DCJTB(0.5 nm)/ Bepp$_2$(20 nm)/ TmPyPb(50 nm)/ LiF(1 nm)/ Al(100 nm)

**Device D3-2**: ITO/ MoO$_3$(3 nm)/ TAPC(40 nm)/ Bepp$_2$(5 nm)/ DCJTB(0.5 nm)/ Bepp$_2$(15 nm)/ TmPyPb(50 nm)/ LiF(1 nm)/ Al(100 nm)

**Device D3-3**: ITO/ MoO$_3$(3 nm)/ TAPC(40 nm)/ Bepp$_2$(15 nm)/ DCJTB(0.5 nm)/ Bepp$_2$(5 nm)/ TmPyPb(50 nm)/ LiF(1 nm)/ Al(100 nm)

**Device D3-4**: ITO/ MoO$_3$(3 nm)/ TAPC(40 nm)/ Bepp$_2$(20 nm)/ DCJTB(0.5 nm)/ TmPyPb(50 nm)/ LiF(1 nm)/ Al(100 nm)

Fig. S4 The normalized EL spectra of the Bepp$_2$ host-based probe devices D3-1 to D3-4 at different voltages of 4V, 5V, 6V, and 7V.
CBP host-based probe devices:

**Device D1-1:** ITO/ MoO₃(3 nm)/ TAPC(40 nm)/ DCJTB(0.5 nm)/ CBP(20 nm)/ TmPyPb(50 nm)/ LiF(1 nm)/ Al(100 nm)

**Device D1-2:** ITO/ MoO₃(3 nm)/ TAPC(40 nm)/ CBP(5 nm)/ DCJTB(0.5 nm)/ CBP(15 nm)/ TmPyPb(50 nm)/ LiF(1 nm)/ Al(100 nm)

**Device D1-3:** ITO/ MoO₃(3 nm)/ TAPC(40 nm)/ CBP(15 nm)/ DCJTB(0.5 nm)/ CBP(5 nm)/ TmPyPb(50 nm)/ LiF(1 nm)/ Al(100 nm)

**Device D1-4:** ITO/ MoO₃(3 nm)/ TAPC(40 nm)/ CBP(20 nm)/ DCJTB(0.5 nm)/ TmPyPb(50 nm)/ LiF(1 nm)/ Al(100 nm)

**Fig. S5** The normalized EL spectra of the CBP-based probe devices D1-1 to D1-4 at different voltages of 4V, 5V, 6V, and 7V.
Fig. S6 The CIE coordinates of the proposed monochrome and white PHOLEDs, (a) for the proposed green, yellow, red, and blue devices G4, Y4, R4, and B4, and (b) for the proposed white devices W1-W4.
**Ir(ppy)₃-based probe devices:**

**Device G:** ITO/MoO₃(3 nm)/ TAPC(40 nm)/ TCTA: 7wt%Ir(ppy)₃ (5 nm)/ TCTA: Bepp₂(1:1): 7wt%Ir(ppy)₃(10 nm)/ Bepp₂: 7wt%Ir(ppy)₃(5 nm)/ TmPyPb(50 nm)/ LiF(1 nm)/ Al(100 nm)

**Device D₄-5:** ITO/ MoO₃(3 nm)/ TAPC(40 nm)/ DCJTB(0.5 nm)/ TCTA: 7wt%Ir(ppy)₃(5 nm)/ TCTA: Bepp₂(1:1): 7wt%Ir(ppy)₃(10 nm)/ Bepp₂: 7wt% Ir(ppy)₃(5 nm)/ TmPyPB(50 nm)/ LiF(1 nm)/ Al(100 nm)

**Device D₄-6:** ITO/ MoO₃(3 nm)/ TAPC(40 nm)/ TCTA: 7wt% Ir(ppy)₃ (5 nm)/ DCJTB(0.5 nm)/ TCTA: Bepp₂(1:1): 7wt% Ir(ppy)₃(10 nm)/ Bepp₂: 7wt% Ir(ppy)₃(5 nm)/ TmPyPB(50 nm)/ LiF(1 nm)/ Al(100 nm)

**Device D₄-7:** ITO/ MoO₃(3 nm)/ TAPC(40 nm)/ TCTA: 7wt%Ir(ppy)₃(5 nm)/ TCTA: Bepp₂(1:1): 7wt%Ir(ppy)₃(10 nm)/ DCJTB(0.5 nm)/ Bepp₂: 7wt% Ir(ppy)₃(5 nm)/ TmPyPB(50 nm)/ LiF(1 nm)/ Al(100 nm)

**Device D₄-8:** ITO/ MoO₃(3 nm)/ TAPC(40 nm)/ TCTA: 7wt%Ir(ppy)₃(5 nm)/ TCTA: Bepp₂(1:1): 7wt%Ir(ppy)₃(10 nm)/ Bepp₂: 7wt%Ir(ppy)₃(5 nm)/ DCJTB(0.5 nm)/ TmPyPB(50 nm)/ LiF(1 nm)/ Al(100 nm)

**Fig. S7** The device structure diagram of the Ir(ppy)₃-based probe devices involved for the detection of carrier recombination zone in the proposed green device.