Supporting Information

## Tunable Full-Color Electroluminescence from All-Organic Optical Upconversion Devices by Near-Infrared Sensing

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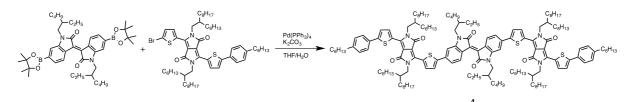
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## Synthesis and Characterization

Commercially available reagents and solvents were used without further purification unless otherwise noted. NMR spectra were recorded on an Avance III 500 spectrometer (Bruker). Chemical shifts of <sup>1</sup>H and <sup>13</sup>C NMR signals were determined relative to the signals of tetramethylsilane ( $\delta = 0.00$ ), CDCl<sub>3</sub> ( $\delta = 77.0$ ) as internal standards. Matrix-assisted laser desorption ionization time-of-flight (MALDI-TOF) mass spectra were collected on an Autoflex III spectrometer (Bruker Daltonics) using dithranol as a matrix. Elemental analysis was carried out with a MT-5 CHN Corder (Yanaco).



Scheme 1. Synthesis of 4.

Synthesis of 4. To a mixture of 6,6'-bis(4,4,5,5-tetramethyl-1,3,2-dioxaborolan-2-yl)-N,N'-(2-ethylhexyl)isoindigo (0.505 g, 0.684 mmol) and 3-(5-bromothiophen-2-yl)-6-[5-(4hexylphenyl)thiophen-2-yl]-2,5-bis(2-hexyldecyl)pyrrolo[3,4-c]pyrrole-1,4-dione (1.55 g, 1.57 mmol) in THF (20 mL) were added Pd(PPh<sub>3</sub>)<sub>4</sub> (0.032 g, 0.027 mmol) and aqueous K<sub>2</sub>CO<sub>3</sub> (2.0 M, 10 mL). The mixture was stirred for 48 h at 60 °C under N<sub>2</sub>. After cooling to room temperature, the reaction mixture was poured into water and then extracted with chloroform. The combined organic layers were washed with water and dried over anhydrous MgSO<sub>4</sub>. After filtration and evaporation, the product was purified by silica gel column chromatography (eluent: chloroform), recrystallized from chloroform/methanol, and dried under vacuum to afford 4 as a dark blue solid (yield = 0.905 g, 57%). This compound was further purified by recycling preparative gel permeation chromatography (eluent: chloroform) prior to use. <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>):  $\delta$  9.17 (d, J = 8.5 Hz, 2 H), 9.02 (d, J = 4.0 Hz, 2 H), 9.01 (d, J = 4.0 Hz, 2 H), 7.55 (d, J = 8.0 Hz, 4 H), 7.47 (d, J = 3.0 Hz, 2 H), 7.39 (d, J = 3.0 Hz, 3 H Hz, 3 H), 7.39 (d, J = 3.0 4.0 Hz, 2 H), 7.24 (d, J = 8.0 Hz, 2 H), 7.18 (d, J = 8.0 Hz, 4 H), 6.96 (s, 2 H), 4.09 (d, J =7.0 Hz, 4 H), 4.01 (d, J = 7.0 Hz, 4 H), 3.82-3.74 (m, 4 H), 2.60 (t, J = 7.5 Hz, 4 H), 2.02-1.95 (m, 4 H), 1.92-1.87 (m, 2 H), 1.63-1.57 (m, 4 H), 1.48-1.22 (m, 124 H), 1.02 (t, J = 7.3 Hz, 6 H), 0.94 (t, J = 7.0 Hz, 6 H), 0.89 (t, J = 6.8 Hz, 6 H), 0.86-0.82 (m, 24 H). <sup>13</sup>C NMR (125 MHz, CDCl<sub>3</sub>): *b* 168.33, 161.50, 161.37, 150.11, 148.44, 145.62, 144.04, 140.03, 138.76, 137.42, 136.91, 135.91, 131.49, 130.67, 130.45, 129.85, 129.01, 128.22, 125.76, 124.87, 123.70, 122.01, 118.94, 108.58, 107.95, 104.42, 46.41, 46.29, 44.08, 38.06, 37.99, 35.75, 31.91, 31.88, 31.70, 31.40, 31.16, 31.03, 30.17, 31.14, 29.86, 29.79, 29.63, 29.60, 29.35, 29.34, 29.03, 28.90, 26.46, 26.40, 26.25, 24.33, 23.12, 22.69, 22.67, 22.64, 22.62. MS (MALDI-TOF): m/z [M]<sup>+</sup> calcd, 2300.55; found, 2301.27. Anal. Calcd (%) for C<sub>148</sub>H<sub>214</sub>N<sub>6</sub>O<sub>6</sub>S<sub>4</sub>: C, 77.23; H, 9.37; N, 3.65; found: C, 77.16; H, 9.30; N, 3.69.

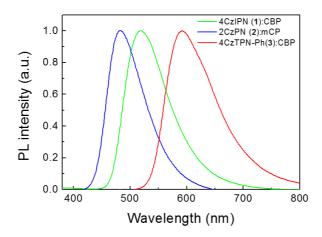


Figure S1. Photoluminescence spectra of 6-wt% doped thin films of 1 in CBP (green), 2 in mCP (blue), and 3 in CBP (red).

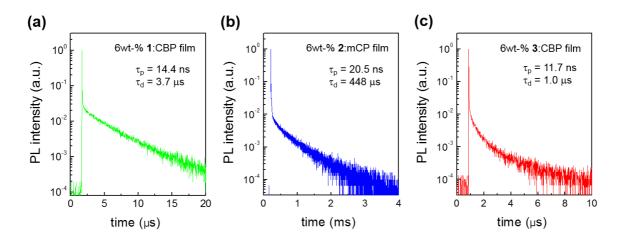


Figure S2. Transient PL decay curves of 6-wt% doped thin films of (a) 1 in CBP, (b) 2 in mCP, and (c) 3 in CBP measured at room temperature under  $N_2$ .

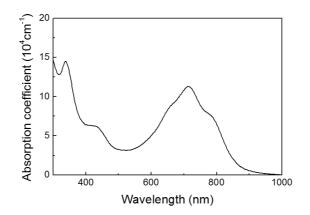


Figure S3. UV-vis-NIR absorption spectrum of a BHJ blend film of 4:PC61BM (1:1, w/w).

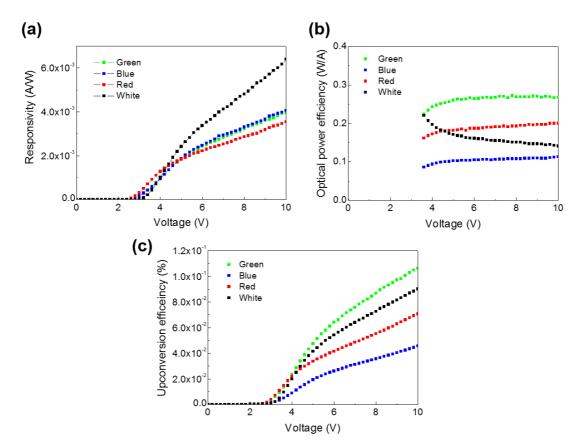
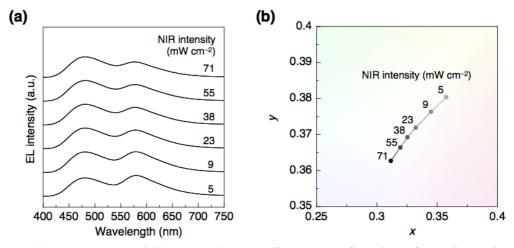


Figure S4. (a) Responsivities, (b) optical power efficiencies, and (c) upconversion efficiencies of organic optical upconversion devices based on TADF emitters 1 (green), 2 (blue), 3 (red), and 2/3 (white).



**Figure S5.** (a) EL spectra and (b) CIE color coordinates as a function of NIR intensity for the white upconversion device.