Supporing Information

Improving Electron Injection of Organic Light-Emitting Transistors via Interface Layer Design

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Figure S1. Schematic illustration for the growth of DPA single crystals by PVT method.



Figure S2. (a, d) The low kinetic energy region of UPS for Au, DPA/Au and DNaDBSO/Au. (b, e) The low binding energy region of UPS for Au, DPA/Au and DNaDBSO/Au. (c, f) Energy level diagrams for Au/DPA system and Au/DNaDBSO/DPA system.



Figure S3. Optical micrographs of DPA-based OLET devices with asymmetric interface layer.



Figure S4. (a) Output characteristics of DPA-based OLET devices symmetrical interface layer for negative $V_{\rm G}$ (from 0 to -120 V). (b) Output characteristics of the DPA-based OLET devices with symmetrical interface layer for positive $V_{\rm G}$ (from 0 to 120 V). Inset: Amplified picture of output curves.



Figure S5. Optical micrographs of DPA-based OLET devices with symmetric interface layer. The red arrow denotes the conductive channel, with a channel length measuring 50 μ m. The thickness of the crystals is less than 1 μ m.



Figure S6. (a, b) Surface morphology and roughness values of the single crystal and injection layer on top of single crystal with AFM.



Figure S7. (a, b) Total device resistance plotted as a function of the channel length for the DPA-transistors with and without injection layers, respectively.



Figure S8. (a b) Output curves and photocurrents of DPA-based OLET devices at negative (a) and positive (b) gate voltage.



Figure S9. (a)Typical transfer curves and EQE of DPA-based OLET devices for P-channel. (b)Typical transfer curves and EQE of dNaAnt-based OLET devices for N-channel.



Figure S10. The luminance and corresponding transfer curve of DPA-based OLET devices.

For planar OLETs, accurately determining the exact luminous area is challenging. Based on literature and emission images¹, we estimated the emission width to be approximately 2 μ m. The DPA light-emitting transistors achieved a maximum luminance of 1438 cd/m² for the n-type transfer curve and 720 cd/m² for the p-type transfer curve.



e S11. Air stability test for dNaAnt-based OLET devices with Au/DNaDBSO and Ca/CsF electrodes. (a, b) Microscopic image of the Ca/CsF electrode: fresh and after 7 hours. (c) P-type and N-type transfer characteristics of devices with Ca/CsF electrode over time. (d, e) Microscopic image of the Au/DNaDBSO electrode: fresh and after 3 days. (f) P-type and N-type transfer characteristics of devices with Au/DNaDBSO electrode over time.

| | Mode 1 | | | | | | | |
|---------|--|---------------------|--------------------------|--|---------------------|------------------------|--|--|
| Devices | μ _h (cm ² V ⁻¹ s ⁻¹) | $V_{\rm t}({ m V})$ | $I_{\rm on}/I_{\rm off}$ | μ _e (cm ² V ⁻¹ s ⁻¹) | $V_{\rm t}({ m V})$ | $I_{ m on}/I_{ m off}$ | | |
| 1 | 0.092 | -105 | 1.4×10 ³ | / | / | / | | |
| 2 | 0.06 | -78 | 8.6×10^{4} | <10-3 | 140 | / | | |
| 3 | 0.35 | -70 | 4.4×10 ³ | / | / | / | | |
| 4 | 0.040 | -35 | 5.8×10 ³ | / | / | / | | |
| 5 | 0.97 | -31 | 1.1×10 ³ | 0.0036 | 92 | 2.5×10^{1} | | |

Table S1. Summary of electrical performance in test mode 1 for DPA-based OLET devices with asymmetric interface layer.

Table S2. Summary of corresponding electrical performance in test mode 2 for DPA-based OLET devices with asymmetric interface layer.

| | Mode 2 | | | | | | | |
|---------|--|---------------------|------------------------|--|---------------------|------------------------|--|--|
| Devices | μ _h (cm ² V ⁻¹ s ⁻¹) | $V_{\rm t}({ m V})$ | $I_{ m on}/I_{ m off}$ | μ _e (cm ² V ⁻¹ s ⁻¹) | $V_{\rm t}({ m V})$ | $I_{ m on}/I_{ m off}$ | | |
| 1 | 0.13 | -66 | 1.6×10^{6} | 0.016 | 60 | 8.5×10 ³ | | |
| 2 | 0.090 | -40 | 2.4×10^{4} | 0.006 | 51 | 1.1×10^{1} | | |
| 3 | 0.27 | -55 | 2.7×10 ⁵ | 0.0024 | 92 | 9.3×10 ¹ | | |
| 4 | 0.31 | -47 | 6.1×10 ⁷ | 0.0038 | 73 | 4.8×10^{1} | | |
| 5 | 0.91 | -30 | 1.1×10 ³ | 0.026 | 85 | 4.9×10 ¹ | | |

Table S3.Summary of transport performance for DPA-based OLET devices with symmetricinterface layer.

| Thickness | μ _h ^{ave} (cm ² V ⁻¹ s ⁻¹) | V _t ^{ave} (V) | I _{on} /I _{off} ave | μ _e ^{ave} (cm ² V ⁻¹ s ⁻¹) | V _t ^{ave} (V) | I _{on} /I _{off} ave | $\mu_{ m h}/\mu_{ m e}$ |
|-----------|---|--------------------------------------|---------------------------------------|---|-----------------------------------|---------------------------------------|-------------------------|
| 0 nm | 0.38 ± 0.51 | -61 | 2.1×10^{6} | 0.0037 ± 0.0061 | 81 | 1.5×10^{3} | 103 |
| 5 nm | 0.48 ± 0.27 | -82 | 1.1×10^{6} | 0.0048 ± 0.0057 | 78 | 1.8×10^{3} | 100 |
| 10 nm | 1.01 ± 0.57 | -54 | 2.2×10^{5} | 0.021 ± 0.016 | 79 | 1.8×10^{3} | 48 |
| 20 nm | 0.73 ± 0.65 | -60 | 1.5×10^{6} | 0.0074 ± 0.0087 | 85 | 1.2×10^{3} | 70 |

It should be noted that the electron mobility in devices which do not show n-type feature is regard as 0.

| Mobility (cm ² V ⁻¹ s ⁻¹) | | | | | | | | |
|---|--------------|--------------|--------------|--------------|--------------|--------------|-----------|--------------|
| dovicos | $\mu_{ m h}$ | $\mu_{ m h}$ | $\mu_{ m h}$ | $\mu_{ m h}$ | $\mu_{ m e}$ | $\mu_{ m e}$ | μ_{e} | $\mu_{ m e}$ |
| uevices | (0 nm) | (5 nm) | (10 nm) | (20 nm) | (0 nm) | (5 nm) | (10 nm) | (20 nm) |
| 1 | 1.02 | 0.88 | 0.54 | 0.57 | 5.30E-04 | 0.01 | 0.02 | 0.0037 |
| 2 | 0.20 | 0.009 | 0.46 | 1.21 | 0.0011 | 0.002 | 0.0060 | 0.013 |
| 3 | 0.15 | 0.0085 | 0.022 | 2.32 | 0.0020 | 9.00E-05 | 0.041 | 0.020 |
| 4 | 1.60 | 0.52 | 0.90 | 1.35 | 0.0016 | 0.0024 | 0.043 | 0.026 |
| 5 | 1.20 | 0.44 | 0.96 | 0.15 | 3.00E-04 | 0.0030 | 0.0060 | 0.0030 |
| 6 | 0.051 | 0.72 | 0.85 | 0.88 | / | 0.0010 | 0.0065 | 0.0040 |
| 7 | 0.074 | 0.61 | 2.17 | 1.27 | 0.016 | 0.0057 | 0.025 | 0.0020 |
| 8 | 0.018 | 0.72 | 1.49 | 0.37 | 2.16E-05 | 0.0026 | 0.011 | 7.00E-04 |
| 9 | 0.28 | 0.61 | 1.45 | 0.046 | 6.43E-05 | 0.0057 | 0.053 | 0.0010 |
| 10 | 0.70 | 0.72 | 1.53 | 0.14 | 2.70E-04 | 0.0026 | 0.020 | 2.00E-04 |
| 11 | 0.0042 | 0.63 | 1.32 | 0.043 | 4.45E-05 | 0.023 | 0.013 | / |
| 12 | 0.082 | 0.20 | 1.10 | 0.24 | 0.0055 | 1.00E-04 | 0.0060 | 0.0060 |
| 13 | 0.032 | 0.69 | 0.093 | 0.30 | 0.017 | 0.0034 | 0.0020 | 1.00E-04 |
| 14 | 0.32 | 0.14 | 0.67 | 1.07 | / | 0.0017 | 0.015 | 0.021 |
| 15 | 0.030 | 0.46 | 1.03 | 1.01 | 0.012 | 0.0080 | 0.043 | 0.010 |

Table S4. Summary of transport performance for DPA-based OLET devices with symmetric interface layer.

Table S5. Summary of the simulated and experimental mobilities of DPA.

| DPA | $\mu_{\rm h} ({\rm cm}^2{\rm V}^{-1}{\rm s}^{-1})$ | $\mu_{\rm e} ({\rm cm}^2{ m V}^{-1}{ m s}^{-1})$ | $\mu_{\rm h}/\mu_{\rm e}$ |
|---------------------------------|--|--|---------------------------|
| Simulation | 2.0 | 0.22 | 9.0 |
| Experiment (with 10 nm DNaDBSO) | 1.1 | 0.053 | 20 |

*The mobility is simulated by transport module in MOMAP software^{2,3}. The calculation was carried out at B3LYP/STO-3G level.

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