Supporting Information

## **Coherent Arrangement of Perylene Diimide Derivative via Adhesion-Controlled Transfer for Noise-Suppressed Light Signal Detection**

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• Functionalized Polymer Delamination Substrate Fabrication for coherent arrangement Assembly NPDI

Fig. S1. Mold fabrication process for the coherent arrangement assembly of NPDI

## NPDI in Methanol

## NPDI in Trifluoroethanol (TFE)



**Fig. S2.** Photographic images of the formation of the coherent arrangement of NPDI layers using methanol or trifluoroethanol (TFE) as the solvent.



Fig. S3. PCE of the NPDI electron transport layer with methanol or trifluoroethanol as the solvent



Fig. S4. (a) UV-visible absorbance spectra and (b) transmittance analysis for PEDOT:PSS/PM6:Y12/NPDI devices with coherent arrangement of NPDI (red line) and conventional NPDI (black line).



**Fig. S5**. Cyclic voltammetry analysis of coherent arrangement of NPDI (red line) and conventional NPDI (black line). (b) Energy level diagram of the device.



**Fig. S6.** (a) Current density–voltage curves under illumination (100 mW/cm<sup>2</sup>) and (b) EQE spectra for PEDOT:PSS/PM6:Y12/NPDI devices with a coherent arrangement of NPDI (red line) and conventional NPDI (black line) (Area: 0.15 cm<sup>2</sup>).



**Fig. S7**. Chart of reported FF and PCE for non-fullerene acceptor based organic photovoltaic devices with 2,2,2- trifluoroethanol.



Fig. S8. (a) Dark current density–voltage curves, (b) responsivity at 0 V, and (c) detectivity at 0 V for an area of 0.15 cm<sup>2</sup> for OPD devices with a coherent arrangement NPDI (red line) and conventional NPDI (black line).



**Fig. S9.** (a) Dark current density–voltage curves, (b) EQE, (c) responsivity at 0 V, and (d) detectivity at 0 V for an area of 0.06 cm<sup>2</sup> for OPD devices with a coherent arrangement NPDI (red line) and conventional NPDI (black line).



**Fig. S10.** FE-SEM cross-sections of PEDOT:PSS/PM6:Y12/NPDI devices with (a) conventional NPDI and (b) coherent arrangement NPDI.



**Fig. S11.** Optical microscopic images of (a) conventional NPDI and (b) coherent arrangement of NPDI films.



Fig. S12. DI water contact angle analysis of BHJ with (a) conventional NPDI and (b) coherent arrangement NPDI films.



Fig. S13. Compensation voltages under illumination of C-NPDI based device and A-NPDI based device.



**Fig. S14**. Photoluminescence analysis of ITO/PEDOT:PSS/PM6:Y12/NPDI devices with coherent arrangement NPDI (red line) and conventional NPDI (black line),



**Fig. S15.** XPS survey spectra of the atomic core levels for the NPDI with methanol and trifluoroethanol; a) F1s, b) N1s.

Table S1. Surface energy parameters for various films.

Matorial		$\gamma_{SV}$	$\gamma_{SV}^{p}$	$\gamma_{SV}^{\ \ d}$
Material		(mJ/m <sup>2</sup> )	(mJ/m <sup>2</sup> )	(mJ/m²)
PUA		55.66	46.12	9.53
BHJ (PM6:)	¥12)	34.11	31.87	2.23
NPDI	Methanol	76.15	40.99	35.16
	Trifluoroethanol	77.30	43.75	33.55

Table S2. Calculated interfacial surface energy for NPDI, BHJ, and PUA.

	Matarial	Interfacial energy
PM6:Y12	wrateriai	[mJ m <sup>-2</sup> ]
	PUA	3.87
NPDI with methanol	PUA	8.23
	PM6:Y12	20.26
NPDI with trifluoroethanol	PUA	7.35
	PM6:Y12	19.43

Table S3. Hansen solubilit	y parameters	for the solvents.
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		<b>∂</b> /M]	Pa <sup>1/2</sup>	
Solvent	$\partial_t$	$\partial_{\mathrm{d}}$	$\partial_{\mathrm{p}}$	$\partial_{ m h}$
o-Xylene	18	17.8	1	3.1
Methanol	29.6	15.1	12.3	22.3
Ethanol	26.5	15.8	8.8	19.4
2,2,2-Trifluoro Ethanol	24.02	15.47	8.3	16.4

 Table S4. Calculated interfacial surface energy for NPDI, BHJ, and PUA.

NPDI	E (oxidation)	E (reduction)	Band gap	НОМО	LUMO
Conventional	2.44	-0.78	-3.23	-7.33	-4.10
Coherent Arrangement	2.46	-0.74	-3.21	-7.35	-4.14

**Table S5.** Photovoltaic parameters for OPVs with a coherent arrangement of NPDI (area:  $0.15 \text{ cm}^2$ ).

	V <sub>oc</sub> (V)	J <sub>SC</sub> (mA/cm <sup>2</sup> )	J <sub>SC,EQE</sub> (mA/cm <sup>2</sup> )	FF	PCE (%)
C- NPDI	0.728	28.14	25.78	0.59	12.04
A- NPDI	0.789	28.14	26.58	0.65	14.45

**Table S6.** Electron mobility of electron-only devices containing a coherent arrangement of

 NPDI interlayer.

	Electron mobility (cm <sup>2</sup> V <sup>-1</sup> s <sup>-1</sup> )
C- NPDI	7.905 × 10 <sup>-6</sup>
A- NPDI	1.730 × 10 <sup>-5</sup>

**Table S7.** Trap density of electron-only devices containing a coherent arrangement of NPDI interlayer.

	V <sub>TFL</sub>	N <sub>trap</sub> (# cm <sup>-3</sup> )
C-NPDI	0.918	$2.117 \times 10^{16}$
A- NPDI	0.731	$1.686 \times 10^{16}$

**Table S8.** Photoresponse rate of OPVs with a coherent arrangement NPDI for on/off photocurrent ratios of 0.1 to 0.9.

	τ <sub>on</sub>	$ au_{ m off}$
	(\$)	(\$)
C-NPDI	1.31 µs	1.25 µs
A- NPDI	1.06 µs	1.22 µs

	-3 dB		Variation
	Pristine	Aged	
C-NPDI	351 kHz	345 kHz	-1.7%
A- NPDI	445 kHz	442 kHz	-0.6%

Table S9. Frequency-dependent responses of OPDs with coherent arrangement of NPDI.