

## Supplementary Materials

### Visible to Near-Infrared Nanocrystalline Organic Photodetector with Ultrafast Photoresponse

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**Table s1** Detectivity of OPD measured under the illumination of monochromatic light at a wavelength ( $\lambda$ ) of 850 nm) with light intensity of 0.0005, 0.005 and 0.05 mW/cm<sup>2</sup> based on the noise current density.

850 nm (mW/cm <sup>2</sup> )	R (0 V) (mA/W)	R (-0.5 V) (mA/W)	$\frac{R\sqrt{A}}{D^*(\sqrt{i_{noise}})}$ (0 V, 10 Hz) (Jones)	$\frac{R\sqrt{A}}{D^*(\sqrt{i_{noise}})}$ (0 V, 10 <sup>5</sup> Hz) (Jones)	$\frac{R\sqrt{A}}{D^*(\sqrt{i_{noise}})}$ (-0.5 V, 10 Hz) (Jones)	$\frac{R\sqrt{A}}{D^*(\sqrt{i_{noise}})}$ (-0.5 V, 10 <sup>5</sup> Hz) (Jones)
0.0005	680	1209	$6.80 \times 10^{10}$	$1.69 \times 10^{13}$	$6.54 \times 10^{10}$	$6.59 \times 10^{12}$
0.005	558	875	$5.58 \times 10^{10}$	$1.22 \times 10^{13}$	$4.73 \times 10^{10}$	$4.77 \times 10^{12}$
0.05	535	723	$5.35 \times 10^{10}$	$1.02 \times 10^{13}$	$3.96 \times 10^{10}$	$3.99 \times 10^{12}$

**Table s2** A summary of the detail performance parameters of OPD based Y6 acceptor

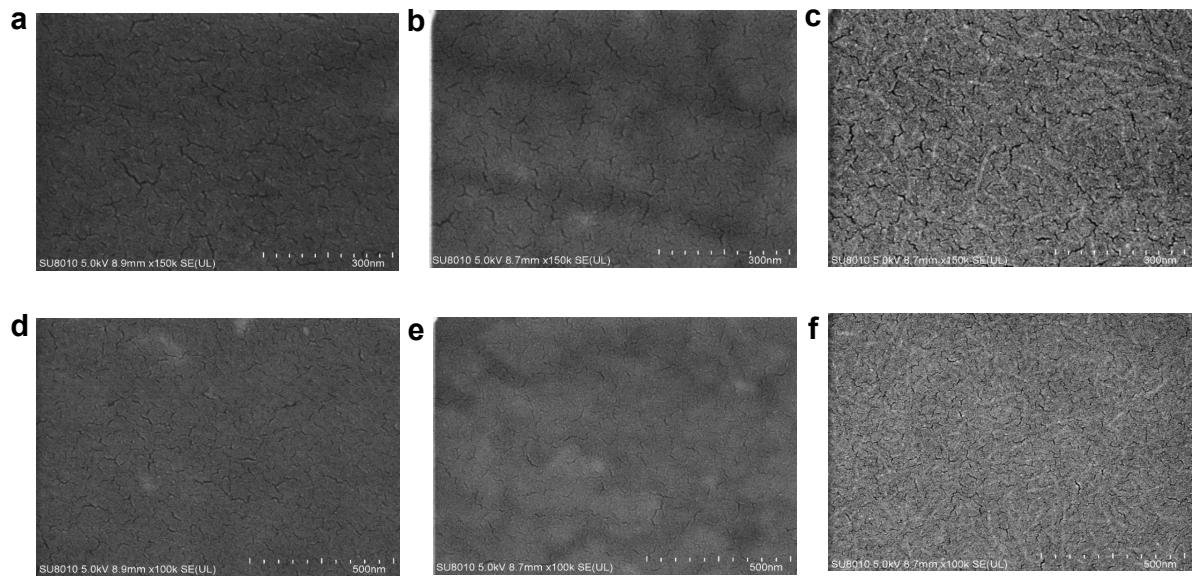
Active Layer	$R$ (mA W <sup>-1</sup> )	D* (Jones)	$J_d$ (A cm <sup>-2</sup> )	Ref.
PFT-OEHp:Y6	500 (-0.1 V)	1.16 x 10 <sup>13</sup> (-0.1 V)	5.81 x 10 <sup>-9</sup> (-0.1 V)	<sup>1</sup>
NT812:Y6 (1:4)	N/A	1.2 x 10 <sup>13</sup> (-0.1 V)	1.00 x 10 <sup>-9</sup> (-0.1 V)	<sup>2</sup>
PM6:Y6/P3HT:PC <sub>71</sub> BM	8.8(10 V)	6.8 x 10 <sup>12</sup> (10 V)	N/A	<sup>3</sup>
D18:Y6 (1:1.6)	680 (0 V)	6.35 x 10 <sup>13</sup> (0 V)	1.28 x 10 <sup>-9</sup> (0 V)	This work

**Table s3** A summary of exciton binding energy of organic materials

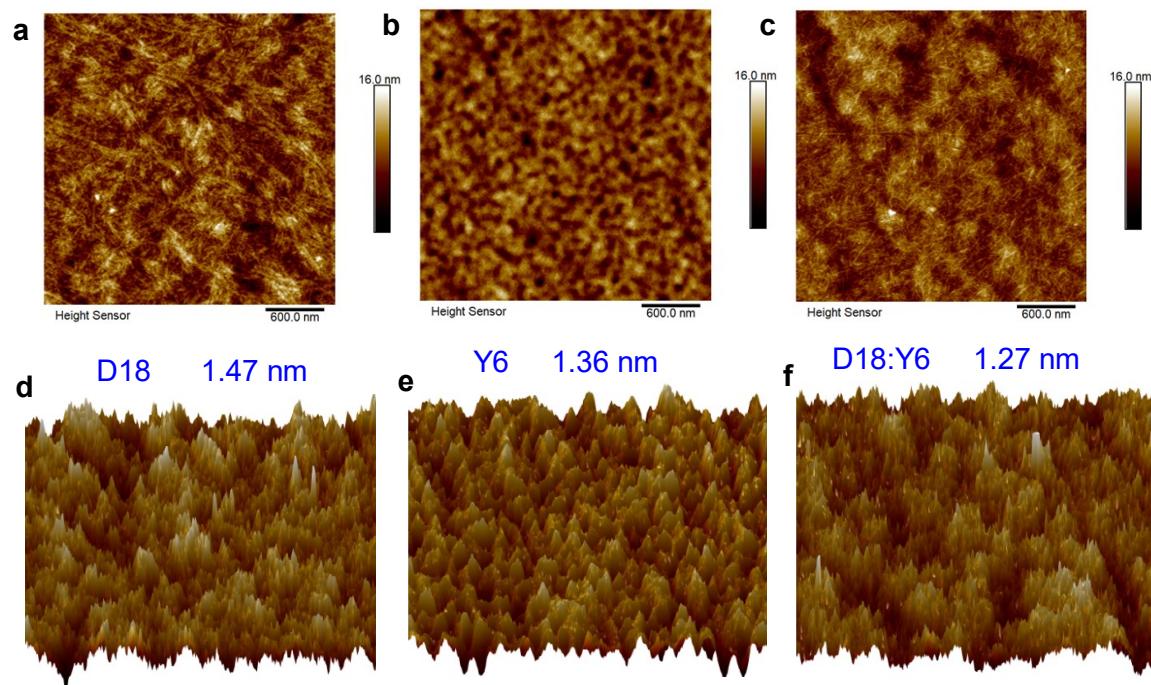
Organic materials	Binding Energy (eV)	Ref.
Alq3	1.4	M. Knupfer, H. Peisert, T. Schwieger: Phys. Rev. B 65, 033 204 (2002)
$\alpha$ -NPD	1.0	I.G. Hill, A. Kahn, Z.G. Soos, R.A. Pascal Jr.: Chem. Phys. Lett. 327, 181 (2000)
Anthracene	1.0	B. Schweitzer, H. Bässler: Synth. Met. 109, 1 (2000)
PTCDA	0.8	C.I. Wu, Y. Hirose, H. Sirringhaus, A. Kahn: Chem. Phys. Lett. 272, 43 (1997)
CuPc	0.6	I.G. Hill, A. Kahn, Z.G. Soos, R.A. Pascal Jr.: Chem. Phys. Lett. 327, 181 (2000)
$\alpha$ -6T	0.4	L.M. Blinov, S.P. Palto, G. Ruani, C. Taliani, A.A. Tevosov, S.G. Yudin, R. Zamboni: Chem. Phys. Lett. 232, 401 (1995)
Alkoxy-PPV	0.36	S.F. Alvarado, P.F. Seidler, D.G. Lidzey, D.D.C. Bradley: Phys. Rev. Lett. 81, 1082 (1998)
PFO	0.3	S.F. Alvarado, P.F. Seidler, D.G. Lidzey, D.D.C. Bradley: Phys. Rev. Lett. 81, 1082 (1998)
PPP	0.4	R. Kersting, U. Lemmer, M. Deussen, R.F. Marth, H. Kurz, V.I. Arkhipov, H. Bässler, E.O. Göbel: Phys. Rev. Lett. 73, 1440 (1994)
DO-PPP	0.2	Y. Yang, Q. Pei, A.J. Heeger: Synth. Met. 78, 263 (1996)
PT	0.6	M. Liess, S. Jeglinski, Z.V. Vardeny, M. Ozaki, K. Yoshino, Y. Ding, T. Barton: Phys. Rev. B 56, 15 712 (1997)
PA	0.5	M. Liess, S. Jeglinski, Z.V. Vardeny, M. Ozaki, K. Yoshino, Y. Ding, T. Barton: Phys. Rev. B 56, 15 712 (1997)
PDA	0.5	G. Weiser: Phys. Rev. B 45, 14 076 (1992)
C <sub>60</sub>	1.4	P.A. Brühwiler, A.J. Maxwell, A. Nilsson, N. Martensson, O. Gunnarsson: Phys. Rev. B 48, 18 296 (1993)
C <sub>70</sub>	1.0	M. Knupfer, D.M. Poirier, J.H. Weaver: Phys. Rev. B 49, 2281 (1994)
Naphthalene	1.67	H. Du, R.-C.A. Fuh, J. Li, L.A. Corkan, J.S. Lindsey, Photochemistry and Photobiology 68 (1998) 141.
Tetracene	0.81	T. Sakurai, S. Hayakawa, Japanese Journal of Applied Physics 13 (1974) 1733
Pentacene	0.55	K. Kim, Y.K. Yoon, M.O. Mun, S.P. Park, S.S. Kim, S. Im, J.H. Kim, Journal of Superconductivity 15 (2002) 595–598.
Hexacene	0.51	H. Angliker, E. Rommel, J. Wirz, Chemical Physics Letters 87 (1982) 208
Pyrene	0.73	H. Du, R.-C.A. Fuh, J. Li, L.A. Corkan, J.S. Lindsey, Photochemistry and Photobiology 68 (1998) 141
Chrysene	1.15	B. Jaeckel, J. Sambur, B.A. Parkinson, The Journal of Physical Chemistry C 113 (2009) 1837
mPtcdi	0.58	A.J. Ferguson, T.S. Jones, Journal of Physical Chemistry B 110 (2006) 6891
F12BBL6	0.43	S. Kraner, R. Scholz, C. Koerner, K. Leo, The Journal of Physical Chemistry C 2015, 119, 22820-22825
BBL3-h-F12BBL3	0.24	S. Kraner, R. Scholz, C. Koerner, K. Leo, The Journal of Physical Chemistry C 2015, 119, 22820-22825
BBL3- $\pi$ -F12BBL3	0.48	S. Kraner, R. Scholz, C. Koerner, K. Leo, The Journal of Physical Chemistry C 2015, 119, 22820-22825
INIC	1.79	L. Zhu, Y. Yi, Z. Wei, The Journal of Physical Chemistry C 2018, 122, 22309-22316
ITIC	1.84	L. Zhu, Y. Yi, Z. Wei, The Journal of Physical Chemistry C 2018, 122, 22309-22316
PC <sub>71</sub> BM	2.73	L. Zhu, Y. Yi, Z. Wei, The Journal of Physical Chemistry C 2018, 122, 22309-22316
PC <sub>61</sub> BM	2.90	L. Zhu, Y. Yi, Z. Wei, The Journal of Physical Chemistry C 2018, 122, 22309-22316
4TIC	0.04	L. Zhu, Z. Tu, Y. Yi, Z. Wei, The Journal of Physical Chemistry Lett 2019, 10, 4888-4894
COi7IC	0.25	L. Zhu, Z. Tu, Y. Yi, Z. Wei, The Journal of Physical Chemistry Lett 2019, 10, 4888-4894
Y6	-0.11-0.15	L. Zhu, J. Zhang, Y. Guo, C. Yang, Y. Yi, Z. Wei, Angew. Chem. Int. Ed. 2021, 60 15348-15353.

**Table s4** Three kinetic components of D18:Y6 thin film at 590 nm and 850 nm by TAS.

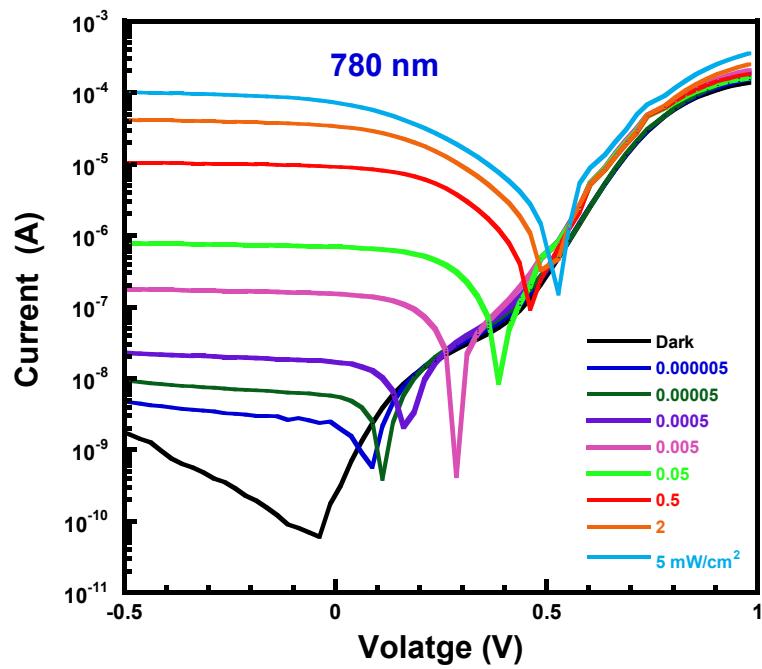
Films	D18:Y6		
Time (ps)	$\tau_1$	$\tau_2$	$\tau_3$
590 nm	0.88	14	1097
850 nm	0.86	58	1258



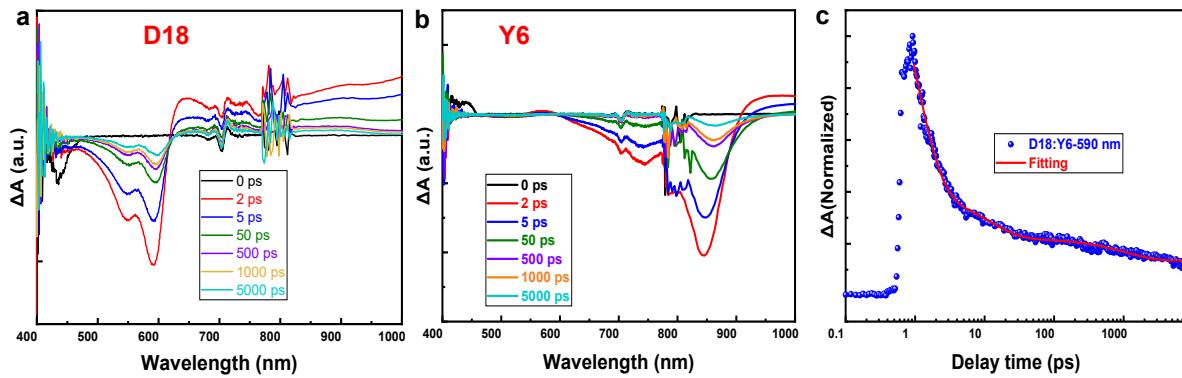
**Fig. s1** The SEM image of D18 (**a** and **d**), Y6 (**b** and **e**) and D18:Y6 (**c** and **f**) thin film.



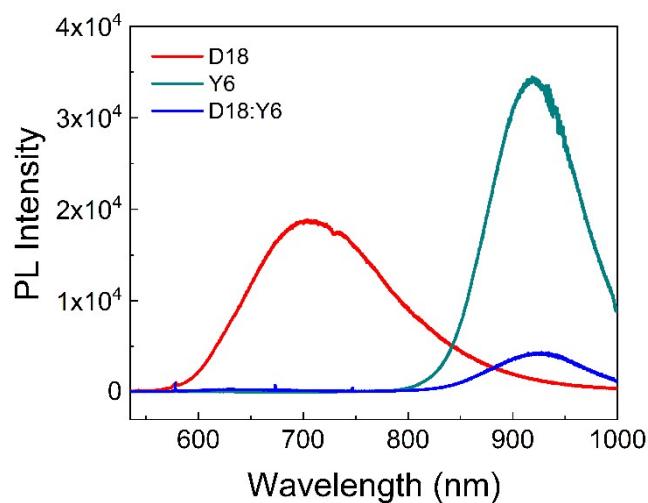
**Fig. s2** The AFM image of D18 (**a** and **d**), Y6 (**b** and **e**) and D18:Y6 (**c** and **f**) thin film.



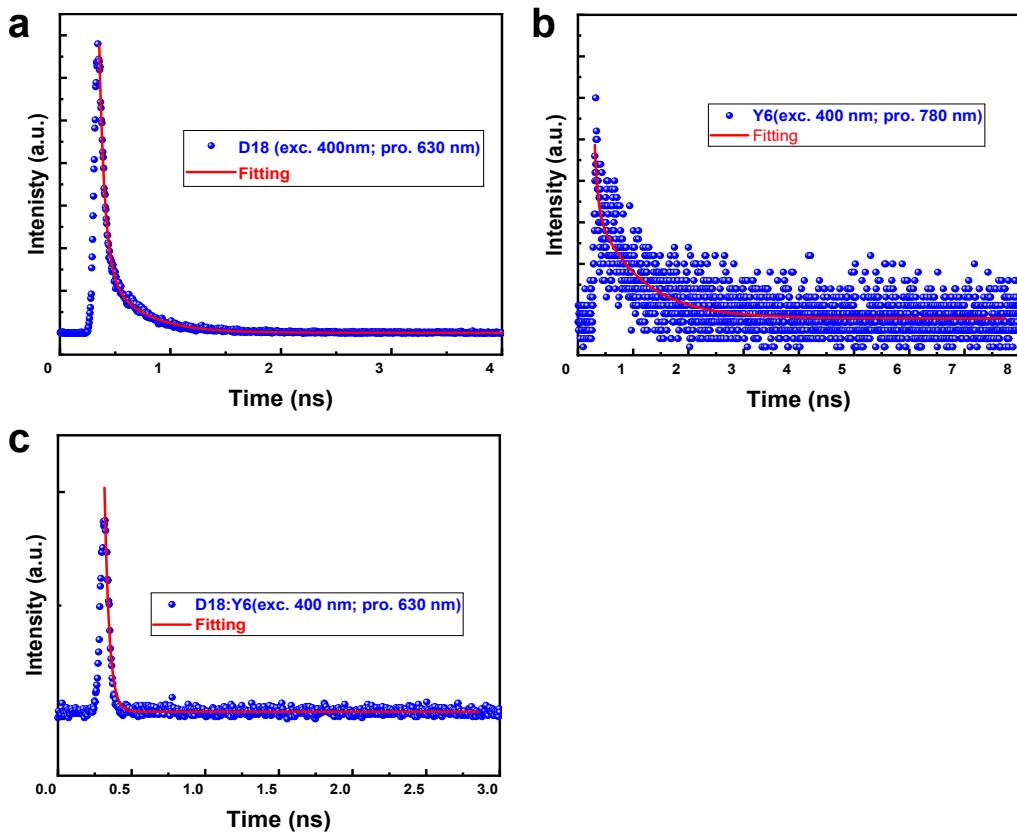
**Fig. s3** The I-V curves of PD dependence at 780 nm on power density



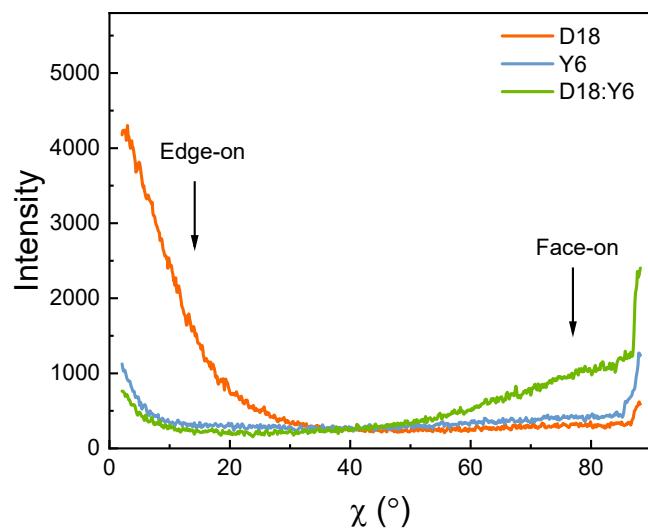
**Fig. s4** **a** The corrected femtosecond TA spectroscopy of D18 film at selected probe delay times. **b** The corrected femtosecond TA spectroscopy of Y6 film at selected probe delay times. **c** TA spectra as a function of delay time for D18:Y6 film at 590 nm.



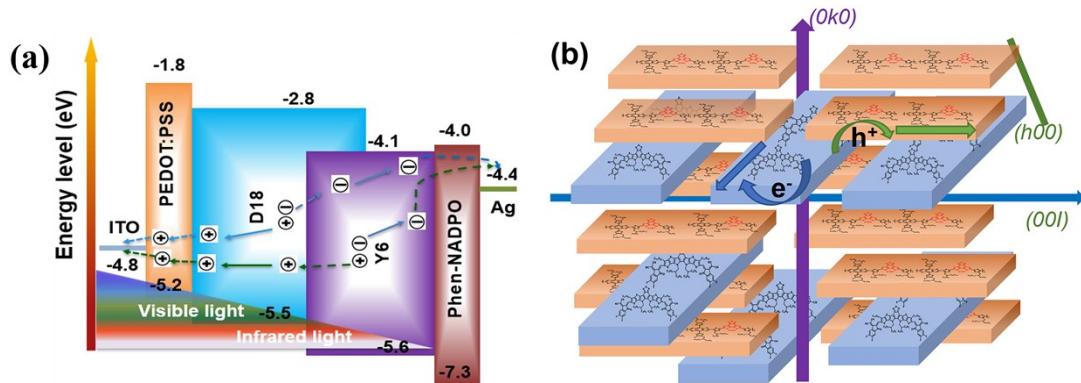
**Fig. s5.** Steady-state PL spectra of of D18, Y6 and D18:Y6 films.



**Fig. s6** **a** The time resolved photoluminescence spectra of D18 film. **b** The time resolved photoluminescence spectra of Y6 film. **c** The time resolved photoluminescence spectra of D18: Y6 film at probed wavelength of 630 nm.



**Fig. s7** The  $\chi$  scattering profiles of D18, Y6 and D18:Y6.



**Figure s8** **(a)** The corresponding energy level alignment of various thin films and charge carrier transport/extraction process. **(b)** Illustration for the lamellar packing,  $\pi-\pi$  stacking, and molecular orientation.

## **Reference**

1. Zhong, Z.; Peng, F.; Huang, Z.; Ying, L.; Yu, G.; Huang, F.; Cao, Y. *ACS Appl. Mater. Interfaces* **2020**, 12, (40), 45092-45100.
2. Xie, B.; Xie, R.; Zhang, K.; Yin, Q.; Hu, Z.; Yu, G.; Huang, F.; Cao, Y. *Nat. Commun.* **2020**, 11, (1), 2871.
3. Zhao, Z.; Wang, J.; Xu, C.; Yang, K.; Zhao, F.; Wang, K.; Zhang, X.; Zhang, F. *J. Phys. Chem. Lett.* **2020**, 11, (2), 366-373.