

## *Supporting Information*

### **A general route towards two-dimensional organic crystal-based functional fibriform transistors for wearable electronic textiles**

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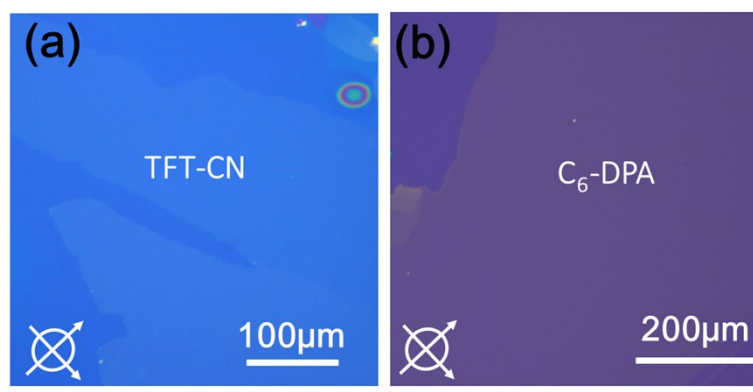
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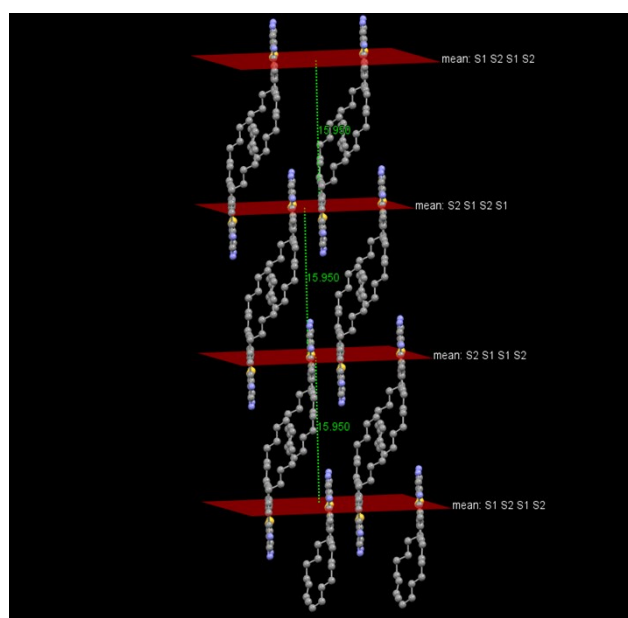
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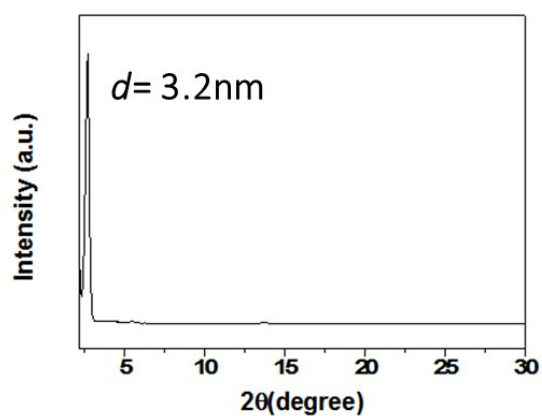
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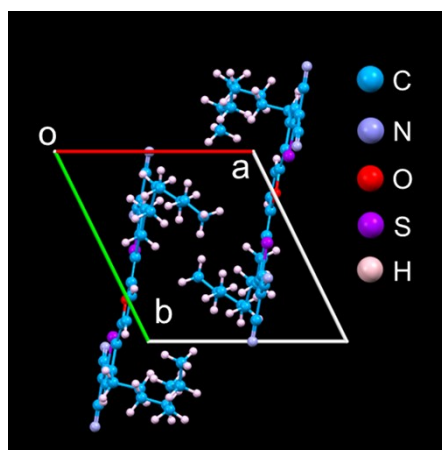
**Fig. S1** a) and b) Polarized optical microscopy images of micrometer-sized TFT-CN and C<sub>6</sub>-DPA 2DCOS on the SiO<sub>2</sub> (300 nm)/Si.



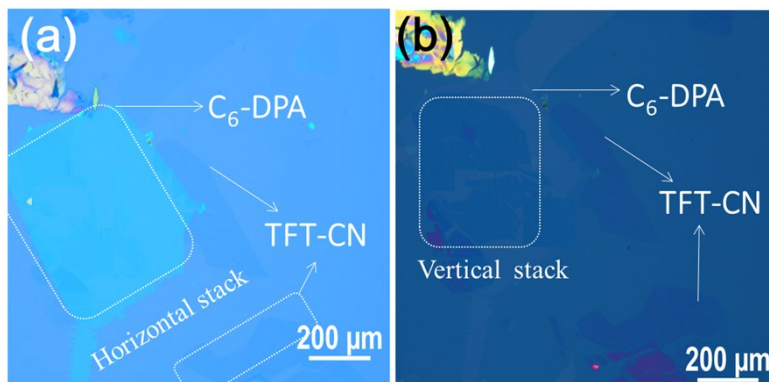
**Fig. S2** The intermolecular distance of TFT-CN



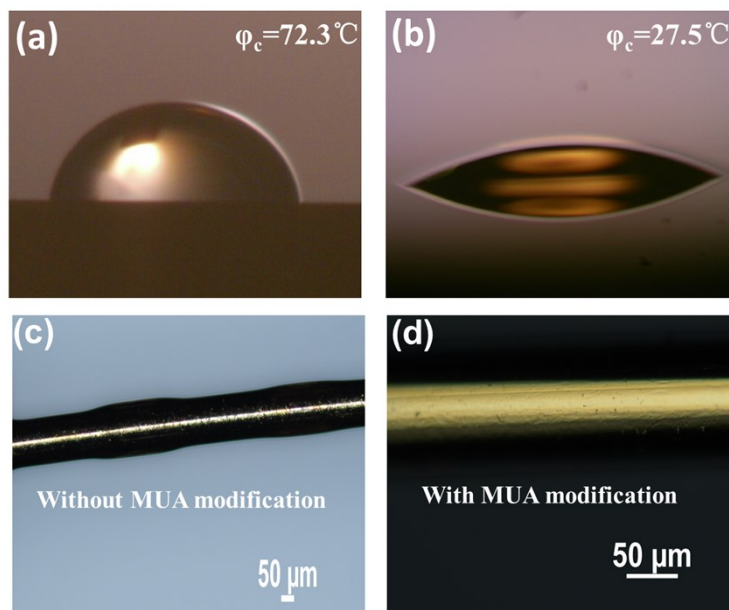
**Fig. S3** XRD patterns of C<sub>6</sub>-DPA which diffraction peaks at  $2\theta = 2.7^\circ$ ,  $5.5^\circ$ , and  $13.5^\circ$  were observed in. (The first sharp diffraction peak corresponded to a  $d$ -spacing of  $\approx 3.2$  nm)



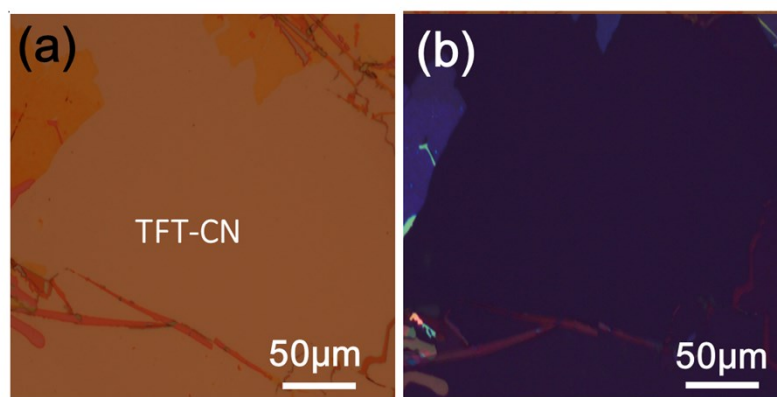
**Fig. S4** The  $\pi$ - $\pi$  interactions in TFT-CN crystal.



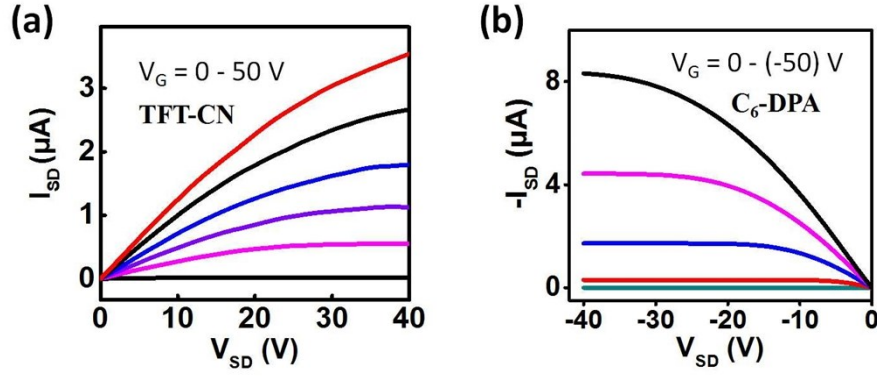
**Fig. S5** The optical microscope (a) and crosspolarized microscope images (b) of TFT-CN and C<sub>6</sub>-DPA 2DCOS with different stacking ways.



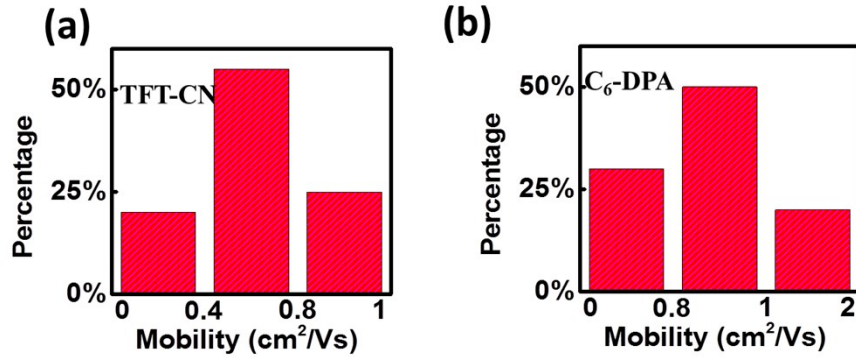
**Fig. S6** a, b) The contact angle of bare Au and MUA modified Au. c-e) The Optical microscopy image of Au wire dip-coated PMMA layer. f) The Optical microscopy image of MUA modified wire with PMMA layer.



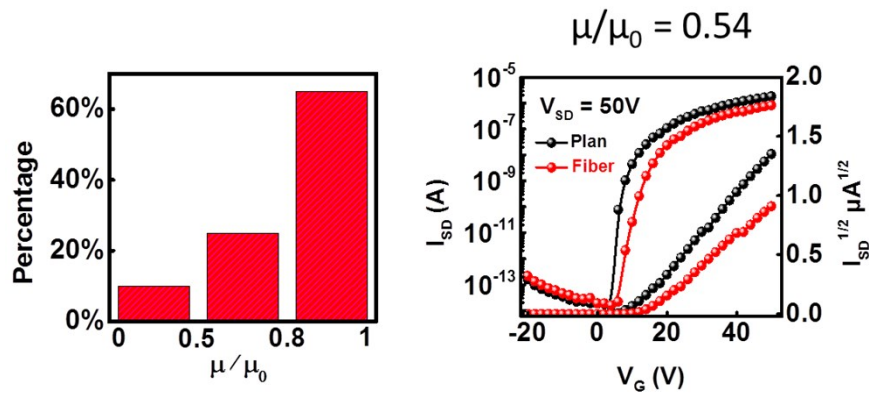
**Fig. S7** The optical microscope (a) and crosspolarized microscope images (b) of 2DCOS on the PMMA/Au flexible substrate.



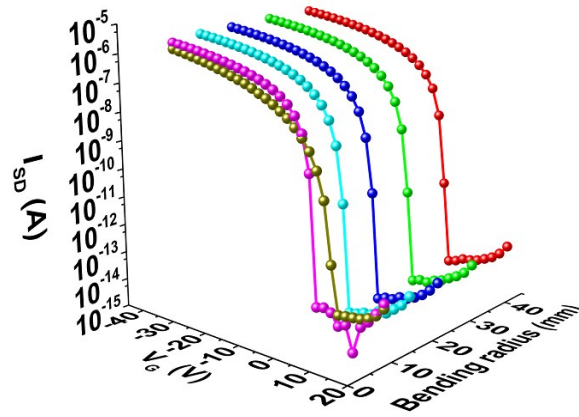
**Fig. S8** a, b) The typical output curves of the fiber-shaped OFETs based on TFT-CN and C<sub>6</sub>-DPA 2DCOS.



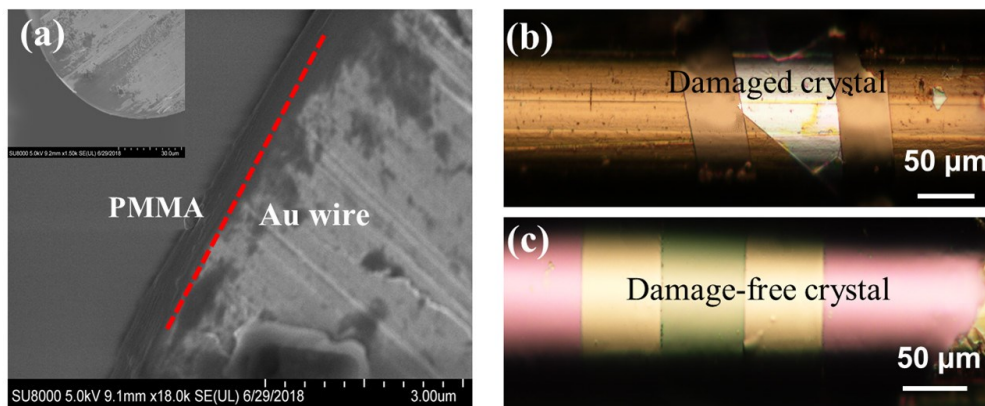
**Fig. S9** Mobility distributions based on 30 devices (a) TFT-CN, (b) C<sub>6</sub>-DPA.



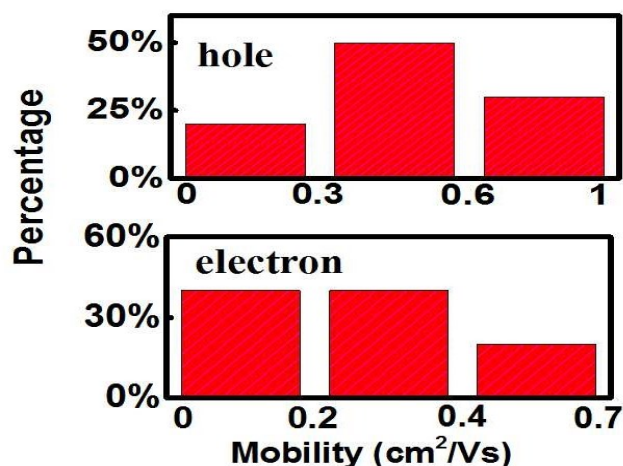
**Fig. S10** a) The variations of mobilities between the before and after transferred devices based on these 30 TFT-CN devices. b) The typical transfer curves of the before and after transferred TFT-CN device.



**Fig.S11** The transfer characteristics plotted against the bending radius of the TFT-CN 2DCOS-based OFET.



**Fig. S12** a) The SEM image of a MUA-modified Au microfiber with 780-nm PMMA layer. b) Bottom-gate/top-contact (BGTC) device fabricated by a traditional method. c) Bottom-gate/top-contact (BGTC) device fabricated by jigsaw puzzle method.



**Fig. S13** Mobility distributions of ambipolar p-n junction devices based on 10 devices.

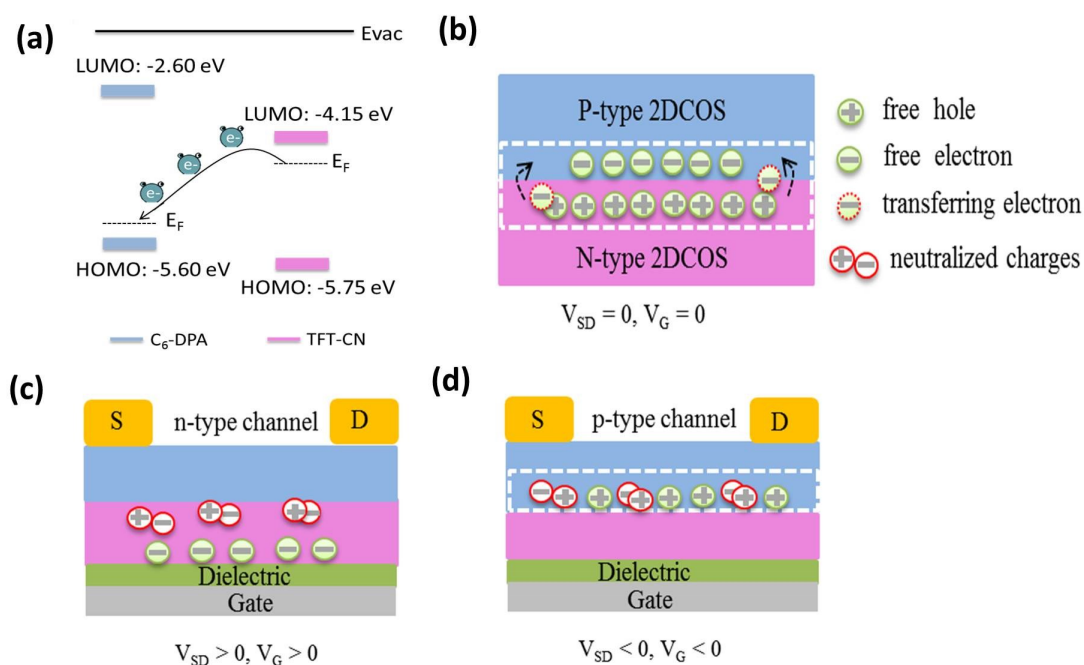
**Table S1.** Reported high performance ambipolar OFETs based on organic single-crystalline double component.

| Type             | p-type material      | n-type material       | Hole mobility (cm <sup>2</sup> V <sup>-1</sup> s <sup>-1</sup> ) | Electron mobility (cm <sup>2</sup> V <sup>-1</sup> s <sup>-1</sup> ) | Ref              |
|------------------|----------------------|-----------------------|------------------------------------------------------------------|----------------------------------------------------------------------|------------------|
| Cocrystal        | DPTTA                | C <sub>60</sub>       | 0.3                                                              | 0.01                                                                 | 1                |
| Cocrystal        | 4M-DSB               | CN-TFPA               | 0.0067                                                           | 0.67                                                                 | 2                |
| Lateral junction | Dif-TES-ADT          | BPE-PTCDI             | 0.32                                                             | 0.43                                                                 | 3                |
| Bilayer          | C <sub>8</sub> -BTBT | C <sub>60</sub>       | 0.16                                                             | 0.17                                                                 | 4                |
| Bilayer          | TIPS-PEN             | PTCDI- C <sub>8</sub> | 0.23                                                             | 0.13                                                                 | 5                |
| Bilayer          | C <sub>6</sub> -DPA  | TFT-CN                | 0.87                                                             | 0.82                                                                 | 6                |
| Bilayer          | C <sub>6</sub> -DPA  | TFT-CN                | 0.9                                                              | 0.65                                                                 | <b>This work</b> |

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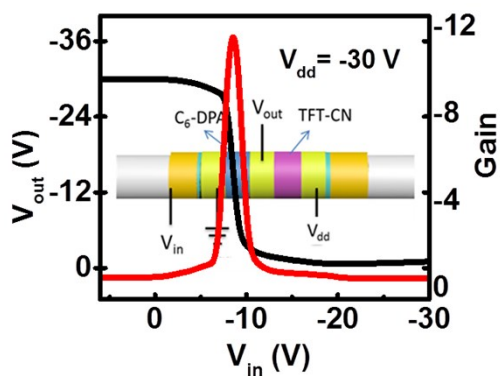


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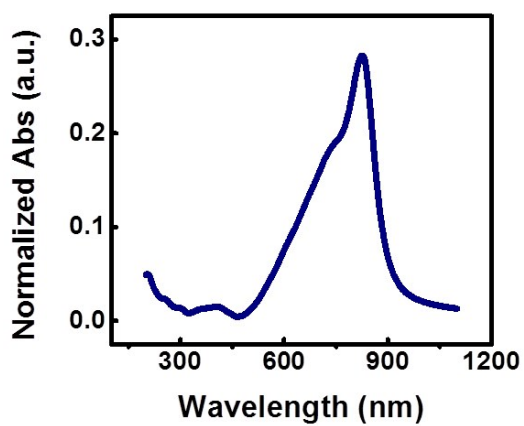


**Fig. S14** The charge transport mechanism of bilayer 2DCOS-based p-n junction. a) Energy level alignment of C<sub>6</sub>-DPA and TFT-CN. b-d) schematic illustrations of electron accumulation and hole accumulation modes of the ambipolar OFETs based on bilayer p-n junctions.

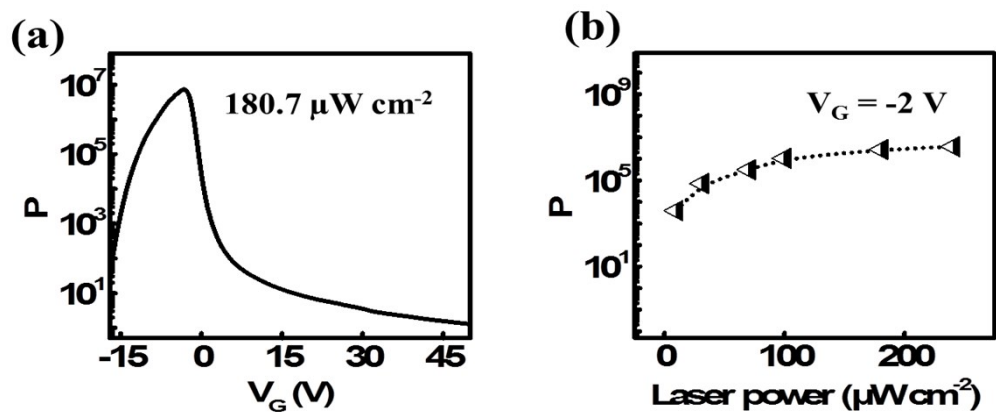




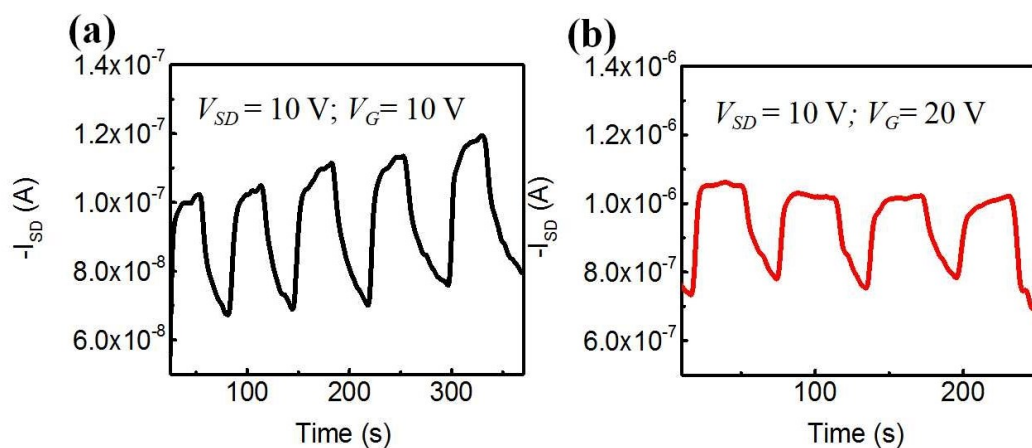
**Fig. S15** Representative static voltage-transfer characteristics of the fibriform inverter with voltage gain operating under -30 V.



**Fig.S16** The absorption spectrum of the TFT-CN film



**Fig. S17** The  $P$  of the fibriform phototransistors as a function of (a)  $V_G$  and (b) illumination intensity.



**Fig. S18** Photoswitch of the fibriform NIR phototransistor operated at  $V_G = 10$  V,  $V_{SD} = 10$  V and  $V_G = 20$  V,  $V_{SD} = 10$  V based on an illumination intensity of  $180.7 \text{ P}$ .