### **Supporting Materials**

### **Type-III Organic/Two-Dimensional multi-layer Phototransistors**

#### with Promoted Operation Speed at Communication Band

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1. The EDS of graphene/HAT-CN/Bi<sub>2</sub>O<sub>2</sub>Se on SiO<sub>2</sub> substrate.



Fig. S1. The EDS of our device can distinguish  $Bi_2O_2Se$  and graphene/HAT-CN.

2. The Raman spectrum of HAT-CN on SiO<sub>2</sub> substrate.



Fig. S2. The Raman spectrum of HAT-CN can distinguish C-C peak and C-N peak.



3. The  $V_g$  modulation on graphene/HAT-CN/Bi\_2O\_2Se phototransistor.

Fig. S3. The IV and photocurrent measurements at different gate voltages under the 850 nm with 60 mW/cm<sup>2</sup>





Fig. S4. Optoelectronic characterization and measurements of graphene/ $Bi_2O_2Se$  phototransistor. (a) The V<sub>ds</sub> curve with varied Vg applying. (b) The energy band schematic of graphene/ $Bi_2O_2Se$ . (c) Transient response performance of graphene/ $Bi_2O_2Se$ .

5. Photocurrent measurements at increasing input power density.



**Fig. S5.** Transient photocurrent measurements at increasing input power density of graphene/Bi<sub>2</sub>O<sub>2</sub>Se and graphene/HAT-CN/Bi<sub>2</sub>O<sub>2</sub>Se control devices.





Fig. S6. (a,b) The dynamic response time of graphene/HAT-CN/ $Bi_2O_2Se$  with the irradiated light at 850 and 1310 nm.

7. The thickness dependent performance of graphene/HAT-CN/Bi<sub>2</sub>O<sub>2</sub>Se.



**Fig. S7.** The responsivity and recover time of thickness various HATCN devices at 1310 nm.

# 8. The noise spectrum of graphene/Bi<sub>2</sub>O<sub>2</sub>Se



Fig. S8. The 1/f noise analysis of the device for different Vg values.

# 9. The stability of graphene/HAT-CN/Bi<sub>2</sub>O<sub>2</sub>Se



Fig. S9. (a) The detection stability of graphene/HAT-CN/ $Bi_2O_2Se$  at 1550 nm. (b) And after 45 days, the stability of graphene/HAT-CN/ $Bi_2O_2Se$ .