# **Supporting Information**

# Ultraviolet Light-Activated Charge Modulation Heterojunction for Versatile Organic Thin Film Transistors

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Outline

- 1. Supplemental photoresponse data of OTFTs
- 2. Photoluminescence spectra of pentacene thin films
- 3. Charge behavior at the heterojunctions of the P3HT-PTCDI bimolecular system

#### 1. Supplemental photoresponse data of OTFTs



**Figure S1**. Time-resolved  $I_D$  variation of the PTCDI-covered P3HT-based OTFTs operated at  $V_D$  and  $V_G$  of -10 V during the continuous on and off UV light.



**Figure S2.** Time-resolved  $I_D$  variation of the Alq3-covered P3HT-based OTFTs operated at  $V_D$  and  $V_G$  of -10 V during the continuous on and off green light.



**Figure S3.** Simulations in the logic operation of (a) PTCDI/Alq3-covered P3HT-based OTFT as an XOR gate and (b) general UV-Vis organic phototransistors as an OR gate under the control of the green and the UV light. The logic 0, 1, and -1 are shown.



**Figure S4.** Green light photoresponse of pentacene/Alq3-covered P3HT based OTFTs: (a) transfer curves ( $V_D = -10$  V); (b) time-resolved  $I_D$  variations ( $V_D = V_G = -10$  V) during the continuous on and off green light.



**Figure S5.** Green light photoresponse of LiF/Alq3-covered P3HT based OTFTs: (a) transfer curves  $(V_D = -10 \text{ V})$ ; (b) time-resolved  $I_D$  variations  $(V_D = V_G = -10 \text{ V})$  during the continuous on and off green light.

## 2. Photoluminescence spectra of pentacene thin films



Figure S6. PL spectra of the pentacene thin film excited by (a) 532 and (b) 633 nm light sources.

### 3. Charge behavior at the heterojunctions of the P3HT-PTCDI bimolecular system



**Figure S7.** Illustration of charge behavior at the heterojunction of the P3HT-PTCDI system. The molecular optimized geometries and energy levels of the bimolecular system computed using Gaussian 16 program are shown.