

## Supporting Information

### **Bilayer Chlorophyll-Based Bio-Photodetector Based on Z-Type Charge Transfer Process**

Yuting Sun<sup>a</sup>, Ziyang Liu<sup>a</sup>, Yuanlin Li<sup>a</sup>, Tianfu Xiang<sup>a</sup>, Aijun Li<sup>a</sup>, Yuhong He<sup>b</sup>, Haotong Wei<sup>b,c</sup>, Shin-ichi Sasaki<sup>d,e</sup>, Hitoshi Tamiaki<sup>d</sup>, and Xiao-Feng Wang<sup>a,\*</sup>

<sup>a</sup>Key Laboratory of Physics and Technology for Advanced Batteries (Ministry of Education), College of Physics, Jilin University, Changchun 130012, P. R. China

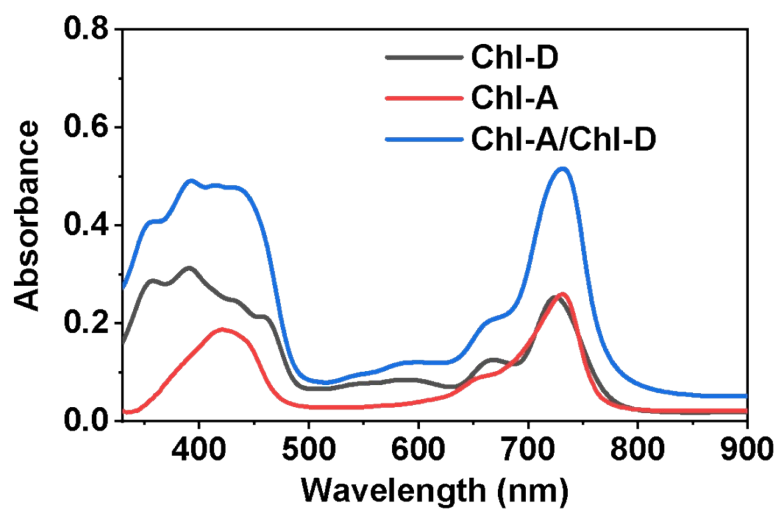
<sup>b</sup>State Key Laboratory of Supramolecular Structure and Materials, College of Chemistry, Jilin University, Changchun 130012, P.R. China

<sup>c</sup>Optical Functional Theranostics Joint Laboratory of Medicine and Chemistry, The First Hospital of Jilin University, Changchun 130012, P.R. China

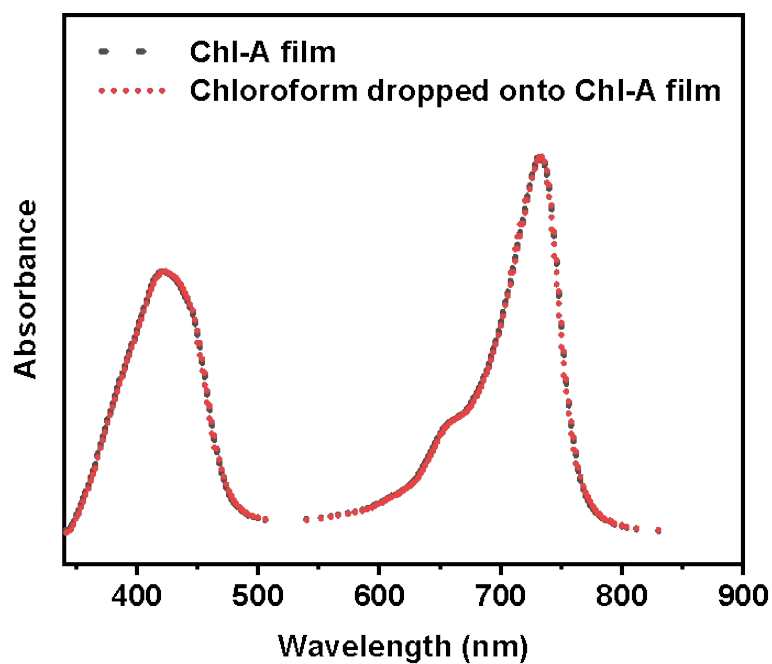
<sup>d</sup>Graduate School of Life Sciences, Ritsumeikan University, Kusatsu, Shiga 525-8577, Japan

<sup>e</sup>Department of Medical Bioscience, Faculty of Bioscience, Nagahama Institute of Bio-Science and Technology, Nagahama, Shiga 526-0829, Japan

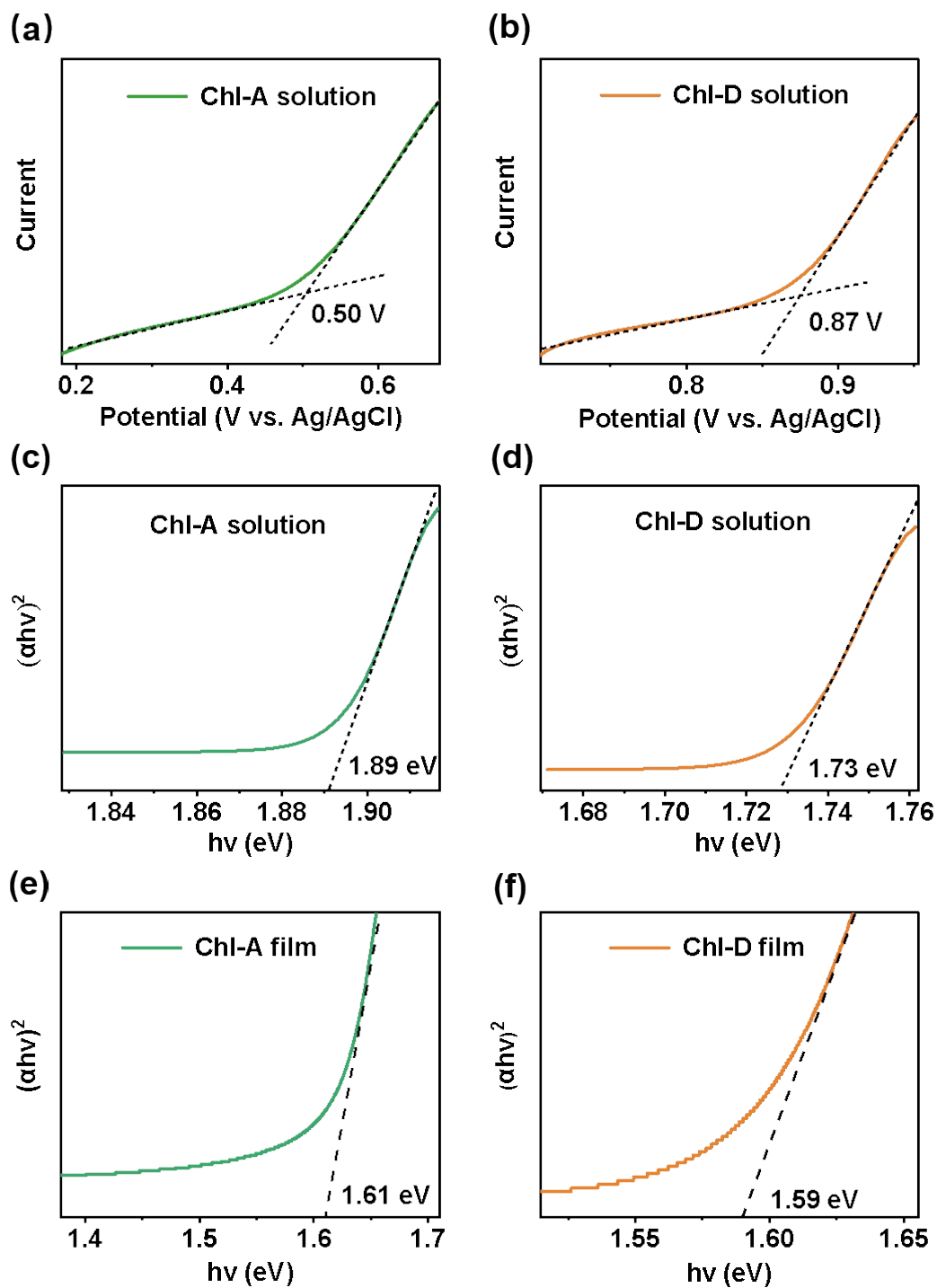
\*Corresponding author. E-mail: xf\_wang@jlu.edu.cn



**Figure S1.** The comparison of absorption spectra of Chl-D film, Chl-A film, and Chl-A/Chl-D film.



**Figure S2.** Absorption spectra of Chl-A film (black dotted line) and the already formed Chl-A film with chloroform dropped onto it (red dotted line) (PS: the operation is the same as that for the device fabrication, except without Chl-D spin-coated from chloroform).

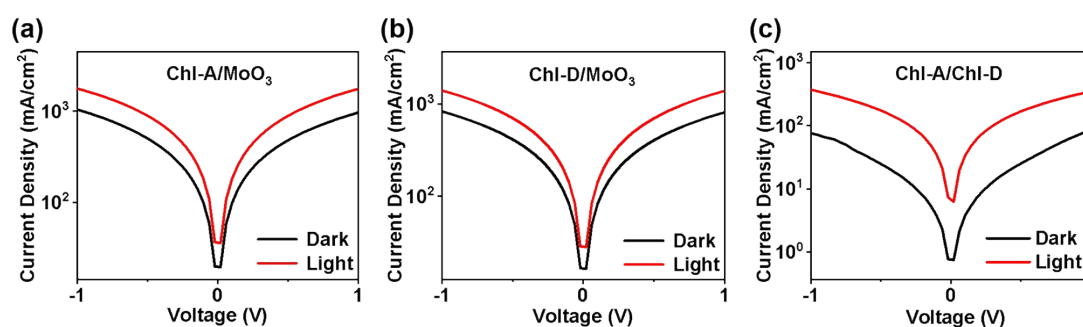


**Figure S3.** Mott-Schottky plots of (a) Chl-A solution, and (b) Chl-D solution. Tauc plots of (c) Chl-A solution, and (d) Chl-D solution. Tauc plots of (e) Chl-A film, and (f) Chl-D film.

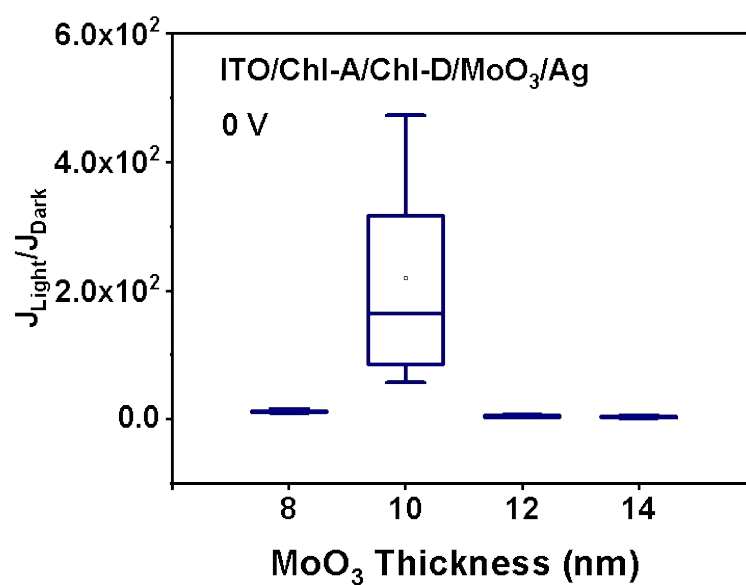
Table S1. Energy levels of Chl-A film and Chl-D film.

Chls	$E_g^{opt}$ (eV)	HOMO (eV)	LUMO (eV)
Chl-A	1.61	-4.95	-3.34
Chl-D	1.59	-5.48	-3.89

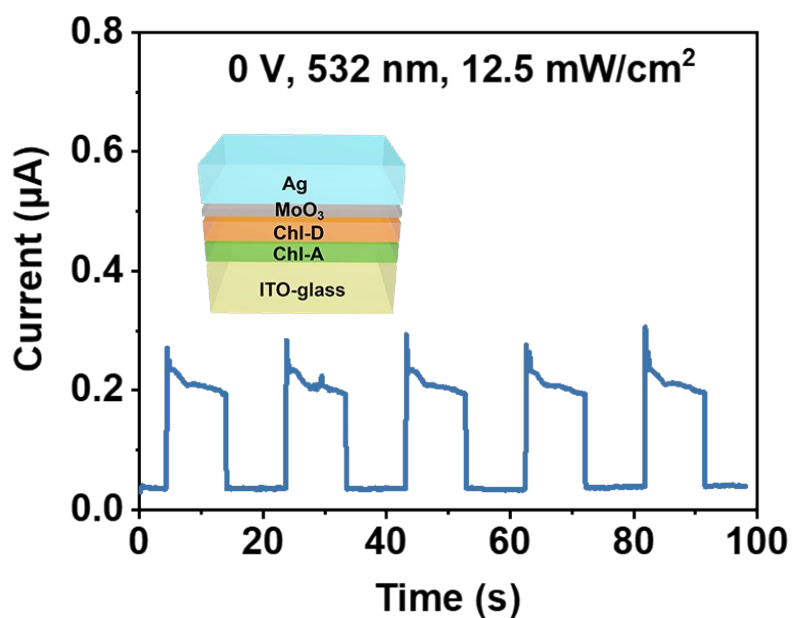
a) Band gap  $E_g$  are derived from the absorption spectra:  $E_g$  (eV) = 1240 /  $\lambda$  (nm). HOMO levels were obtained from previous studies.<sup>1</sup> LUMO levels are estimated from HOMO +  $E_g$ .



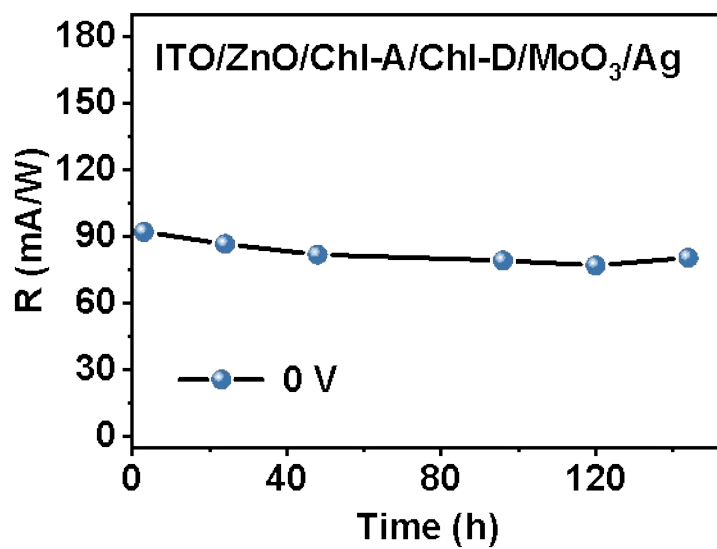
**Figure S4.** J–V curves of (a) ITO/Chl-A/MoO<sub>3</sub>/Ag, (b) ITO/Chl-D/MoO<sub>3</sub>/Ag, and (c) ITO/Chl-A/Chl-D/Ag device in light (using a solar simulator with an optical power of 100 mW/cm<sup>2</sup>) and darkness.



**Figure S5.**  $J_{\text{Light}}/J_{\text{Dark}}$  statistics of the ITO/Chl-A/Chl-D/MoO<sub>3</sub>/Ag device at various MoO<sub>3</sub> thickness.



**Figure S6.** I-t curves of the ITO/Chl-A/Chl-D/MoO<sub>3</sub>/Ag device under 532 nm laser.



**Figure S7.** Stability of the device responsivity at different time for the device at 0 V.

## References

- [1] W. Zhao, C. Dall'Agnese, S. Duan, Y. Sanehira, Y. Wei, H. Tamiaki, S. Sasaki and X.-F. Wang, Trilayer Chlorophyll-Based Cascade Biosolar Cells, *ACS Energy Lett.* 2019, **4**, 384-389.