Supporting Information

Atomic-Layer Tailored Organic Photodetectors: Harnessing Intermolecular Charge-Transfer Absorption for Expanded Spectral Sensitivity up to Telecommunication Band

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Calculations

As shown in Fig. S3(a), using the Versa STAT3 electrochemical workstation, the C-V curve of ZnPc was measured at room temperature using cyclic voltammetry in a three electrode battery. The required values for calculating the HOMO of ZnPc were obtained by analyzing the C-V curve. HOMO can be derived from equation (1),

$$HOMO = -(x - 0.22 + 4.8) \quad (1)$$

Where x is the voltage abscissa value obtained from the C-V curve calculation of ZnPc.

As shown in Fig. S1(b), by analyzing the absorption curve, the absorption band edge of ZnPc can be calculated. The LUMO energy level of ZnPc can be derived from equation (2),

$$LUMO = HOMO - (-y) \quad (2)$$

Where y is the absorption band boundary value of ZnPc.

Supplementary Figures

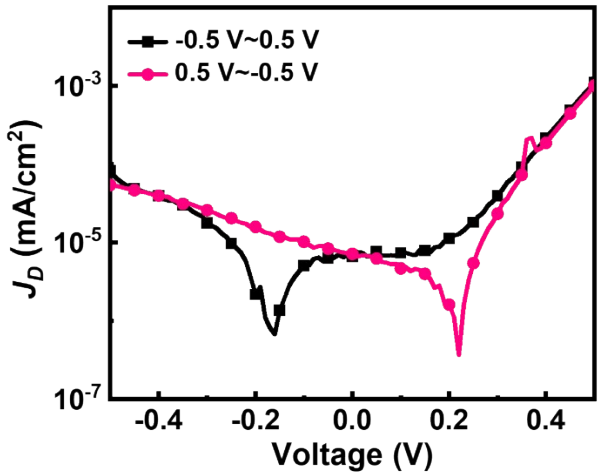

Fig. S1 The J_D-V curve of the device scanned from -0.5 V~0.5 V and 0.5 V~ -0.5 V.

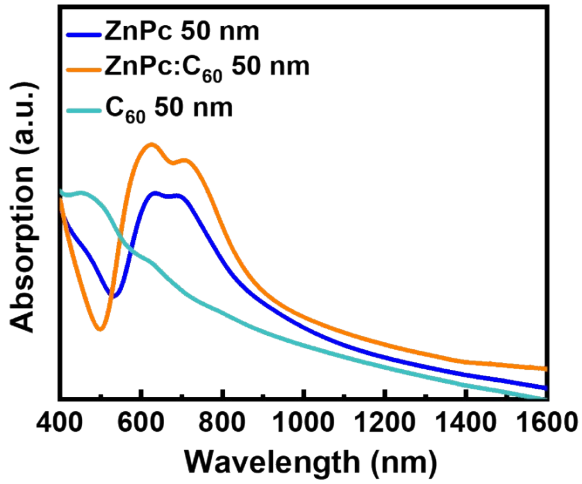

Fig. S2 Absorption spectra of ZnPc, C_{60}, and ZnPc: C_{60} films with a thickness of 50 nm.

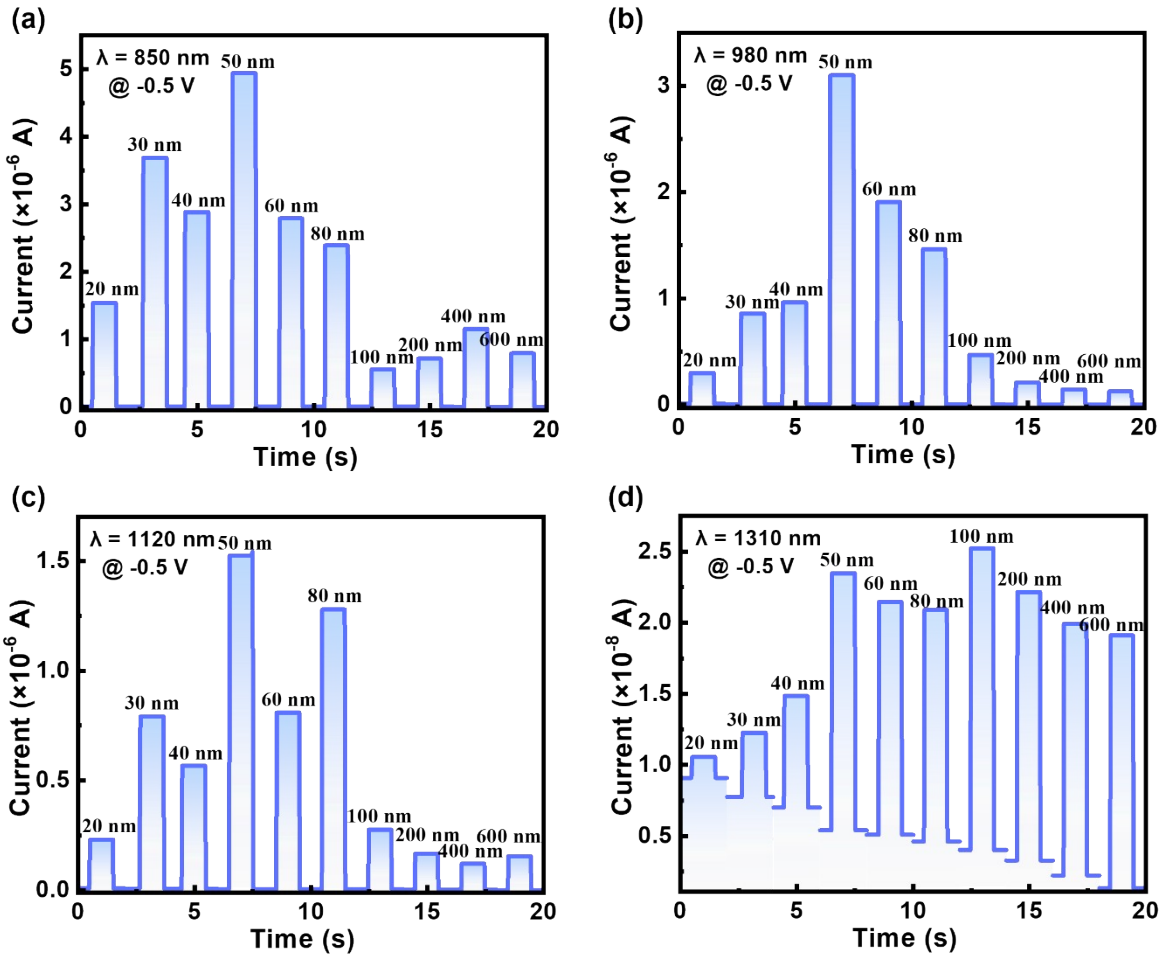

Fig. S3 Transient photocurrent responses of the OPD with different thicknesses of the ZnPc:C_{60} layer at 850 mm, 980 nm, 1120 nm, 1310 nm.

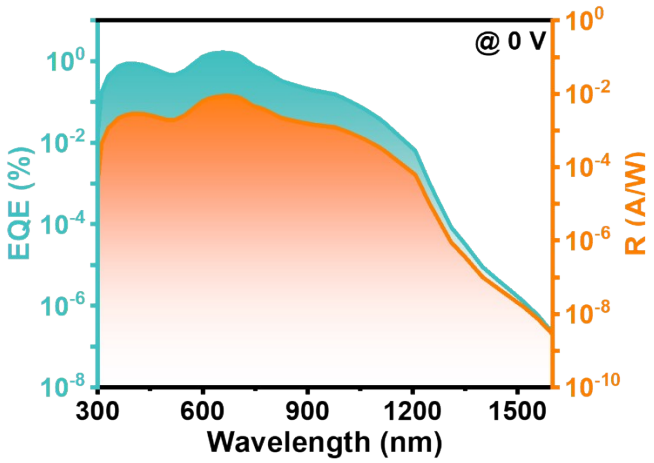

Fig. S4 EQE and R spectra of the Al_2O_3-OPD at 0 V.

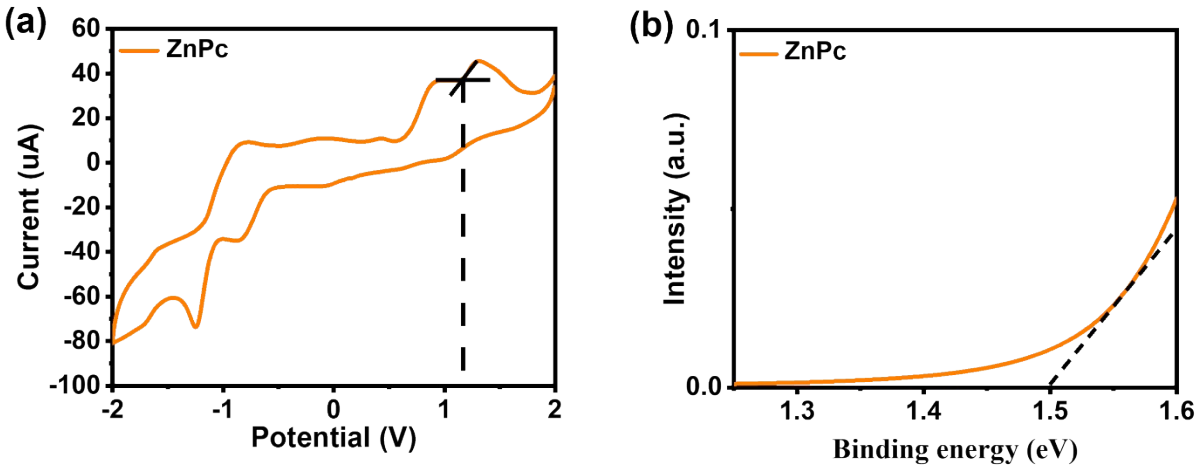

Fig. S5 (a) C-V curves of ZnPc. (b) The absorption band edge of ZnPc.

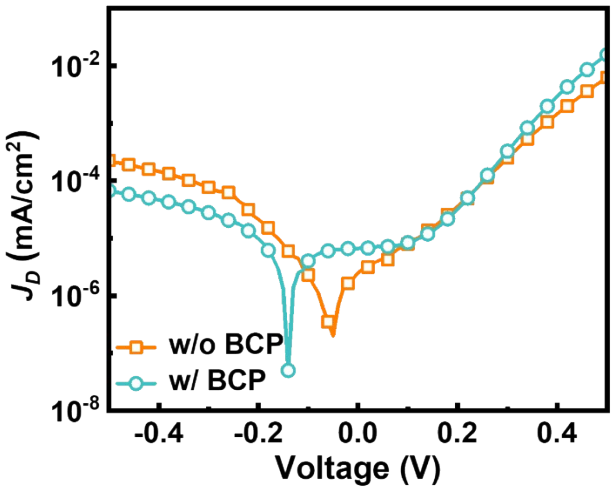

Fig. S6 *J-V* curves of the control and Al_2O_3-OPD with/without BCP hole blocking layer in dark.

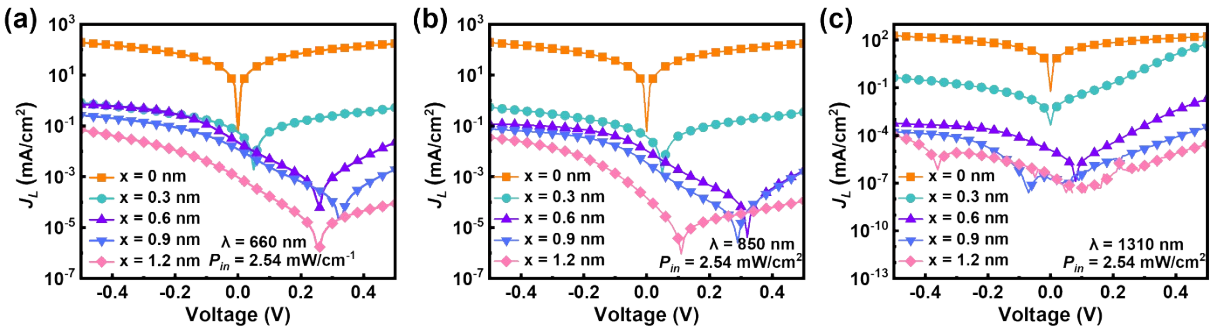

Fig. S7 (a, b) Photocurrent density curves of Al_2O_3-OPD with different thicknesses of Al_2O_3 tested under 660 nm, 850 nm and 1310 nm LEDs.

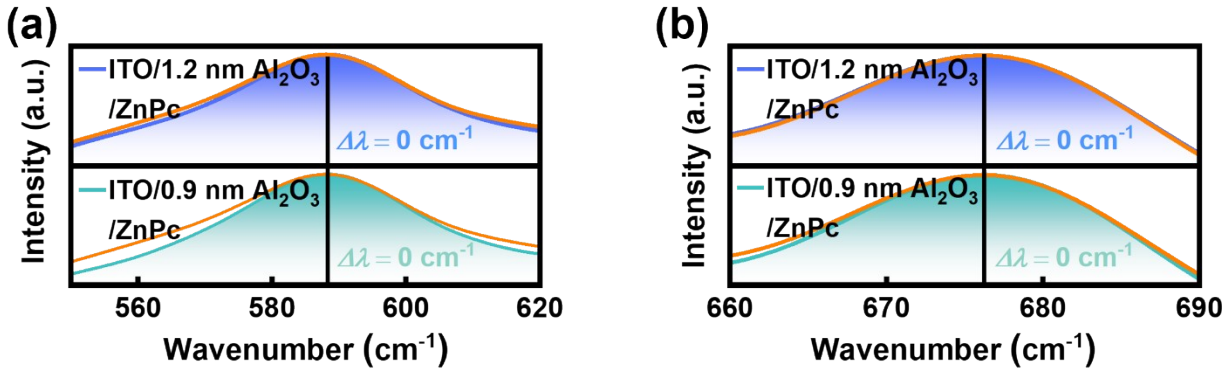

Fig. S8 (a, b) Raman spectra of the ITO/ZnPc films treated with 0.9 nm and 1.2 nm thick Al_2O_3.

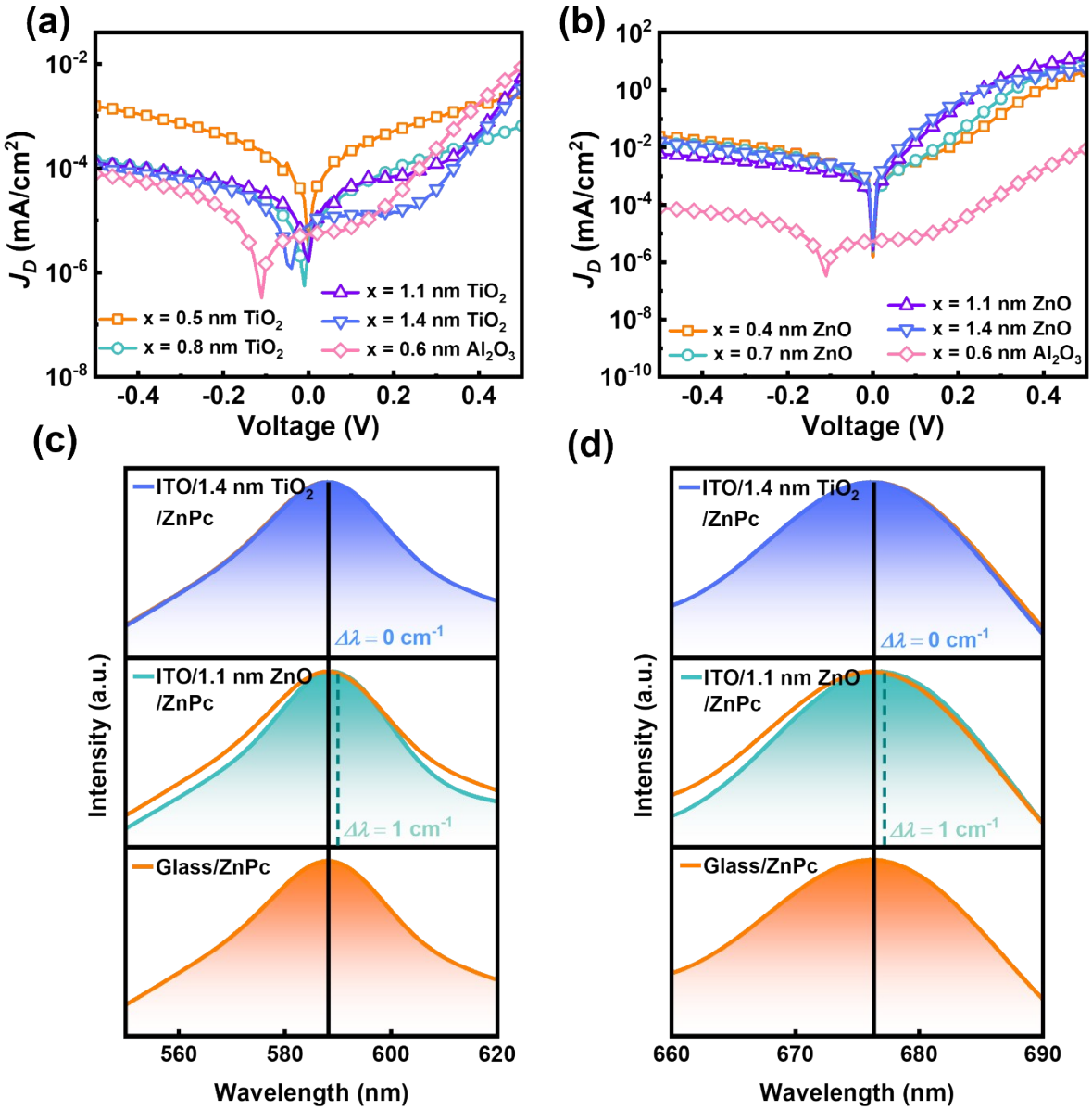

Fig. S9 (a, b) *J-V* curves of devices modified with ZnO and TiO_2 interfacial layers. (c, d) Raman spectra of the glass/ZnPc and ITO/ZnPc films treated with TiO_2 and ZnO.

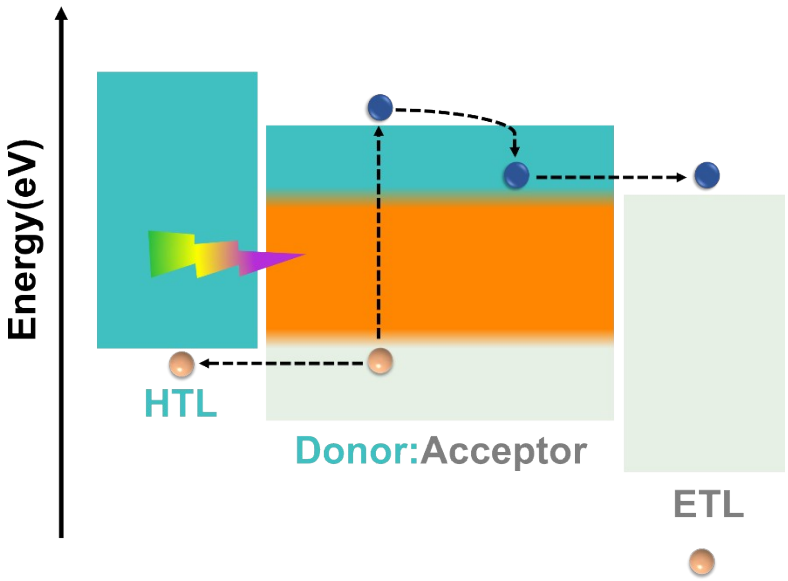

Fig. S10 Generation and dissociation of excitons when the device is illuminated by a visible light. For simplicity, only absorption of ZnPc is illustrated.

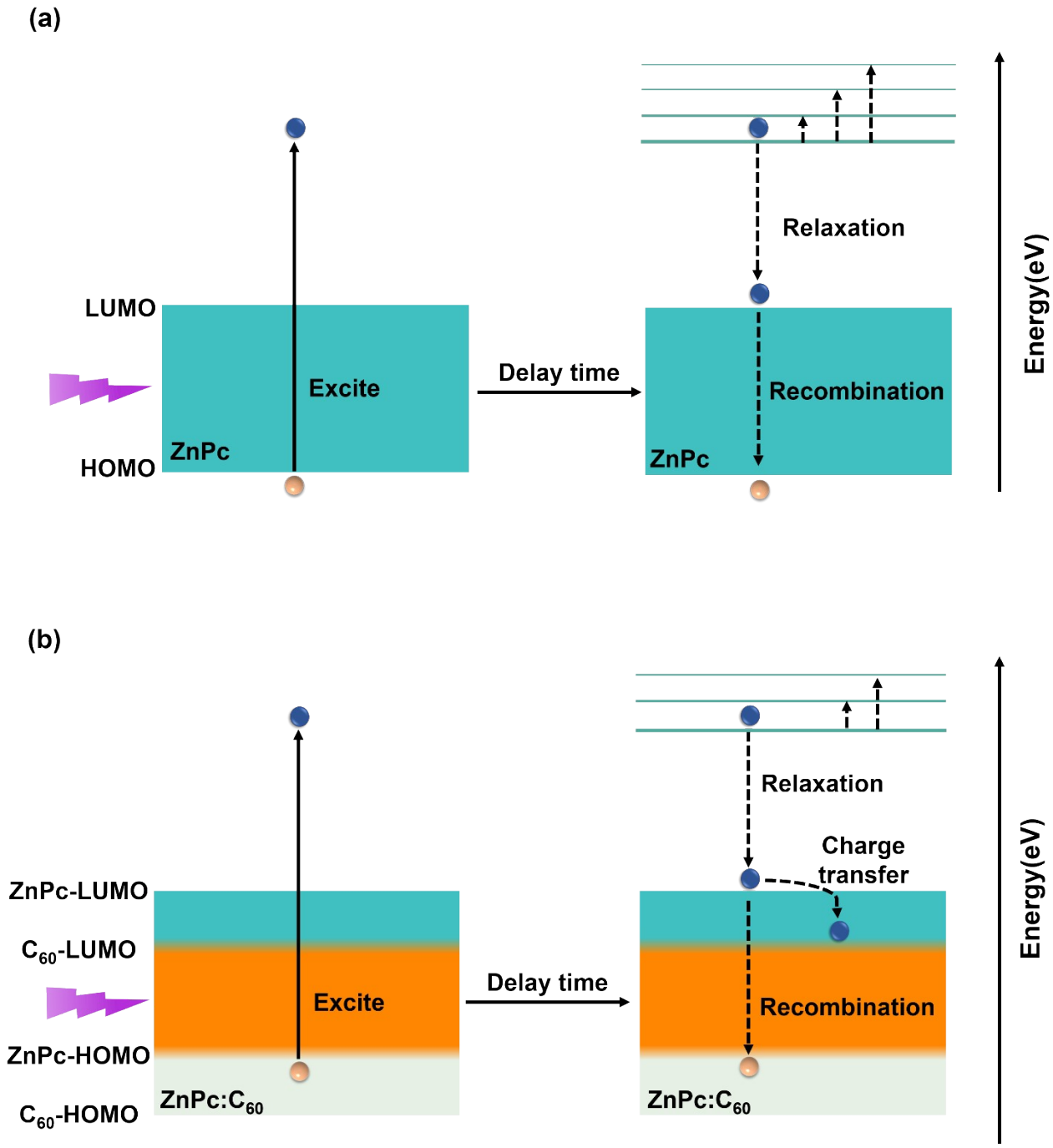

Fig. S11 Excitation and relaxation processes of electrons in (a) ZnPc film, and (b) ZnPc:C_{60} blend film after photoexcitation.