## Supporting Information

# Improved performance of ZnO based inverted organic photodetectors with morphological and interfacial modification

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### Device characterization

The current density-voltage curve (J-V curve) of the organic photodetector was measured by Keithley 4200 digital source meter. The external quantum efficiency IPCE spectrum was measured by the solar cell quantum efficiency test system, and the additional bias was applied by the Keithley 2400. Impedance spectrum (Is) of the device was measured by electrochemical workstation CHI760E. The response speed was measured by 685 nm pulsed laser, DLPCA-200 current amplifier and DSOS204A oscilloscope. Mott-Schottky curves, linear dynamic range, C-F curves and capacitance-voltage curves were measured using the multifunctional organic photoelectric measurement system (Paios). The thickness of the film was measured by Bruker Step-meter-Surface Profiler. The surface morphology and roughness of the film were measured by the Bruker atomic force microscope Dimension Icon. The noise spectrum density was measured by the low frequency noise test system (LFN-1000), Wuxi Xinjian semiconductor-Tech Co,.Ltd

### Device optimization using DCB as solvent

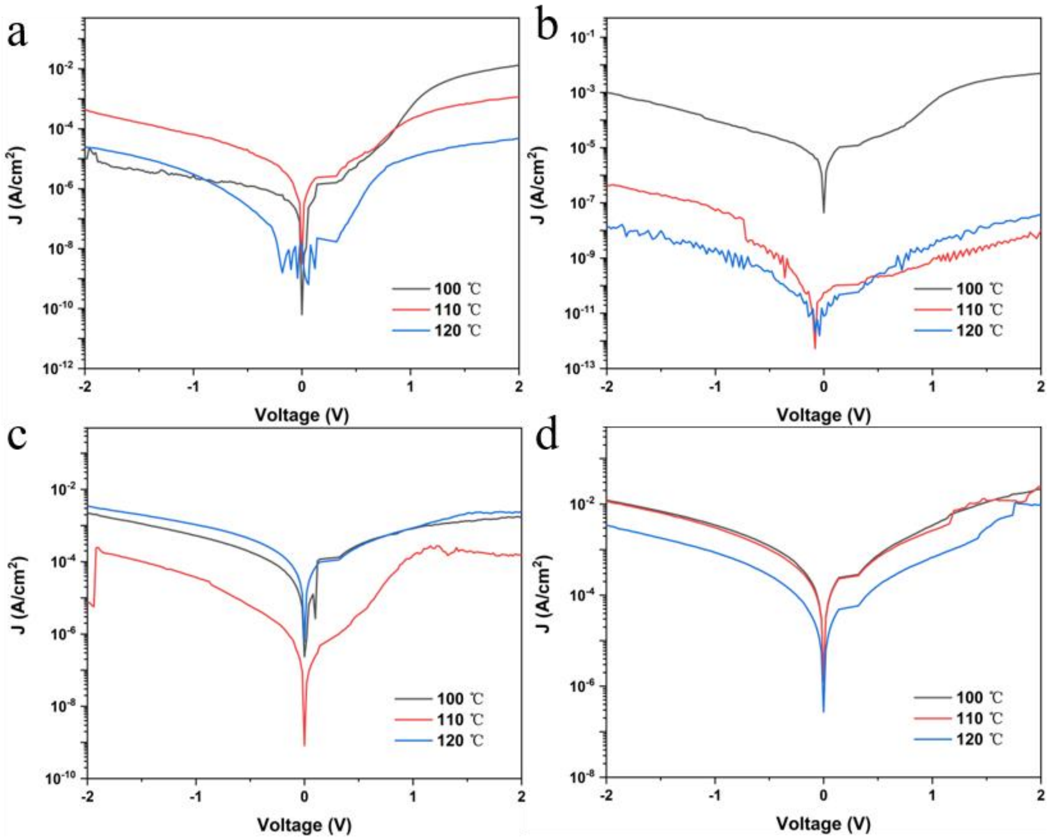


**Figure S1.** Dark current density of the pristine ZnO (p-ZnO) based devices using DCB as solvent with the thermal annealing of 100 °C~120 °C and the active layer thickness of (a)195 nm, (b)170 nm, (c)145 nm, (d)125 nm.

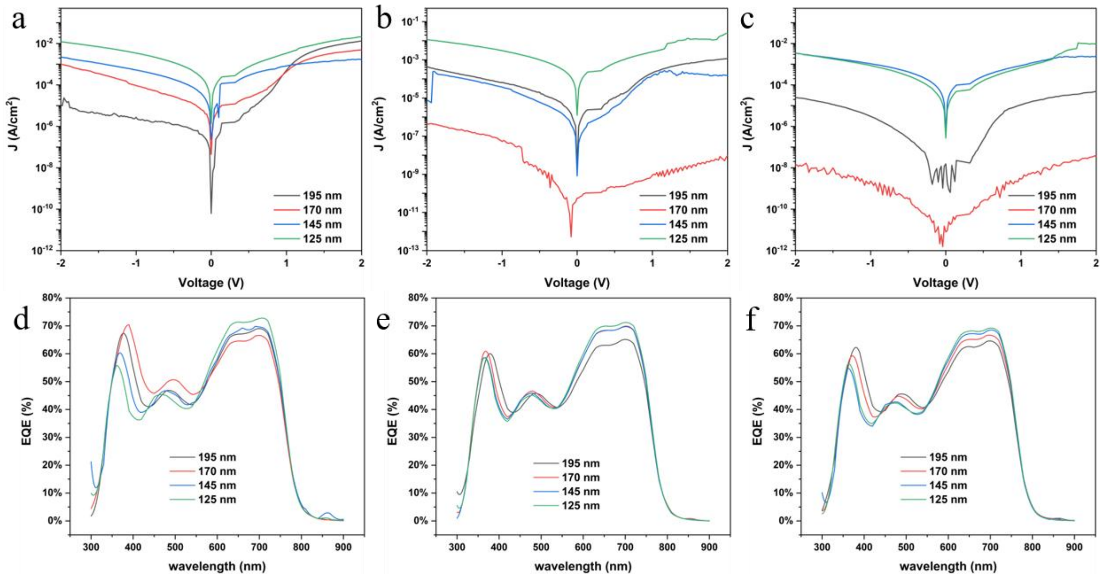


**Figure S2.** Dark current density (a) 100 °C, (b)110 °C, (c)120 °C, and EQE (d) 100 °C, (e)110 °C, (f)120 °C, of the p-ZnO based devices using DCB as solvent with the thermal annealing of 100 °C~120 °C.

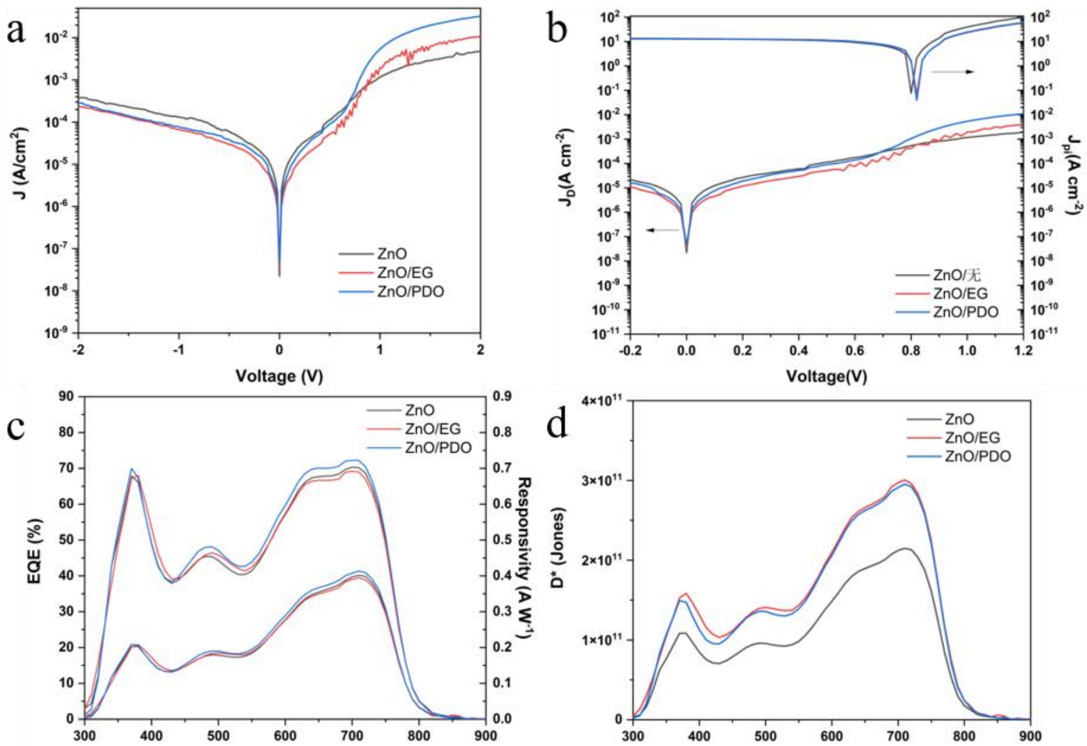


**Figure S3.** Performance of p-ZnO and modified ZnO based devices using DCB as solvent: (a) Dark current density, (b) Dark current density and photocurrent density, (c) External quantum efficiency and responsivity, (d)Specific detectivity.

**Table S1.** Main performance parameters of organic photodetectors at 730 nm

| ETL | Bias (V) | $J_D$ (A/cm$^2$) | EQE (%) | R (A/W) | D*(Jones) |
|---|---|---|---|---|---|
| **ZnO** | -0.1 | $1.09*10^{-5}$ | 70.3 | 0.397 | $2.12*10^{11}$ |
| **ZnO/EG** | -0.1 | $5.40*10^{-6}$ | 69.1 | 0.390 | $2.97*10^{11}$ |

| | | | | | |
|---|---|---|---|---|---|
| **ZnO/PDO** | -0.1 | $6.13*10^{-6}$ | 72.2 | 0.407 | $2.91*10^{11}$ |

## Device optimization using CB as solvent

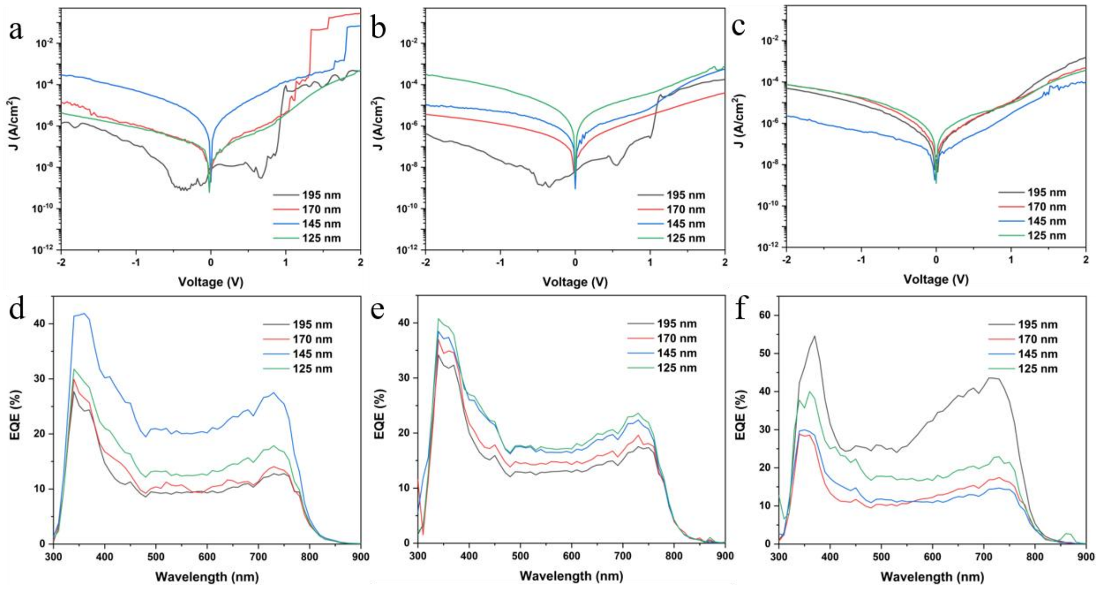


**Figure S4.** Dark current density (a) 100 °C, (b)110 °C, (c)120 °C, and EQE (d) 100 °C, (e)110 °C, (f)120 °C, of the p-ZnO based devices using CB as solvent with the thermal annealing of 100 °C~120 °C.

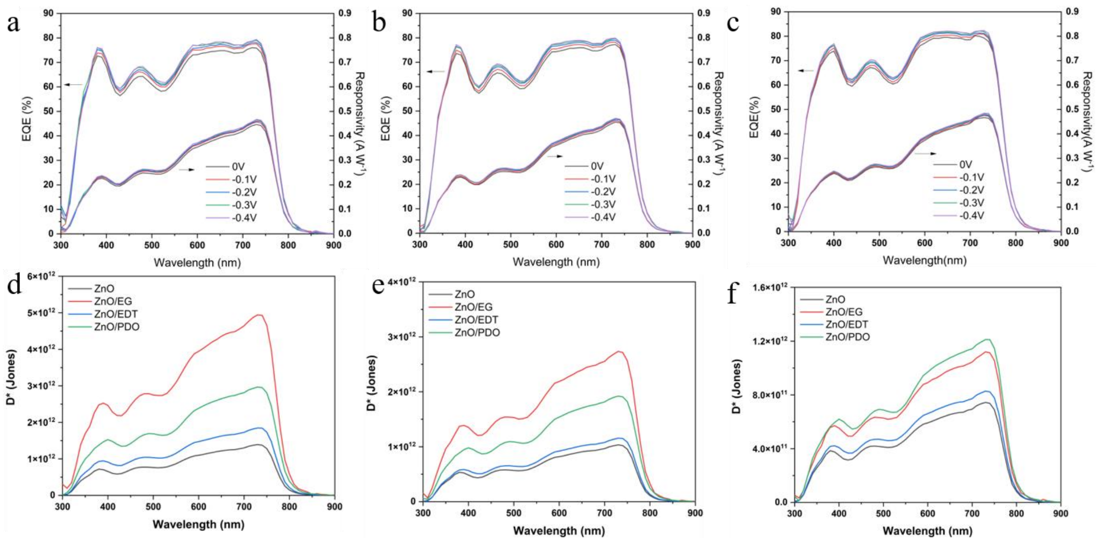


**Figure S5.** The EQE of (a) ZnO/EG, (b) ZnO/EDT, (c) ZnO/PDO based devices using CB as solvent under different bias; Specific detectivity of p- and m-ZnO based devices under the bias of (d) -0.2 V, (e) -0.3 V, (f) -0.4 V.

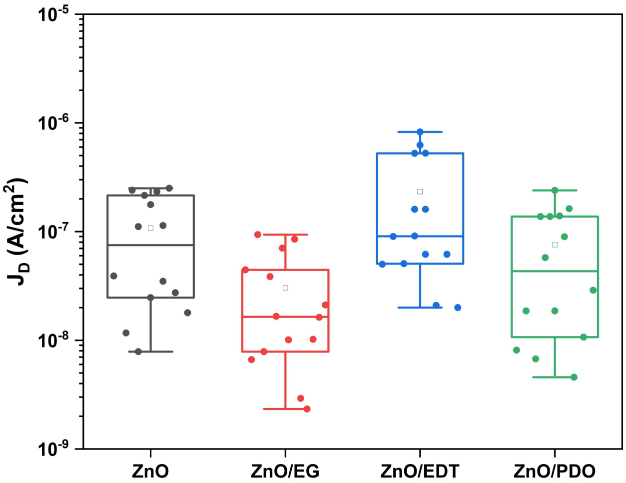


**Figure S6.** The boxplot of dark current density of p- and m-ZnO based devices under -0.1 V bias.

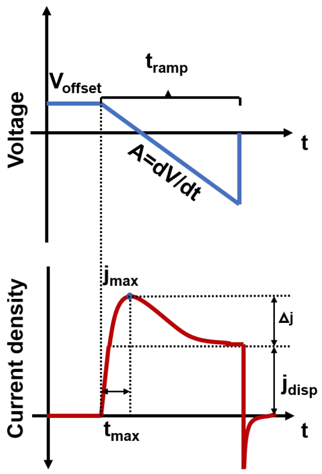


**Figure S7.** Schematic illustration of the photo-CELIV method.

In photo-CELIV, free charge carriers are generated by a light pulse and are subsequential by a voltage ramp. A light emitting diode was used as a light source. Carrier mobility is calculated by the following equation,

$$\boldsymbol{\mu} = \frac{2 \cdot d^2}{3 \cdot A \cdot t_{max}^2} \cdot \frac{1}{1+0.36\frac{\Delta j}{j_{disp}}} \tag{1}$$

where μ is the carrier mobility, d is the active layer thickness, A is the ramp rate of 200 V/ms, which is defined as $\boldsymbol{A} = \frac{dV}{dt}$, $t_{max}$ is the time of peak current, $j_{disp}$ is displacement current, Δj is peak current minus displacement current. The $t_{max}$, the $j_{disp}$ and the Δj can be estimated using the photo-CELIV transient currents.

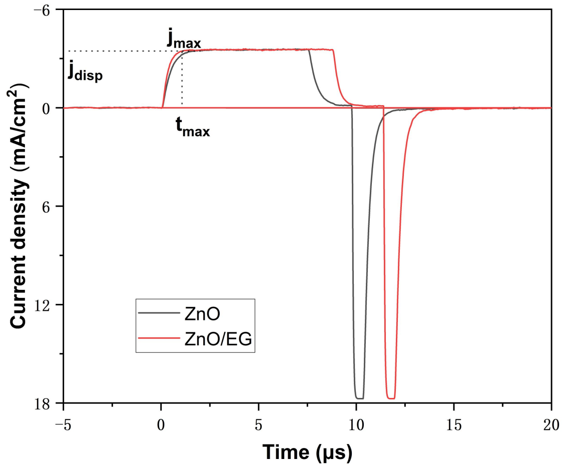


**Figure S8.** Current density-Time curves of ZnO and ZnO/EG based devices.

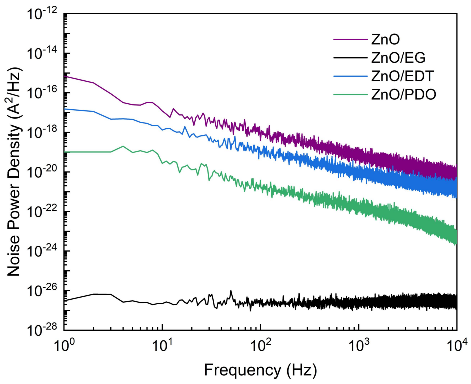


**Figure S9.** The noise current spectral densities of the p- and m-ZnO based devices at -0.1 V.

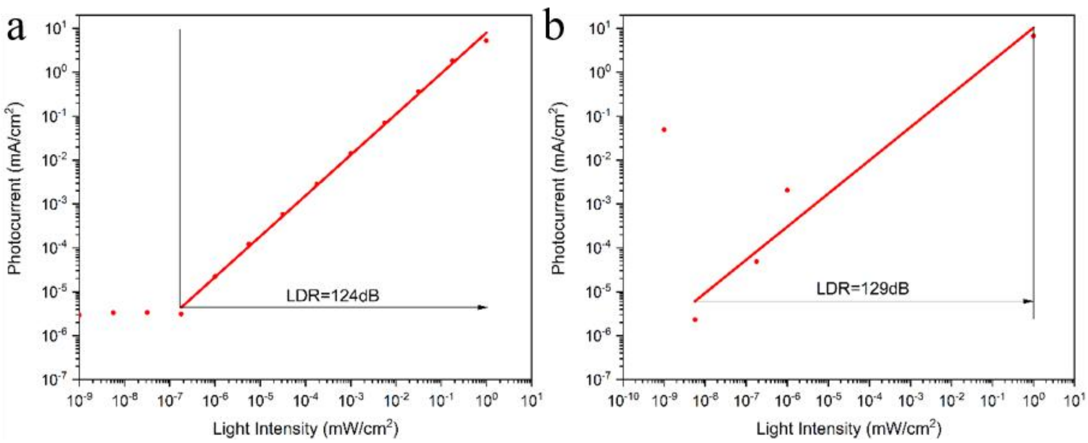


**Figure S10.** Linear dynamic range of (a) ZnO/EDT, and (b) ZnO/PDO based OPDs under -0.1 V bias.