Supporting Information

# Sign Reversal of Magneto-Capacitance in an Organic Heterojunction based Opto-Spintronic System

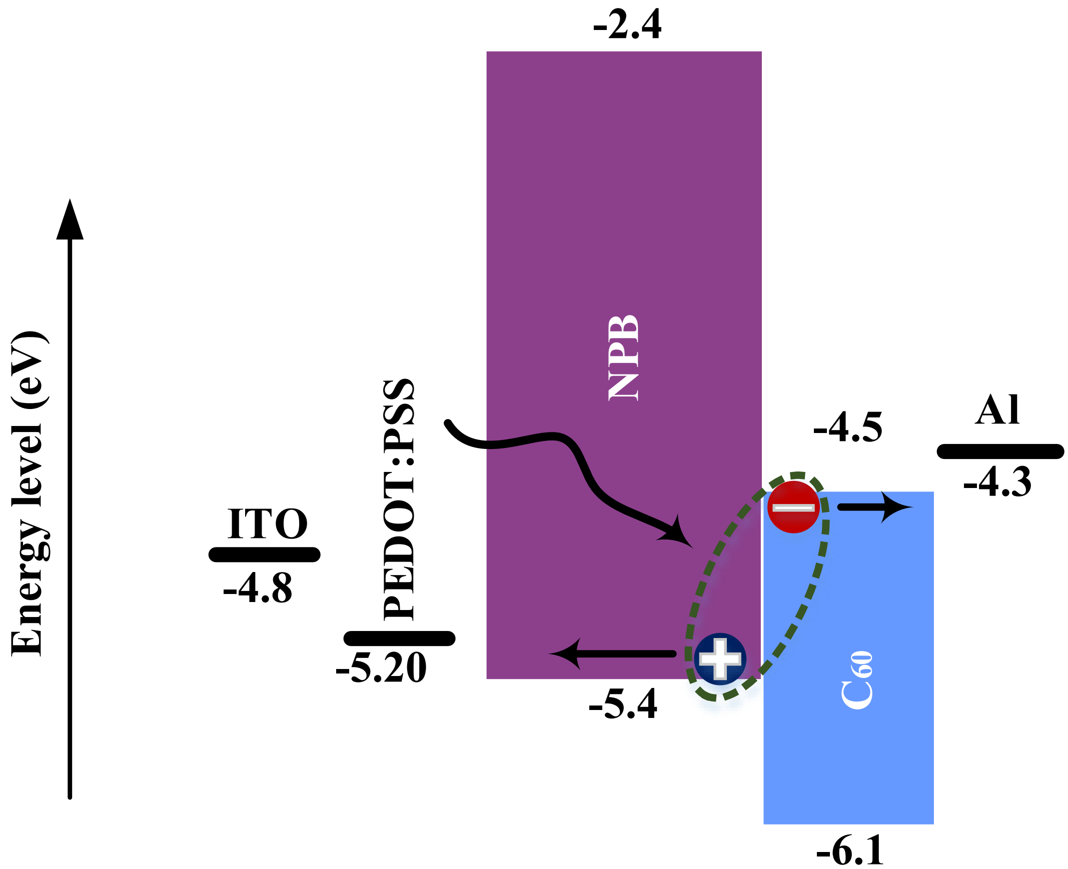
Lin Luan,[1] Kai Wang,[2]* and Bin Hu[1,2,3]*

1. Wuhan National Laboratory for Optoelectronics and School of Optical and Electronic Information, Huazhong University of Science and Technology, Wuhan 430074, China

2. Key Laboratory of Luminescence and Optical Information, Ministry of Education, Beijing Jiaotong University, Beijing 100044, China

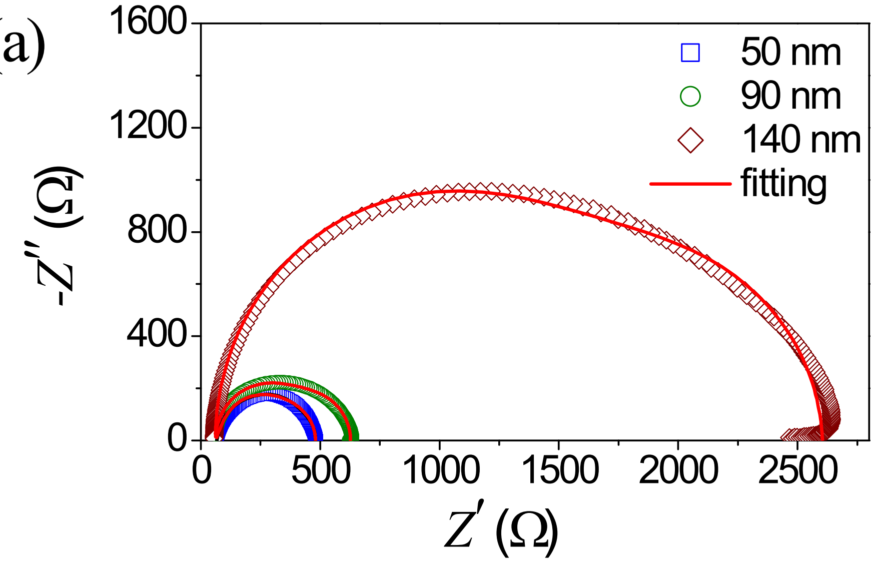
3. Department of Materials Science and Engineering, University of Tennessee, Knoxville, Tennessee 37996, USA

* Corresponding authors: Kai Wang (Dr.) (email: kaiwang@bjtu.edu.cn); Bin Hu (Prof.) (email: bhu@utk.edu)

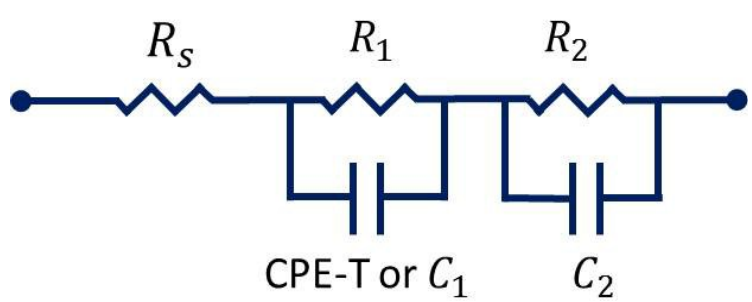



**S-Fig. 1** The schematic picture illustrates the energy alignment of the materials for the device configuration (ITO(glass)/PEDOT:PSS(30 nm)/NPB(50 nm, 90 nm, 140 nm)/$C_{60}$(30 nm)/Al(100 nm)). The electron-hole pair was created under photo-excitation and further diffuse to the two separated electrodes.

(a)




(b)




**S-Fig. 2** (a) The cole-cole (Nyquist) plots of the devices (ITO(glass)/PEDOT:PSS(30 nm)/NPB(50 nm, 90 nm, 140 nm)/$C_{60}$(30 nm)/Al(100 nm)) measured under 1000 mW/cm$^2$ illumination with the absence of the magnetic field, the red lines are the fitting curves. (b) The equivalent electronic circuit for the devices. $R_s$ represents the series resistance. $R_1$ and $C_1$ are the corresponding resistance and capacitance measured at lower *ac*-modulation frequencies due to the NPB surface resistance and capacitance. As we can see from S-Table 1, the decrease of the NPB layer thickness from 140 nm to 50 nm leads to the reduction of $C_1$. $R_2$ and $C_2$ are the corresponding resistance and capacitance measured at higher frequencies due to the bulk resistance/geometric capacitance $(C_{geo})$. Apparently, as it is indicated in S-Table 1, the increase of the NPB layer thickness leads to the decrease of $C_2$ because $C_2 = C_{geo} = \varepsilon_r \varepsilon_0 \frac{A}{d}$. CPE stands for constant phase element, and it contains two parts CPE-T and CPE-P. Conventionally, the semi-circle of the impedance spectrum can be modeled by a simple *RC* component. However, the lower frequency part may require a constant phase element (*CPE*) under a certain circumstance and it contains two major parts, a pseudo-capacitance (*CPE-T*) represented by *Q*, and a semi-circle depression element (*CPE-P*) denoted by *n*. The *CPE* can thus be written as $CPE = R^{(\frac{1-n}{n})} \cdot Q^{(\frac{1}{n})}$, in which $R$ represents resistance. If *n* approaches 1, the *CPE* turns theoretically to a capacitor.

**S-Tab. 1** The fitting parameters for the equivalent circuit of S-Figure 2(b).

| NPB Thickness | $R_s$ | $R_1$ | CPE-T or $C_1$ | CPE-P | $R_2$ | $C_2$ |
|---|---|---|---|---|---|---|
| 140 nm | 60.26 | 1005 | $2.03 \times 10^{-8}$ | 0.968 | 1509 | $1.78 \times 10^{-9}$ |
| 90 nm | 60.85 | 224 | $1.88 \times 10^{-8}$ | - | 342.3 | $2.49 \times 10^{-9}$ |
| 50 nm | 65.40 | 15.4 | $8.10 \times 10^{-9}$ | - | 399 | $4.50 \times 10^{-9}$ |