Supporting Information

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

Lin Luan,¹ Kai Wang,²* and Bin Hu¹,²,³*

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.
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/cm² 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

\[
CPE = R \left( \frac{1}{n} \right) \cdot Q^n,
\]

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).

<table>
<thead>
<tr>
<th>NPB Thickness</th>
<th>(R_s)</th>
<th>(R_1)</th>
<th>CPE-T or (C_1)</th>
<th>CPE-P</th>
<th>(R_2)</th>
<th>(C_2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>140 nm</td>
<td>60.26</td>
<td>1005</td>
<td>(2.03 \times 10^{-8})</td>
<td>0.968</td>
<td>1509</td>
<td>(1.78 \times 10^{-9})</td>
</tr>
<tr>
<td>90 nm</td>
<td>60.85</td>
<td>224</td>
<td>(1.88 \times 10^{-8})</td>
<td>-</td>
<td>342.3</td>
<td>(2.49 \times 10^{-9})</td>
</tr>
<tr>
<td>50 nm</td>
<td>65.40</td>
<td>15.4</td>
<td>(8.10 \times 10^{-9})</td>
<td>-</td>
<td>399</td>
<td>(4.50 \times 10^{-9})</td>
</tr>
</tbody>
</table>