

# Fe<sub>3</sub>O<sub>4</sub> based hole transport bilayer for efficient and stable perovskite solar cells

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## Supplementary Information

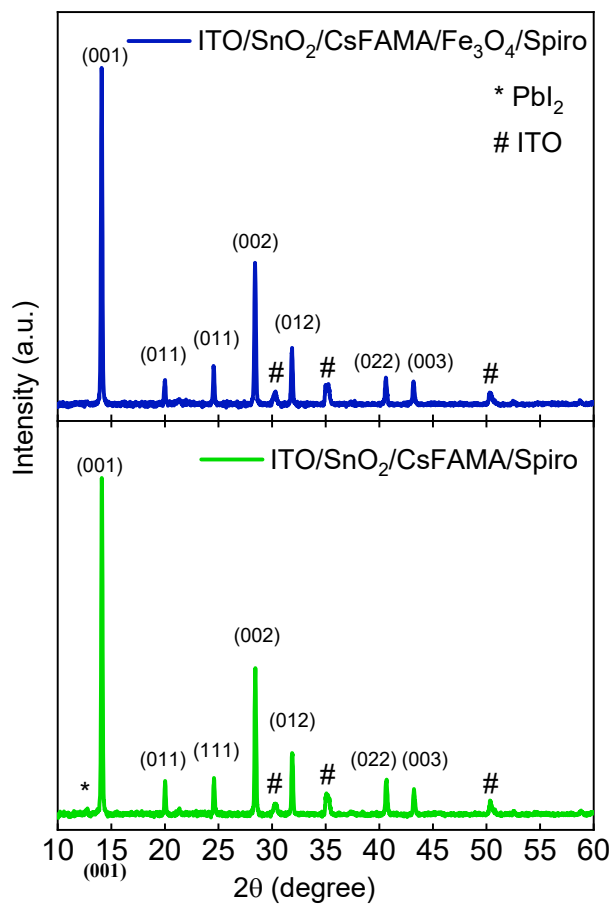


Figure S1. XRD diffractogram of ITO/SnO<sub>2</sub>/CsFAMA/Fe<sub>3</sub>O<sub>4</sub>/Spiro and ITO/SnO<sub>2</sub>/CsFAMA/Spiro thin films.

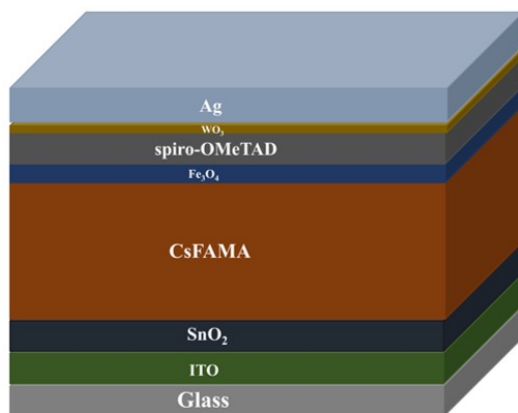
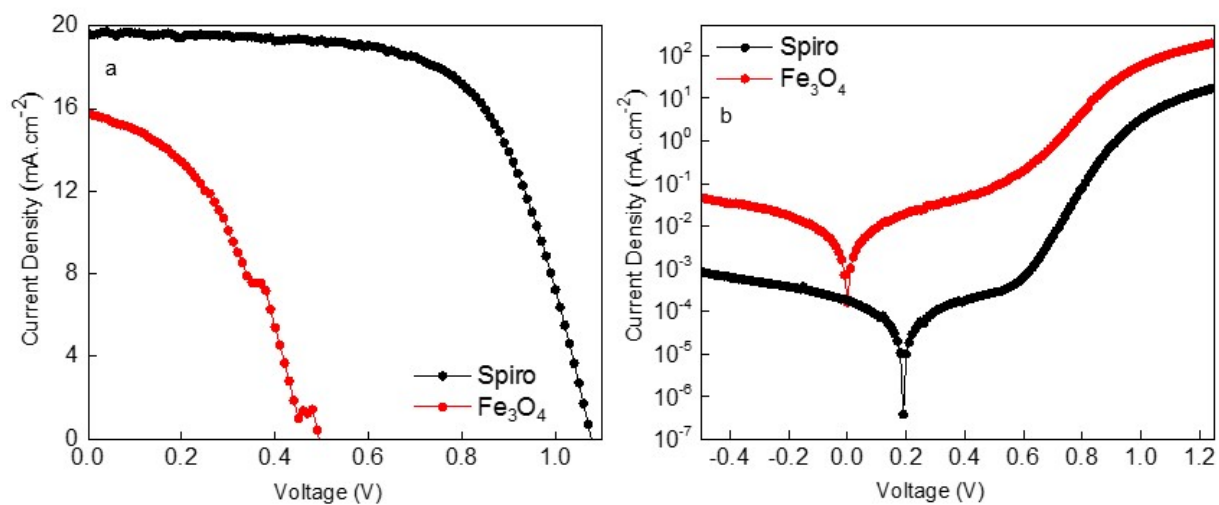
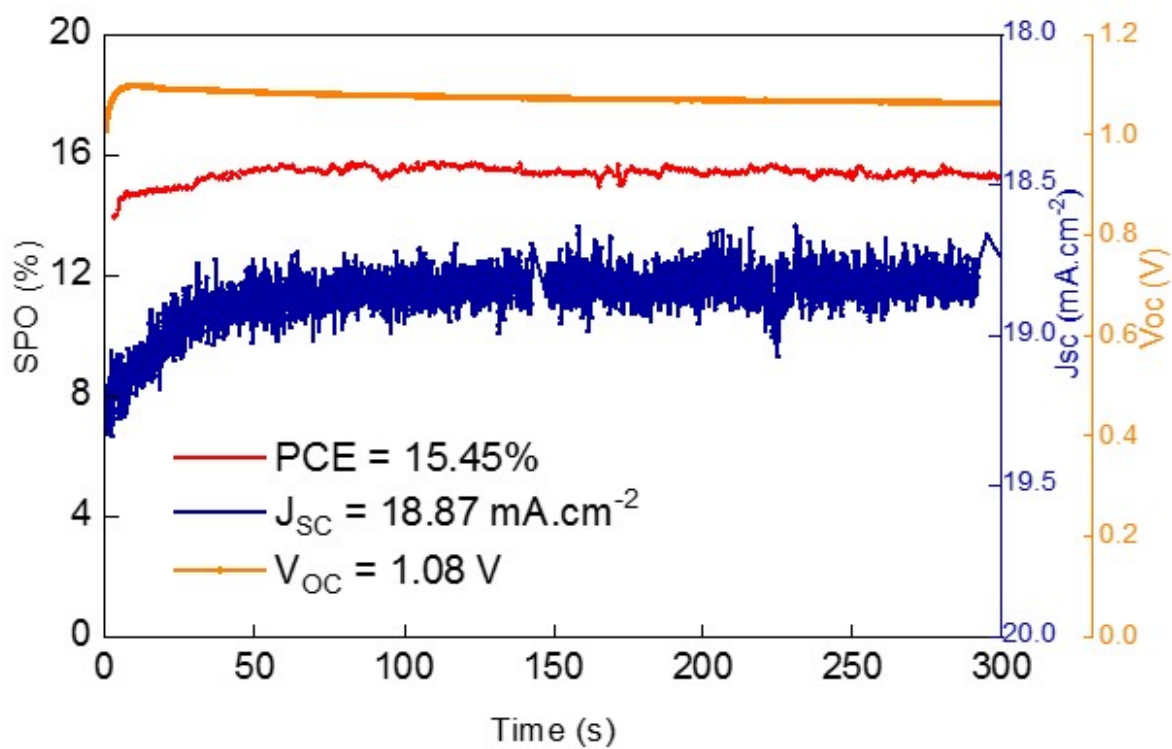


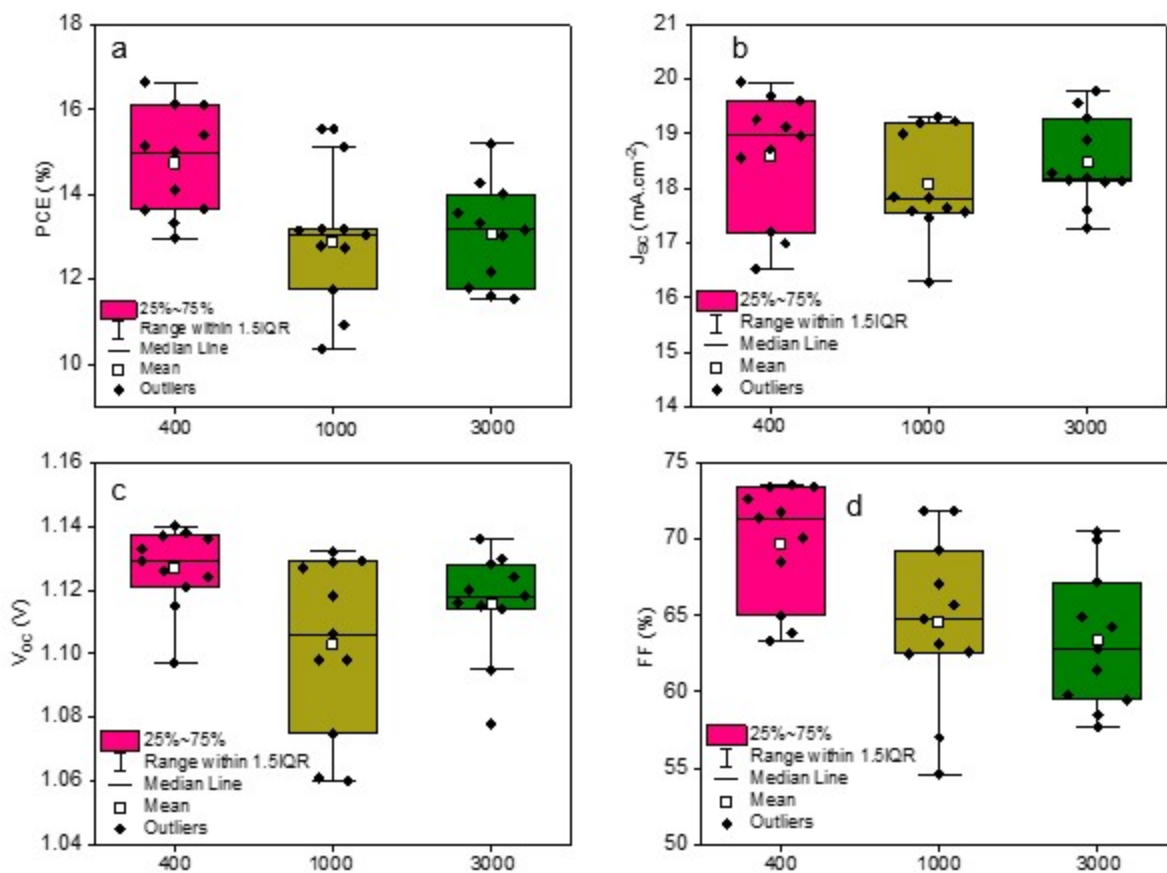
Figure S2. Device architecture of the PSCs in this work



**Figure S3.** (a) Light J–V curves of the perovskite solar cells with Fe<sub>3</sub>O<sub>4</sub> and spiro-OMeTAD hole transport layers, and (b) Dark J–V curves of the perovskite solar cells with Fe<sub>3</sub>O<sub>4</sub> and spiro-OMeTAD hole transport layers

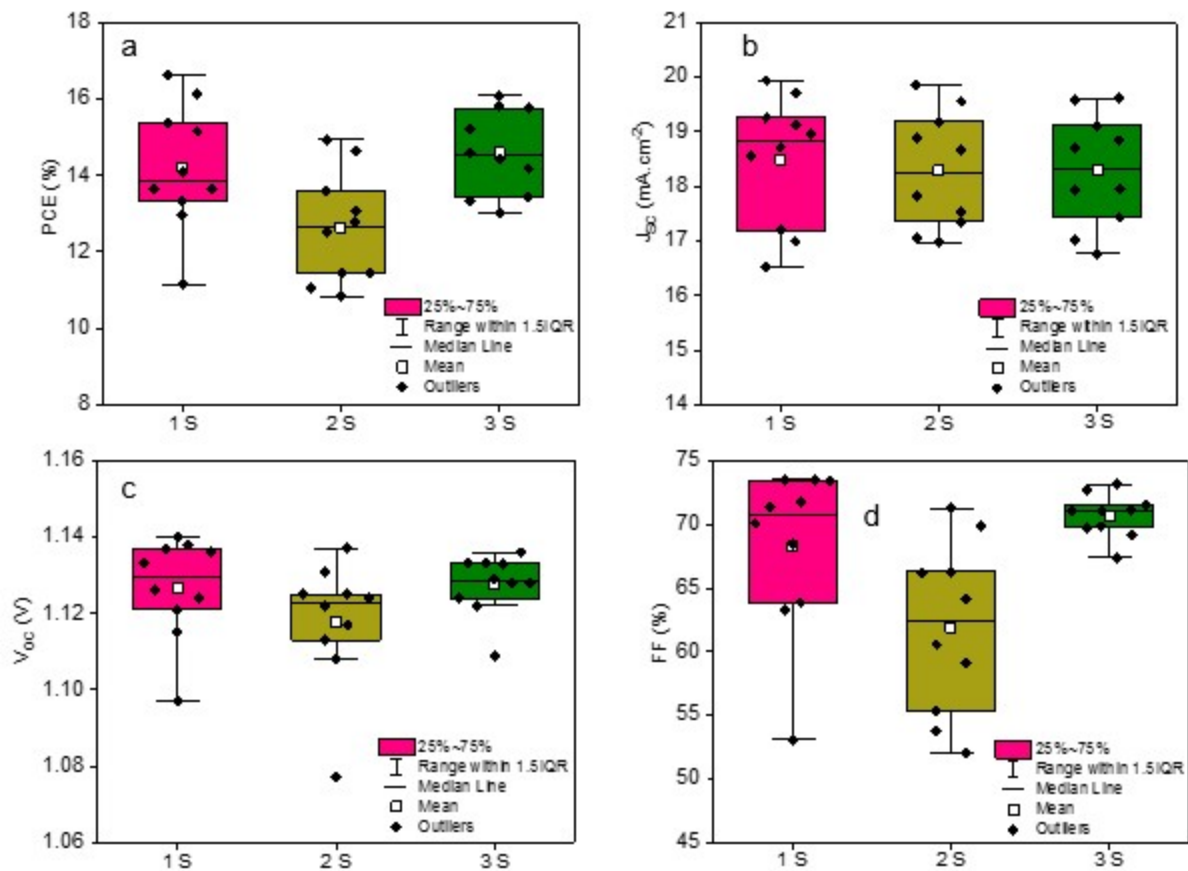


**Figure S4.** Steady-state current, voltage and stabilized power output at a maximum power point (0.65 V) of the champion device.

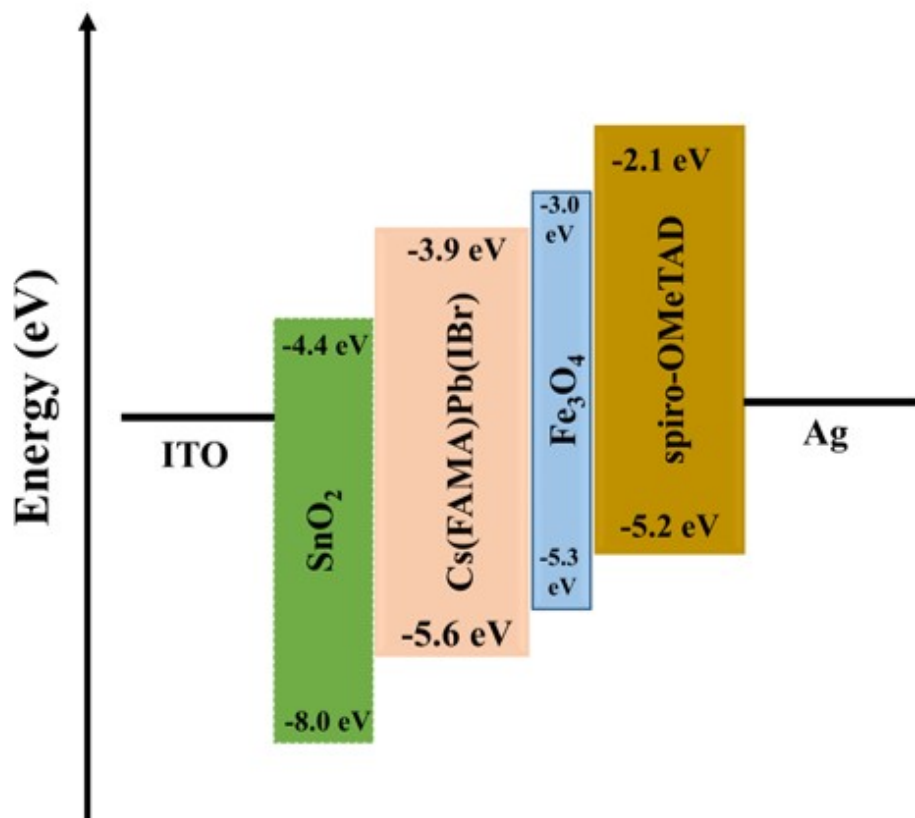


**Figure S5.** Impact of spinning acceleration on photovoltaic performance of ITO/SnO<sub>2</sub>/CsFAMA/Fe<sub>3</sub>O<sub>4</sub>/spiro-OMeTAD/WO<sub>3</sub>/Ag devices (Spin rpm was kept constant @3000rpm for Fe<sub>3</sub>O<sub>4</sub> bilayer HTL)

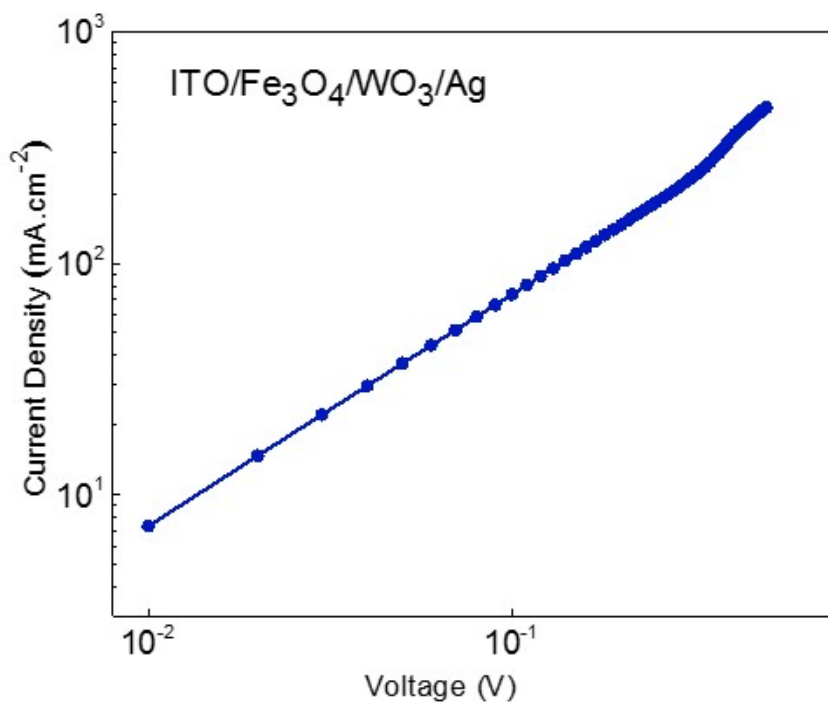
In addition, to optimize the spin parameters, we fabricated several devices for the modified HTL based on spinning acceleration and the number of spinning steps (one step, two steps, and three steps). The statistical results for the optimization processes are also displayed in Figure S5 and Figure S6 in supporting information. The 400 rpm.s<sup>-1</sup> spinning acceleration at 3000 rpm for 30 s and three-step spin coating showed much better performance mainly due to better surface coverage of the thin layer of magnetite nanoparticles onto the perovskite layer.



**Figure S6.** Impact of spin coating steps on photovoltaic performance of ITO/SnO<sub>2</sub>/CsFAMA/Fe<sub>3</sub>O<sub>4</sub>/spiro-OMeTAD/WO<sub>3</sub>/Ag devices (Spin rpm was kept constant @3000rpm with spinning acceleration @400 rpm.s<sup>-1</sup> for Fe<sub>3</sub>O<sub>4</sub> bilayer HTL)



**Figure S7.** Energy level diagram of the same. The values for the different energy levels shown in the Figure are taken from literature (The figure is drawn free of scale).



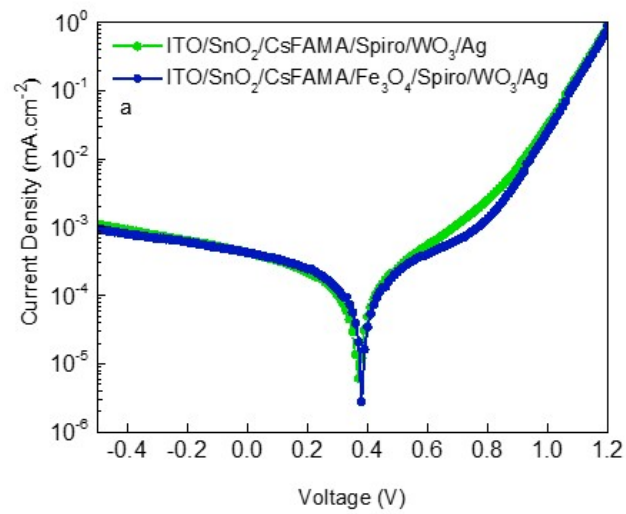
**Figure S8.** SCLC measurement of hole only device with configuration ITO/ Fe<sub>3</sub>O<sub>4</sub>/WO<sub>3</sub>/Ag.

**Table S1.** Photovoltaic parameters of perovskite devices with Fe<sub>3</sub>O<sub>4</sub> and spiro-OMeTAD HTLs.

| <b>Configuration</b>  | <b>J<sub>sc</sub></b><br><b>(mA cm<sup>-2</sup>)</b> | <b>V<sub>oc</sub></b><br><b>(V)</b> | <b>FF</b><br><b>(%)</b> | <b>PCE</b><br><b>(%)</b> |
|---|--|-------------------------------------|-------------------------|--------------------------|
| ITO/SnO <sub>2</sub> /CsFAMA/Spiro/Ag                           | 18.64  | 1.08                                | 68.60                   | 13.81                    |
| ITO/SnO <sub>2</sub> /CsFAMA/Fe <sub>3</sub> O <sub>4</sub> /Ag | 15.70  | 0.49                                | 40.17                   | 3.11                     |

**Table S2.** Fast and slow components for the PL decay and their corresponding proportions.

| <b>Description (ITO/SnO<sub>2</sub>)</b>     | <b>τ<sub>1</sub> (ns)</b> | <b>A<sub>1</sub> (%)</b> | <b>τ<sub>2</sub> (ns)</b> | <b>A<sub>2</sub> (%)</b> | <b>τ<sub>ave</sub> (ns)</b> |
|--|---------------------------|--------------------------|---------------------------|--------------------------|-----------------------------|
| CsFAMA                                       | 6.85                      | 49.38                    | 142                       | 50.62                    | 135.93                      |
| CsFAMA/Fe <sub>3</sub> O <sub>4</sub>        | 9.73                      | 33.31                    | 66.09                     | 66.69                    | 62.23                       |
| CsFAMA/Spiro                                 | 1.55                      | 92.17                    | 14.59                     | 7.83                     | 7.34                        |
| CsFAMA/Fe <sub>3</sub> O <sub>4</sub> /Spiro | 1.03                      | 98.55                    | 31.21                     | 1.45                     | 10.51                       |



**Figure S9.** Dark J-V measurements of reference and target perovskite devices.