Supporting Information

Extremely thin and robust interconnecting layer providing 76% Fill Factor in a tandem polymer solar cell architecture

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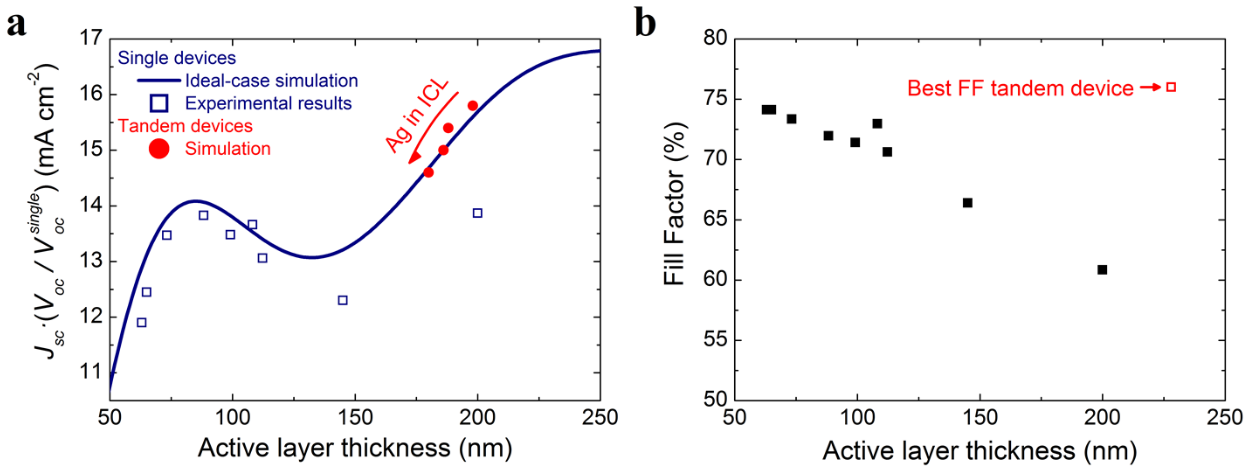


Figure S1. a) $J_{sc}{\cdot}V_{oc}$ as a function of the active layer thickness for PTB7:PC_{71}BM-based cells normalized to the single-junction V_{oc} under the assumption that $V_{oc}^{tandem} = 2\,V_{oc}^{single}$. J_{sc} was computed using the transfer matrix formalism (see Refs. 23 and 24 on the main text) to obtain the optimal single-junction device values (blue solid line) and the optimal values for tandem devices with both sub-cells featuring the same PTB7:PC_{71}BM absorber (red solid circles). The latter are computed for four Ag nominal contents in the interconnecting layer (0.5, 1, 1.5, and 2 nm). The experimental J_{sc} for single-junction devices is shown with blue empty squares. A deviation from the ideal case, mostly caused by the decrease in fill factor (FF) when increasing thickness (Figure S1(b)), is clearly seen beyond 100 nm. b) Experimentally measured FF for different active layer thicknesses for PTB7:PC_{71}BM-based single-junction devices (black solid dots) and best value for tandem devices (red hollow square). For all tandem devices in the figures the thickness corresponds to the total active layer thickness.

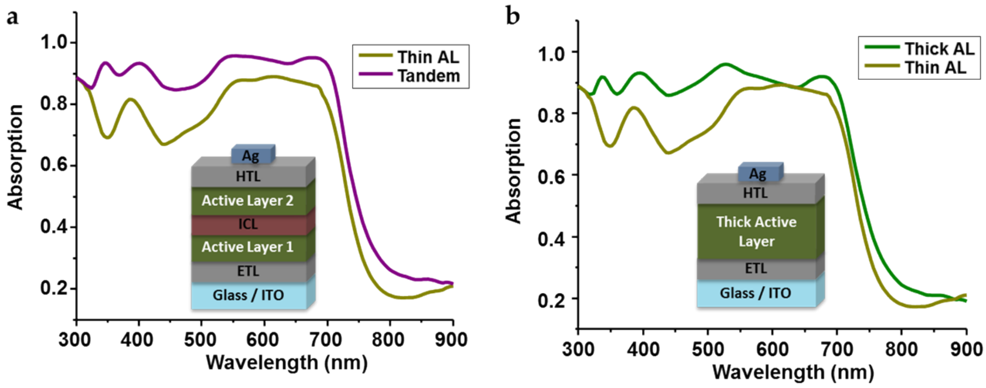


Figure S2: Having tandem solar cells with two subcells based on the same material combination has proven effective in overcoming the low absorption of the thin film PSCs. Here we show a calculated absorption comparison between a) a thin (100 nm) single junction PTB7 solar cell and a tandem solar cell with two layers of PTB7 (total polymer thickness 220 nm), and b) the same thin single junction PTB7 solar cell with a 220 nm single-junction PTB7 solar cell. Although the tandem device and the thick layer single junction device show similar absorption, the thick layer single junction solar cell will perform poorly because charge transport limitations (see also **Figure S1b**).

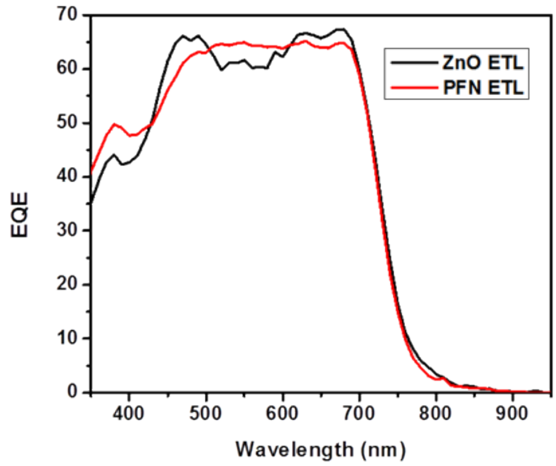


Figure S3: External quantum efficiency (EQE) spectra for an inverted device with 12 nm-thick PFN as ETL (red line) and for an inverted device with 35 nm-thick ZnO as ETL (black line) showing a calculated J_{sc} of 14.0 mA/cm^2 and 14.1 mA/cm^2, respectively. The experimental value in both cases was 14.3 mA/cm^2.

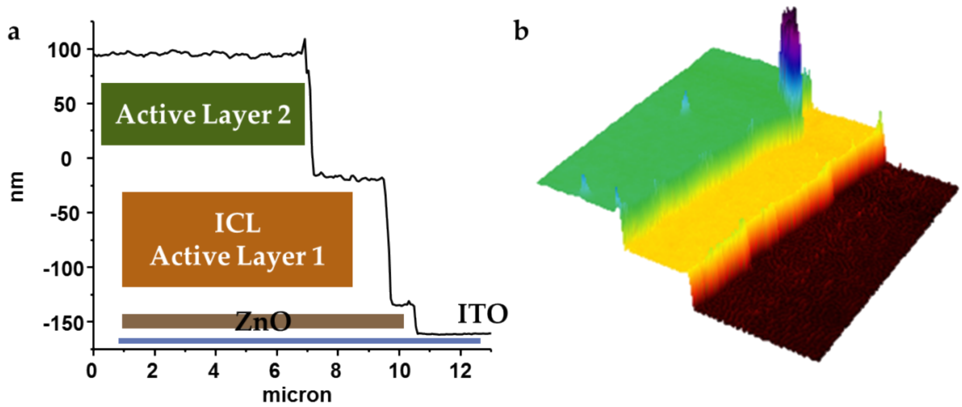
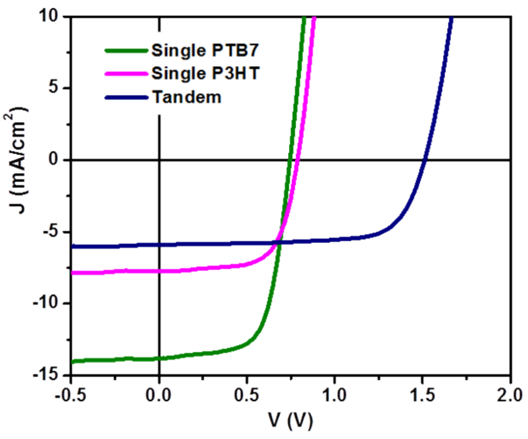


Figure S4: a) AFM profile of a tandem solar cell after scratching. The thicknesses of the ZnO layer, active layer 1 plus ICL and active layer 2 can be easily measured due to the formation of steps. b) 3D AFM image of a scratched area where different layers can be seen.



	Jsc [mA/cm2]	Voc [mV]	FF [%]	Eta [%]
Single PTB7	13.8	744	65.7	6.8
Single P3HT	7.7	788	66.1	4.0
Tandem	5.9	1511	71.5	6.4

Figure S5: *J-V* plot and parameters of single junction devices with active layers of P3HT:ICBA and PTB7:$PC_{71}BM$ and tandem solar cell with configuration glass/ITO/ZnO/P3HT:ICBA/MoO_3/Ag/PFN/PTB7:$PC_{71}BM$/MoO_3/Ag measured under AM1.5 100 mW cm^{-2} illumination.

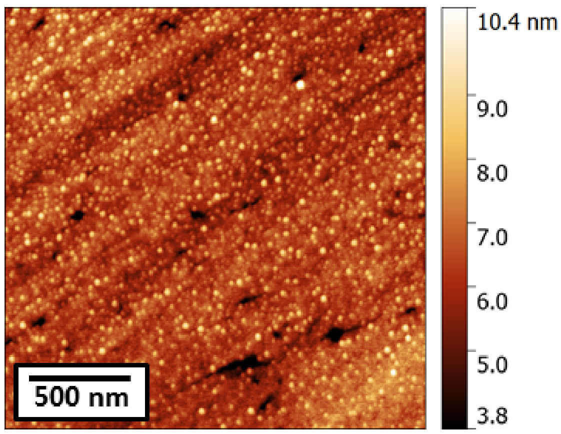


Figure S6. AFM image of a glass/ITO/MoO_3/Ag(0.5 nm)/PFN(10 nm) sample. RMS roughness is 0.8 nm.

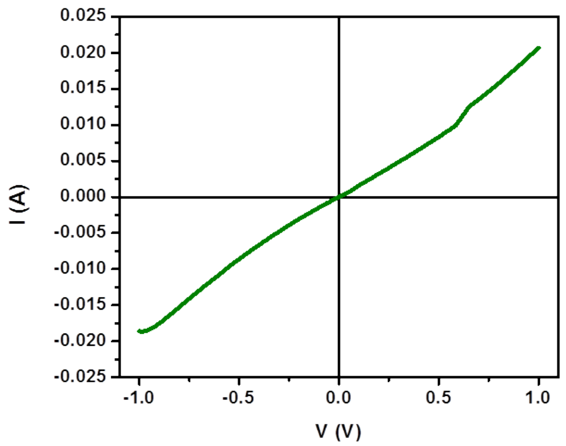


Figure S7: *J-V* characteristic of the device with the following structure: ITO/MoO_3/Ag/PFN/Ag under AM1.5 100 mW cm^{-2} illumination.

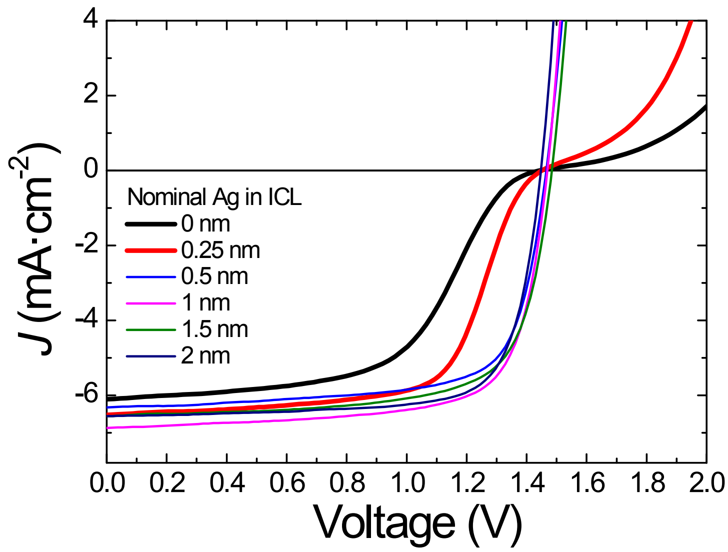


Figure S8. *J-V* curves for tandem devices with varying nominal Ag content in the ICL as indicated.