## Supporting Information

# 2D/1A Strategy to Regulate Film Morphology for Efficient and Stable Nonfullerene Organic Solar Cells 

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Figure S1. Optical absorption of binary and ternary blend films. Symbols: $\mathbf{D}_{\mathbf{1}}=$ PTB7-Th, $\mathbf{D}_{\mathbf{2}}=\operatorname{PffBT} 4 \mathrm{~T}-2 \mathrm{OD}$ and $\mathbf{A}=$ ITIC.


Figure S2. Optical absorption spectra of PTB7-Th, PffBT4T-2OD and ITIC neat films.


Figure S3. Photoluminescence spectra of binary and ternary blend films in comparison to neat films. Symbols: $\mathbf{D}_{\mathbf{1}}=$ PTB7-Th, $\mathbf{D}_{\mathbf{2}}=$ PffBT4T-2OD and $\mathbf{A}=$ ITIC.


Figure S4. GIWAXS patterns of binary and ternary blend films. Symbols: $\mathbf{D}_{\mathbf{1}}=$ PTB7-Th, $\mathbf{D}_{\mathbf{2}}=\operatorname{PffBT} 4 \mathrm{~T}-2 \mathrm{OD}$ and $\mathbf{A}=$ ITIC.


Figure S5. GIWAXS line-cut profiles of binary and ternary blend films. Note that $\mathbf{D}_{\mathbf{1}}=$ PTB7-Th, $\mathbf{D}_{\mathbf{2}}=$ PffBT4T-2OD and $\mathbf{A}=$ ITIC.


Figure S6. GIWAXS patterns of PTB7-Th, PffBT4T-2OD and ITIC neat films.


Figure S7. TEM images of binary and ternary blend thin films. Symbols: $\mathbf{D}_{\mathbf{1}}=$ PTB7 $-\mathrm{Th}, \mathbf{D}_{\mathbf{2}}$ $=$ PffBT4T-2OD and $\mathbf{A}=$ ITIC.


Figure S8. $J-V$ characteristics of binary and ternary blend OSCs. Symbols: $\mathbf{D}_{1}=$ PTB7-Th, $\mathbf{D}_{2}=\operatorname{PffBT} 4 T-2 \mathrm{DD}$ and $\mathbf{A}=$ ITIC .


Figure S9. EQE curves of binary and ternary blend OSCs. Symbols: $\mathbf{D}_{\mathbf{1}}=$ PTB7-Th, $\mathbf{D}_{\mathbf{2}}=$ PffBT4T-2OD and $\mathbf{A}=$ ITIC.


Figure S10. $J-V$ curves of ternary PTB7-Th:PffBT4T-2OD (80:20, w/w):ITIC blend based OSCs in $\mathrm{N}_{2}$ and air atmosphere, respectively.


Figure S11. Dark CELIV $j-t$ profiles of binary and ternary blend OSCs. Symbols: $\mathbf{D}_{\mathbf{1}}=$ PTB7-Th, $\mathbf{D}_{\mathbf{2}}=$ PffBT4T-2OD and $\mathbf{A}=$ ITIC.


Figure S12. Light intensity dependence on $J_{\mathrm{SC}}$ of binary and ternary blend OSCs. Symbols: $\mathbf{D}_{1}=$ PTB7-Th, $\mathbf{D}_{2}=$ PffBT4T-2OD and $\mathbf{A}=$ ITIC.

Table S1. Summary of photovoltaic parameters of PTB7-Th:PffBT4T-2OD:ITIC ternary devices under simulated light irradiation of $100 \mathrm{~mW} \mathrm{~cm}^{-2}$

| $\begin{gathered} \mathbf{D}_{1}: \mathbf{D}_{2}: \mathbf{A}^{\mathrm{a})} \\ (\mathrm{w} / \mathrm{w} / \mathrm{w}) \end{gathered}$ | $\begin{gathered} \boldsymbol{J}_{\mathbf{S C}} \\ \left(\mathrm{mA} / \mathrm{cm}^{2}\right) \end{gathered}$ | $V_{\text {OC }}$ <br> (V) | $\mathbf{F F}$ (\%) | PCE <br> (\%) | Best PCE <br> (\%) |
| :---: | :---: | :---: | :---: | :---: | :---: |
| 1:0:1.5 | $15.06 \pm 0.13$ | $0.82 \pm 0.01$ | $51.21 \pm 0.78$ | $6.35 \pm 0.13$ | 6.48 |
| 0.9:0.1:1.5 | $15.62 \pm 0.21$ | $0.83 \pm 0.01$ | $56.90 \pm 0.67$ | $7.37 \pm 0.19$ | 7.56 |
| 0.8:0.2:1.5 | $15.36 \pm 0.16$ | $0.84 \pm 0.01$ | $62.62 \pm 0.60$ | $8.05 \pm 0.17$ | 8.22 |
| 0.7:0.3:1.5 | $11.77 \pm 0.15$ | $0.85 \pm 0.01$ | $56.48 \pm 0.63$ | $5.66 \pm 0.16$ | 5.82 |
| 0.6:0.4:1.5 | $10.76 \pm 0.09$ | $0.86 \pm 0.01$ | $55.59 \pm 0.83$ | $5.12 \pm 0.13$ | 5.25 |
| 0:1:1.5 | $8.91 \pm 0.10$ | $0.88 \pm 0.01$ | $56.25 \pm 0.53$ | $4.42 \pm 0.09$ | 4.51 |

