Supporting Information

Synergistic Optimization of Mechanical and Photovoltaic Properties in Ternary Organic Solar Cells from a Two-donor Polymer Blend

Mingfei Li[1], Zhongxiang Peng[1], Kaihu Xian[1], Wenchao Zhao[2], Chenxu Liu[3], Yu Chen[3,], Chaohua Cui[4], Long Ye[1, 5*]*

1. School of Materials Science and Engineering, Tianjin Key Laboratory of Molecular Optoelectronic Sciences, Tianjin University, and Collaborative Innovation Center of Chemical Science and Engineering (Tianjin), Tianjin, 300350, China

2. Co-Innovation Center of Efficient Processing and Utilization of Forest Resources, College of Materials Science and Engineering, Nanjing Forestry University, Nanjing 210037, China

3. Beijing Synchrotron Radiation Laboratory, Institute of High Energy Physics, Chinese Academy of Sciences, Beijing 100049, China

4. Laboratory of Advanced Optoelectronic Materials, Suzhou Key Laboratory of Novel Semiconductor-Optoelectronics Materials and Devices, College of Chemistry, Chemical Engineering and Materials Science, Soochow University, Suzhou, 215123, China

5. Hubei Longzhong Laboratory, Xiangyang 441000, China

Corresponding authors: yelong@tju.edu.cn; chen_yu@ihep.ac.cn

Instruments and Measurements

Device Fabrication. The conventional device structure of ITO/ PEDOT:PSS/ active layer/ PNDIT-F3N/ Ag was adopted in this study. The optimal D/A ratio was kept constant at 1:1.2 (wt/wt), in which the donor concentration is 7 mg/ml. The blend solutions were stirred at 60 °C for 2h to fully dissolve. Prior to spin-coating the active layer solutions, 0.5% CN (v/v) was added to the CF. PNDIT-F3N was dissolved in methanol at a concentration of 0.5 mg/mL. The mixed solutions were spin-coated onto the PEDOT:PSS layers, and the thicknesses of all active layers were about 100 nm. Then the films were treated with thermal annealing at 100 °C for 10 min. PNDIT-F3N

was spin-coated on the top of the active layers. Finally, 100 nm thick Ag was deposited on the top of PNDIT-F3N layer under high vacuum.

Optical Absorption. Absorption spectra of all the materials in the solid thin films were measured on Shimadzu UV3600 plus spectrometer. Thin films were prepared by spin coating from the chloroform solutions.

Photovoltaic Properties. The *J-V* measurements were performed via the AAA solar simulator (SS-F5-3A, Enli Technology Co. Ltd, Taiwan) along with AM 1.5G spectra whose intensity was calibrated by the certified standard silicon solar cell at 100 mW/cm^2. The EQE spectra were measured through the Solar Cell Spectral Response Measurement System QE-R3011 (Enli Technology Co. Ltd, Taiwan). The thickness of the blend layers was measured via the surface profilometer Bruker Dektak XT.

Morphology Characterizations. The surface morphology of films was measured by a Nanoscope V AFM (Bruker Multimode 8) in tapping mode. The type of AFM cantilever is RTESPA-300, which possesses a *k* constant of about 40 N/m. The scanning area was 2 μm × 2 μm and the resolution is 256×256 pixels.
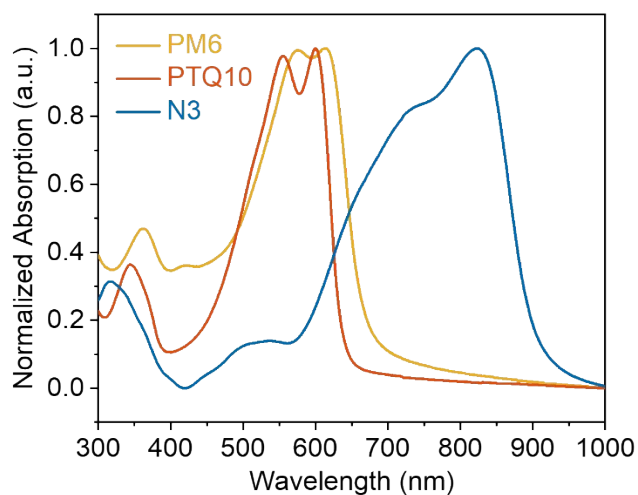
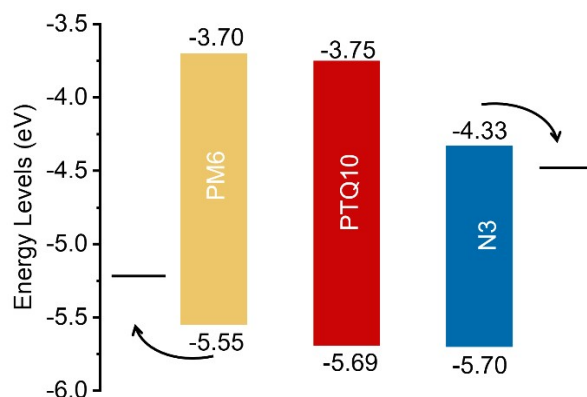**Figure S1.** The UV-vis absorption spectra of the neat films.

Figure S2. The energy level diagram of three neat materials.

Table S1. The peak position and *CL* in IP directions of neat PM6 and PTQ10.

Sample	Position ($Å^{-1}$)	Stacking Distance (Å)	FWHM ($Å^{-1}$)	*CL* (Å)[a]
PM6	0.34	18.57	0.199	28.4
PTQ10	0.28	22.74	0.116	48.4

[a] Coherence lengths (*CL*) were calculated using the Scherrer equation of diffraction peaks: $CL = 2\pi k/\Delta q$, where k is the dimensionless shape factor (herein $k = 0.9$) and Δq was the full width at half maximum (FWHM) of the given peak.

Table S2. The peak position and CL in OOP directions of neat PM6 and PTQ10.

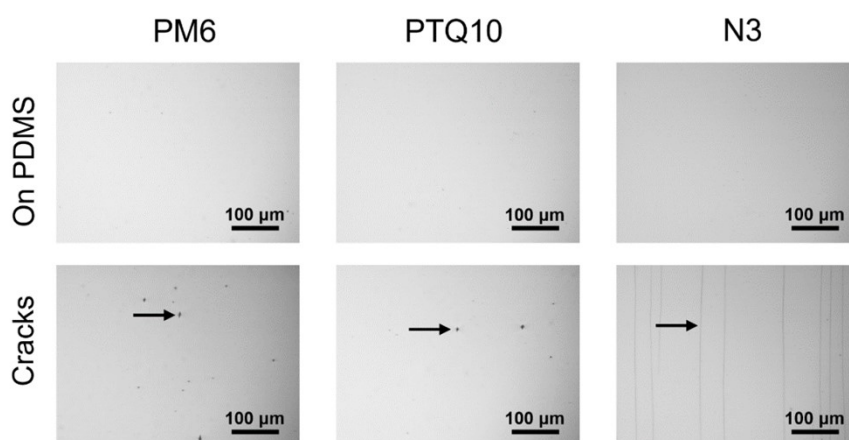| Sample	Position (Å^{-1})	Stacking Distance (Å)	FWHM (Å^{-1})	*CL* (Å)
PM6	1.69	3.72	0.356	15.9
PTQ10	1.78	3.53	0.258	21.9

Figure S3. Optical micrographs of PM6, PTQ10, and N3 neat films were on PDMS by transferred from glass. The cracks on the films are highlighted by black arrows.
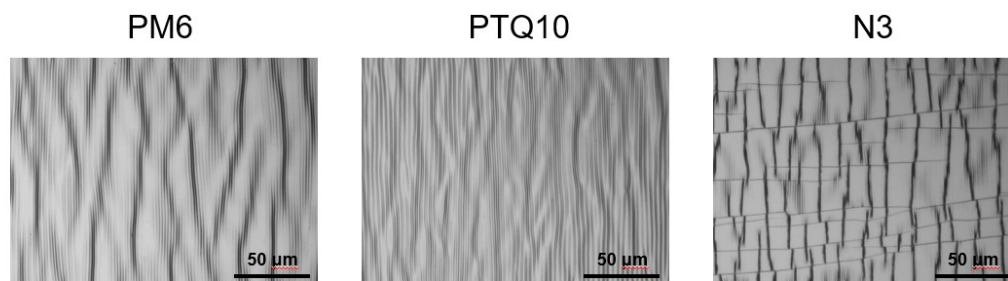

Figure S4. Optical micrographs of PM6, N3, and P(NDI2OD-T2) neat films under 5% strain compression.

Table S3. The relative degree of crystallinity (*rDoC*) values of (010) π–π stacking peaks of PM6 and PTQ10.

Neat Films	PM6	PTQ10
rDoC	0.84	1

Table S4. The crack-onset strain and elastic modulus of PM6, PTQ10, and N3 neat films obtained by the FOE method.

Neat Films	PM6	PTQ10	N3
COS (%)	24.2±1.3	25.2±1.7	1.5±0.3
E (GPa)	1.05±0.10	0.71±0.07	1.49±0.16

Table S5. The parameters extracted from the PSD profiles of PM6:PTQ10:N3 blend films with varied PTQ10 contents.

PTQ10 in Donors	0%	20%	40%	60%	80%	100%
q (nm^{-1})	0.243	0.211	0.186	0.221	0.224	0.236
d (nm)	25.8	29.8	33.8	28.4	28.0	26.6

Table S6. The peak position and CL in OOP directions of PM6:N3 blend film.

(010) peak	Position (Å^{-1})	Stacking distance (Å)	FWHM (Å^{-1})	*CL* (Å)

(010) peak of PM6	1.70	3.69	0.27	20.9
(010) peak of N3	1.80	3.49	0.24	23.3

Table S7. The peak position and CL in OOP directions of PM6:PTQ10:N3 blend film.

(010) peak	position(Å^{-1})	Stacking distance (Å)	FWHM (Å^{-1})	*CL* (Å)
(010) peak of PM6	1.69	3.71	0.20	28.1
(010) peak of PTQ10 and N3	1.80	3.49	0.21	27.3

Table S8. The peak position and CL in OOP directions of PTQ10:N3 blend film.

Sample	Position (Å^{-1})	Stacking Distance (Å)	FWHM (Å^{-1})	*CL* (Å)
PTQ10:N3	1.79	3.49	0.25	22.7

Table S9. The *rDoC* values of (100) lamellar stacking peaks and (010) π–π stacking peaks of three blend films.

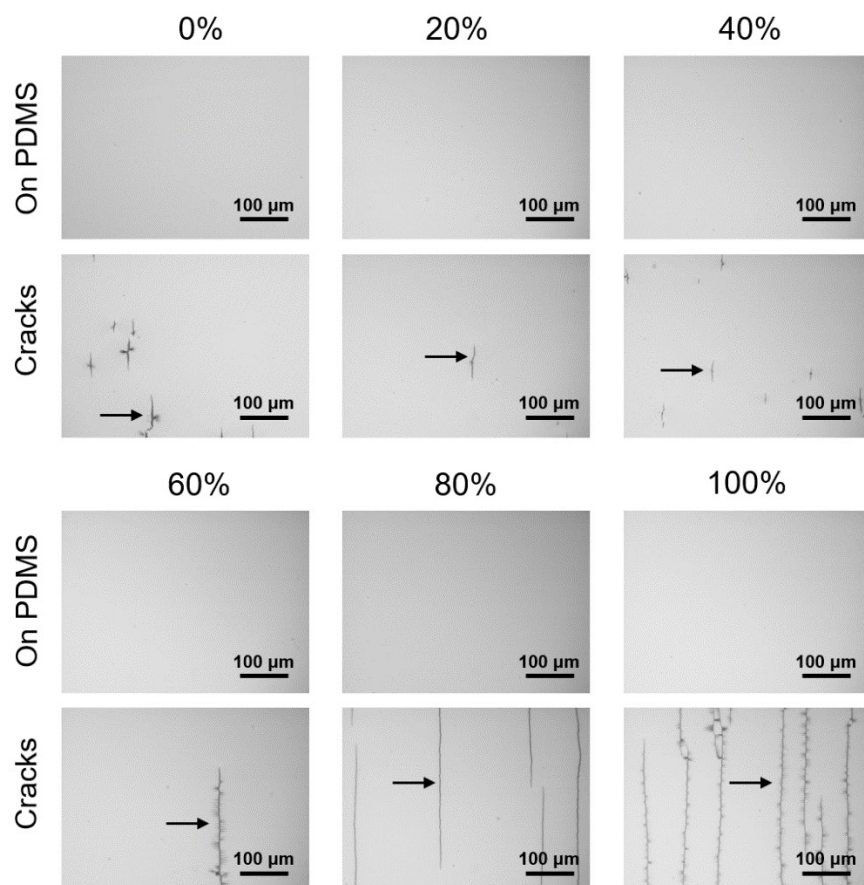| Sample	PM6:N3	PM6:PTQ10:N3	PTQ10:N3
rDoC(100)	0.76	1	0.80
rDoC(010)	0.78	1	0.82

Figure S5. Optical micrographs of PM6:PTQ10:N3 blend films with various PTQ10 contents were on PDMS transferred from glass. The cracks in the films are highlighted by black arrows.
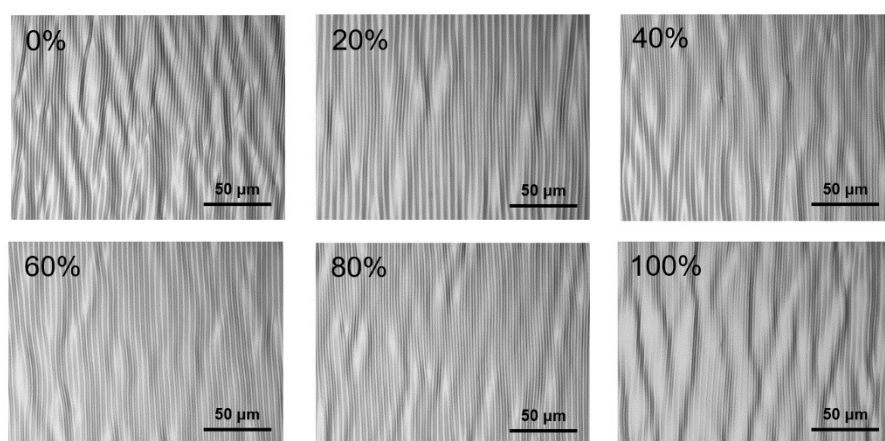

Figure S6. Optical micrographs of PM6:PTQ10:N3 blend films with different PTQ10 contents under 5% strain compression.
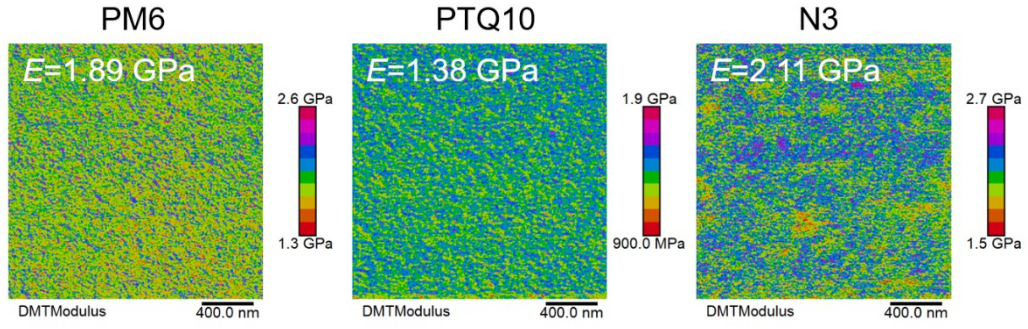
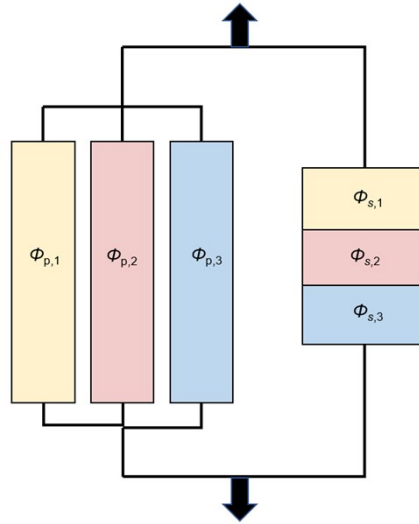**Figure S7.** Representative modulus images of the neat films obtained by the PFQNM method.

Figure S8. The schematic of equivalent box model (EBM) for three-component blends.
$\Phi_p=\Phi_{p,1}+\Phi_{p,2}+\Phi_{p,3}$, $\Phi_s=\Phi_{s,1}+\Phi_{s,2}+\Phi_{s,3}$