SHORT COMMUNICATION

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18.69% PCE from organic solar cells

Ke Jin, Zuo Xiao†, and Liming Ding†

Center for Excellence in Nanoscience (CAS), Key Laboratory of Nanosystem and Hierarchical Fabrication (CAS), National Center for Nanoscience and Technology, Beijing 100190, China

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SUPPORTING INFORMATION

1. Device fabrication and measurements

Conventional solar cells

A 30 nm thick PEDOT:PSS layer was made by spin-coating an aqueous dispersion onto ITO glass (4000 rpm for 30 s). PEDOT:PSS substrates were dried at 150 °C for 10 min. A blend solution of D18-Cl (M_n: 70.5 kDa, PDI: 2.27), N3 and $PC_{61}BM$ in chloroform (13 mg/mL) with 0.3 vol% DPE additive was spin-coated onto PEDOT:PSS layer. PDIN (2 mg/mL) in MeOH:AcOH (1000 : 3) was spin-coated onto active layer (5000 rpm for 30 s). Ag (~80 nm) was evaporated onto PDIN through a shadow mask (pressure ca. 10^{-4} Pa). The device area is 4 mm^2. The thicknesses of the active layers were measured by using a KLA Tencor D-120 profilometer. The illumination intensity of solar simulator was determined by using a monocrystalline silicon solar cell (Enli SRC2020, 2 × 2 cm^2) calibrated by the National Institute of Metrology (NIM). *J–V* curves were measured by using a computerized Keithley 2400 SourceMeter and a Xenon-lamp-based solar simulator (Enli Tech, AM 1.5G, 100 mW/cm^2). When doing *J–V* measurements, a metal mask with an aperture (2.56 mm^2) was used to define the effective area. The external quantum efficiency (EQE) spectra were measured by using a QE-R3011 measurement system (Enli Tech). The best cells were further tested at NIM for certification. A metal mask with an aperture (2.580 mm^2) was used to define the effective area.

Hole-only devices

The structure for hole-only devices is ITO/PEDOT:PSS/active layer/MoO_3/Al. A 30 nm thick PEDOT:PSS layer was made by spin-coating an aqueous dispersion onto ITO glass (4000 rpm for 30 s). PEDOT:PSS substrates were dried at 150 °C for 10 min. A D18-Cl:N3:$PC_{61}BM$ (or D18-Cl:N3) blend in CF was spin-coated onto PEDOT:PSS. Finally, MoO_3 (~6 nm) and Al (~100 nm) were successively evaporated onto the active layer through a shadow mask (pressure ca. 10^{-4} Pa). *J–V* curves were measured by using a computerized Keithley 2400 SourceMeter in the dark.

Electron-only devices

The structure for electron-only devices is ITO/ZnO/active layer/PDIN/Al. The ZnO precursor solution was spin-coated onto ITO glass (4000 rpm for 30 s). The films were annealed at 200 °C in air for 20 min. ZnO film thickness is ~30 nm. A D18-Cl:N3:$PC_{61}BM$ (or D18-Cl:N3) blend in CF was spin-coated onto ZnO. PDIN (2 mg/mL) in MeOH:AcOH (1000 : 3) was spin-coated onto active layer (5000 rpm for 30 s). Al (~100 nm) was evaporated onto the active layer through a shadow mask (pressure ca. 10^{-4} Pa). *J–V* curves were measured by using a computerized Keithley 2400 SourceMeter in the dark.

2. Optimization of device performance

Table S1. Optimization of D : A_1 : A_2 ratio for D18-Cl:N3:$PC_{61}BM$ solar cells[a].

D : A_1 : A_2 (w/w/w)	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	PCE (%)
1 : 1.4 : 0	0.847	27.56	76.5	17.85 (17.82)[b]
1 : 1.4 : 0.1	0.846	27.86	77.2	18.19 (18.14)
1 : 1.4 : 0.2	0.843	27.72	77.3	18.05 (17.90)
1 : 1.4 : 0.3	0.841	27.59	76.8	17.82 (17.71)

[a]Blend solution: 13 mg/mL in CF with 0.3 vol% DPE; spin-coating: 4000 rpm for 30 s.
[b]Data in parentheses stand for the average PCEs for 10 cells.

Table S2. Optimization of DPE content for D18-Cl:N3:$PC_{61}BM$ (1 : 1.4 : 0.1) solar cells[a].

DPE (vol%)	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	PCE (%)
0	0.853	26.83	77.6	17.75 (17.69)[b]
0.3	0.846	27.86	77.2	18.19 (18.14)
0.6	0.842	27.77	77.0	18.00 (17.84)
0.9	0.841	27.70	76.2	17.75 (17.70)

[a]Blend solution: 13 mg/mL in CF; spin-coating: 4000 rpm for 30 s.
[b]Data in parentheses stand for the average PCEs for 10 cells.

Correspondence to: Z Xiao, xiaoz@nanoctr.cn; L M Ding, ding@nanoctr.cn
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Table S3. Optimization of the active layer thickness for D18-Cl:N3:$PC_{61}BM$ (1 : 1.4 : 0.1) solar cells[a].

Thickness (nm)	V_{oc} (V)	J_{sc} (mA/cm^2)	FF (%)	PCE (%)
158	0.837	27.61	76.7	17.73 (17.51)[b]
130	0.846	27.86	77.2	18.19 (18.14)
114	0.849	28.22	78.0	18.69 (18.47)
103	0.847	27.45	78.3	18.21 (18.17)

[a]Blend solution: 13 mg/mL in CF with 0.3 vol% DPE.
[b]Data in parentheses stand for the average PCEs for 10 cells.

3. *J–V*

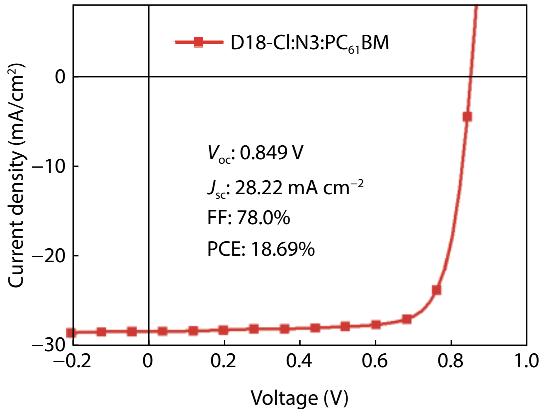

Fig. S1. The *J–V* curve for the best D18-Cl:N3:$PC_{61}BM$ cell.

4. EQE

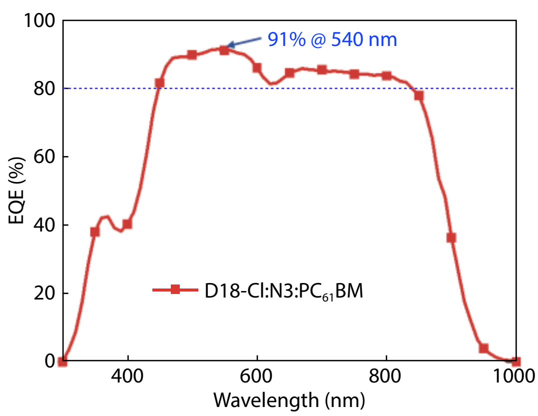

Fig. S2. The EQE spectrum for the best D18-Cl:N3:$PC_{61}BM$ cell.

5. NIM certification

Appendix: Summary of the Report
Report No.: GXgf2021-10527
Client: The National Center for Nanoscience and Technology
Sample: Organic Solar Cells
Type/Model: Organic Solar Cells
DUT S/N: 11-M1-F-1
Manufacturer: The National Center for Nanoscience and Technology
Date of Test: 03/18/2021
Temperature Sensor/Control System: None
Environmental conditions: (23.1±1) C°, RH (36.3±2) %
Mask: An aperture area of 2.580 mm² (M1, Certificate No.: CDjc2019-01321)

The test has been conducted by the PV Metrology Lab of NIM (National Institute of Metrology, China). Measurement of irradiance intensity and all other measurements are traceable to the International System of Units (SI). Data in this report apply only at the time of the test for the sample. For more details, please refer to the text of the report.

Forward Scan:

Area (mm^2)	I_{sc}(mA)	V_{oc}(V)	P_{max}(mW)
2.580	0.706	0.854	0.466
I_{max}(mA)	V_{max}(V)	FF (%)	η (%)
0.648	0.720	77.3	18.1

I-V Characterization Methods:
JJF 1622-2017: Calibration Specification of Solar Cells：Photoelectric Properties
Secondary Reference Cell:
Device S/N: 81#; Device Material: Mono-Si
Solar Simulator:
Classification: AAA (Double-light source: Xeon and Halogen);
Total irradiance: 1000 W/m² based on I_{sc} of the above Secondary Reference Cell.

Fig. S3. NIM (Beijing) report for D18-Cl:N3:$PC_{61}BM$ solar cells.

6. SCLC

Charge carrier mobility was measured by SCLC method. The mobility was determined by fitting the dark current to the model of a single carrier SCLC, which is described by:

$$J = \frac{9}{8}\varepsilon_0\varepsilon_r\mu\frac{V^2}{d^3},$$

where J is the current density, μ is the zero-field mobility of holes (μ_h) or electrons (μ_e), ε_0 is the permittivity of the vacuum, ε_r is the relative permittivity of the material, d is the thickness of the blend film, and V is the effective voltage ($V = V_{appl} - V_{bi}$, where V_{appl} is the applied voltage, and V_{bi} is the built-in potential determined by electrode work function difference). Here, V_{bi} = 0.1 V for hole-only devices, V_{bi} = 0 V for electron-only devices[1]. The mobility was calculated from the slope of $J^{1/2}$–V plot.

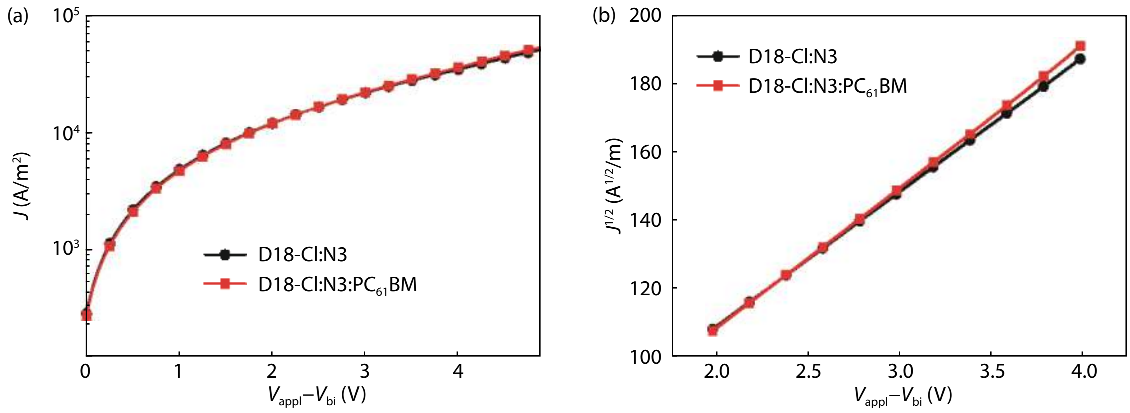

Fig. S4. (a) J–V curves and (b) corresponding $J^{1/2}$– V plots for the hole-only devices (in dark). The thicknesses for D18-Cl:N3 (1 : 1.4) and D18-Cl:N3:$PC_{61}BM$ (1 : 1.4 : 0.1) films are 118 and 115 nm, respectively.

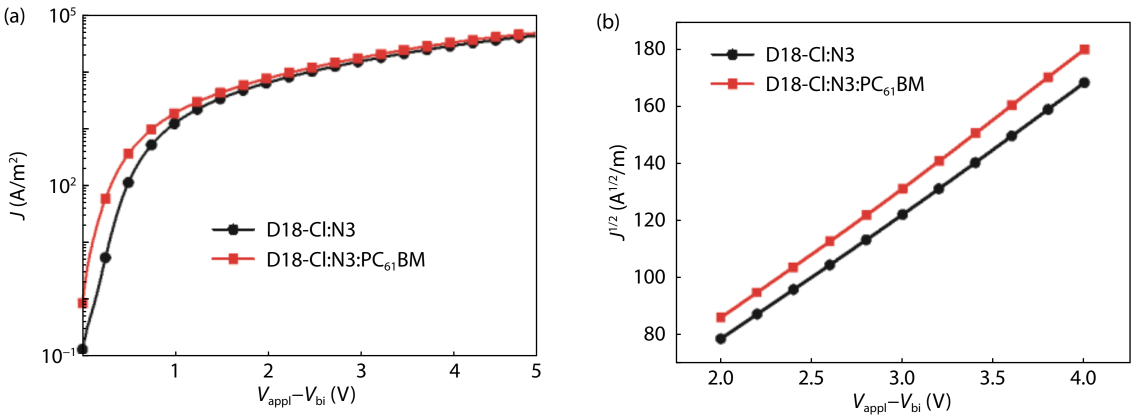

Fig. S5. (a) J–V curves and (b) corresponding $J^{1/2}$– V plots for the electron-only devices (in dark). The thicknesses for D18-Cl:N3 (1 : 1.4) and D18-Cl:N3:$PC_{61}BM$ (1 : 1.4 : 0.1) films are 98 and 100 nm, respectively.

Table S4. Hole and electron mobilities.

Film	μ_h (cm²/(V·s))	μ_e (cm²/(V·s))	μ_h/μ_e
D18-Cl:N3 (1 : 1.4)	8.18×10^{-4}	6.32×10^{-4}	1.29
D18-Cl:N3:$PC_{61}BM$ (1 : 1.4 : 0.1)	8.34×10^{-4}	7.42×10^{-4}	1.12

References

[1] Duan C, Cai W, Hsu B, et al. Toward green solvent processable photovoltaic materials for polymer solar cells: the role of highly polar pendant groups in charge carrier transport and photovoltaic behavior. Energy Environ Sci, 2013, 6, 3022