Manuscript Template

Supplementary Information

Incredible PCE Enhancement Induced by Damaged Perovskite Layers: Deeply Understanding the Working Principle of Additives in Bulk Heterojunction Perovskite Solar Cells

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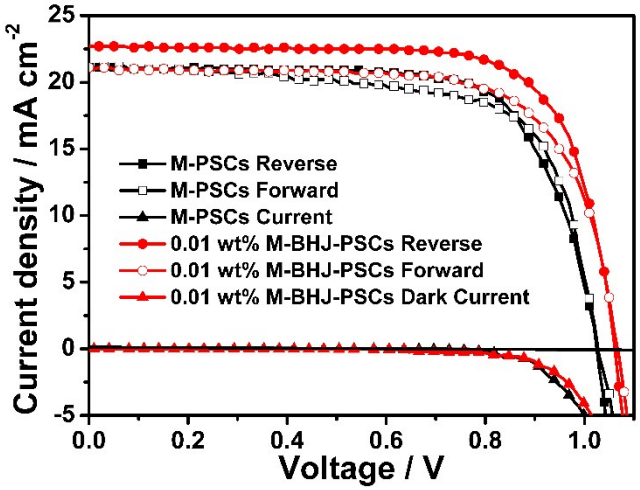

Fig. S1. J-V curves of the champion M-PSCs and M-BHJ-PSCs with 0.01 wt% Spiro-OMeTAD additive.

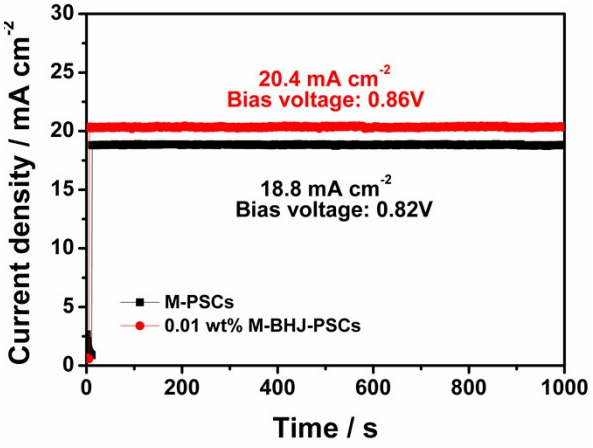

Fig. S2 Steady-state efficiencies measurements in M-PSCs and M-BHJ-PSCs.

The crystallization of perovskite layer was deterred by Spiro-OMeTAD additive, even an extremely low concentration (0.01 wt%), which could be observed from XRD results shown as Fig. S3 and Table 2. With more Spiro-OMeTAD additive, the full width of peaks at half maxima (FWHM) of the perovskite crystal in XRD increased. From equation of Debye-Scherrer:

$$D = \frac{K\lambda}{B\cos\theta}$$

Where D is size of perovskite crystal, K is constant of Scherrer, B is FWHM, θ is angle of diffraction. Therefore, the Spiro-OMeTAD additive shows a negative role in the crystallization of perovskite film, because the size of perovskite layer decreased with FWHM increasing.

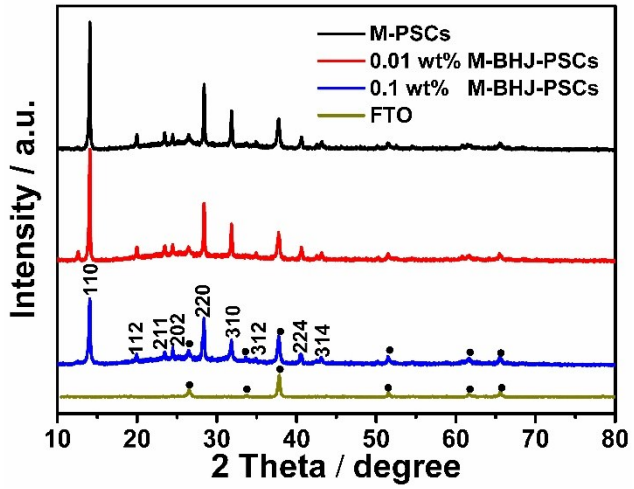

Fig. S3 XRD patterns of perovskite layer in M-PSCs and M-BHJ-PSCs with different Spiro-OMeTAD additive.

Table 2 XRD parameter full width of peaks at half maxima (FWHM) of perovskite layer in M-PSCs and M-BHJ-PSCs with different Spiro-OMeTAD additive.

Spiro-OMeTAD additive (wt%)	FWHM(2θ=14.1°)	FWHM(2θ=28.4°)	FWHM(2θ=31.8°)
0	0.178	0.170	0.186

0.01	0.183	0.194	0.177
0.1	0.237	0.251	0.280

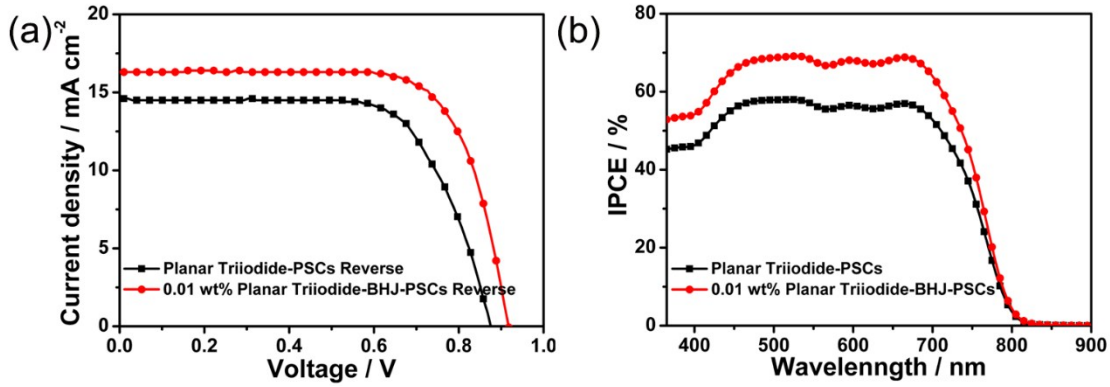

Fig. S4 (a) J-V curves of the champion planar PSCs ($MAPbI_3$) and planar BHJ-PSCs ($MAPbI_3$) with 0.01 wt% Spiro-OMeTAD additive measured under simulated AM 1.5G (100 mW cm^{-2}) illumination and dark with a reverse voltage scanning mode; (b) IPCE curves of the champion planar PSCs ($MAPbI_3$) and planar BHJ-PSCs ($MAPbI_3$) with 0.01 wt% Spiro-OMeTAD additive.

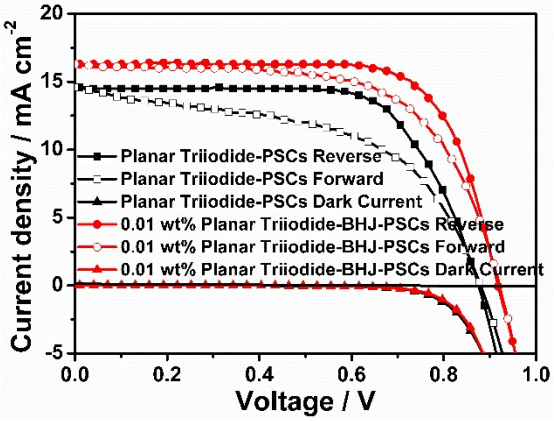

Fig. S5 J-V curves of the champion planar PSCs ($MAPbI_3$) and planar BHJ-PSCs ($MAPbI_3$) with 0.01 wt% Spiro-OMeTAD additive.

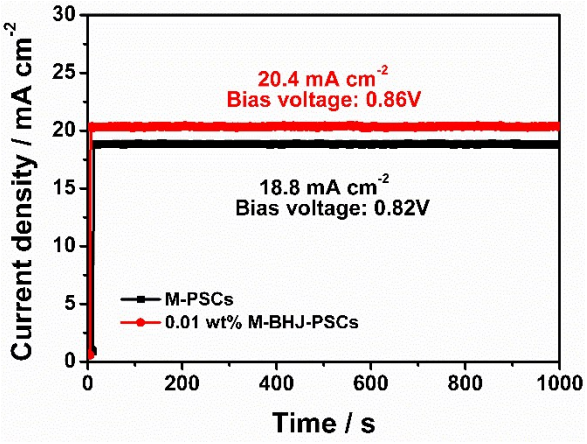

Fig. S6 Steady-state efficiencies measurements in planar PSCs ($MAPbI_3$) and planar BHJ-PSCs ($MAPbI_3$).

Table 3. Photovoltaic parameters based on planar BHJ-PSCs ($MAPbI_3$) with different Spiro-OMeTAD additive.

Spiro-OMeTAD additive		Voc [V]	Jsc [mA cm^{-2}]	FF	PCE [%]
0 wt%	Average	0.83±0.03	14.54±1.27	0.68±0.05	8.21±0.59
	Champion	0.85	14.98	0.71	8.99
0.01 wt%	Average	0.87±0.04	16.68±1.19	0.71±0.03	10.17±0.53
	Champion	0.91	16.28	0.74	10.99
0.03 wt%	Average	0.83±0.03	14.50±1.88	0.69±0.04	8.39±0.51
	Champion	0.87	14.59	0.70	8.82
0.05 wt%	Average	0.83±0.03	10.59±2.08	0.68±0.09	6.19±0.48
	Champion	0.84	12.02	0.69	6.97
0.1 wt%	Average	0.78±0.05	7.77±2.67	0.67±0.06	4.22±0.48
	Champion	0.81	8.89	0.69	4.97

Data in parentheses are the average values of the fabricated 30 PSCs.

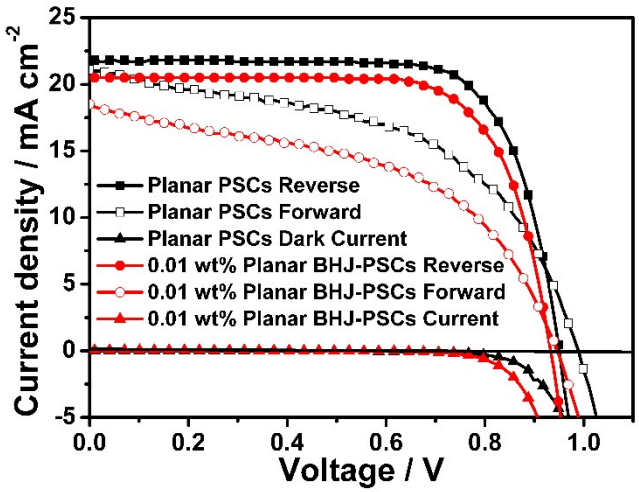

Fig. S7. J-V curves of the champion planar PSCs and planar BHJ-PSCs ($MAPbI_{3-x}Cl_x$) with 0.01 wt% Spiro-OMeTAD additive.

Table 4 Photovoltaic parameters based on planar BHJ-PSCs with different Spiro-OMeTAD additive.

Spiro-OMeTAD additive		Voc [V]	Jsc [mA cm^{-2}]	FF	PCE [%]
0 wt%	Average	0.94±0.02	20.39±0.79	0.73±0.01	13.97±0.78
	Champion	0.95	21.72	0.74	15.32
0.01 wt%	Average	0.91±0.02	18.28±0.89	0.73±0.02	12.06±0.80
	Champion	0.93	20.54	0.73	13.91
0.03 wt%	Average	0.89±0.04	16.76±1.29	0.73±0.01	10.85±1.16
	Champion	0.91	19.16	0.74	12.95
0.05 wt%	Average	0.87±0.03	15.38±0.91	0.73±0.01	9.76±0.70
	Champion	0.93	16.89	0.71	11.17
0.1 wt%	Average	0.86±0.03	14.35±1.22	0.73±0.02	8.90±0.68
	Champion	0.90	15.81	0.72	10.22

Data in parentheses are the average values of the fabricated 50 PSCs.

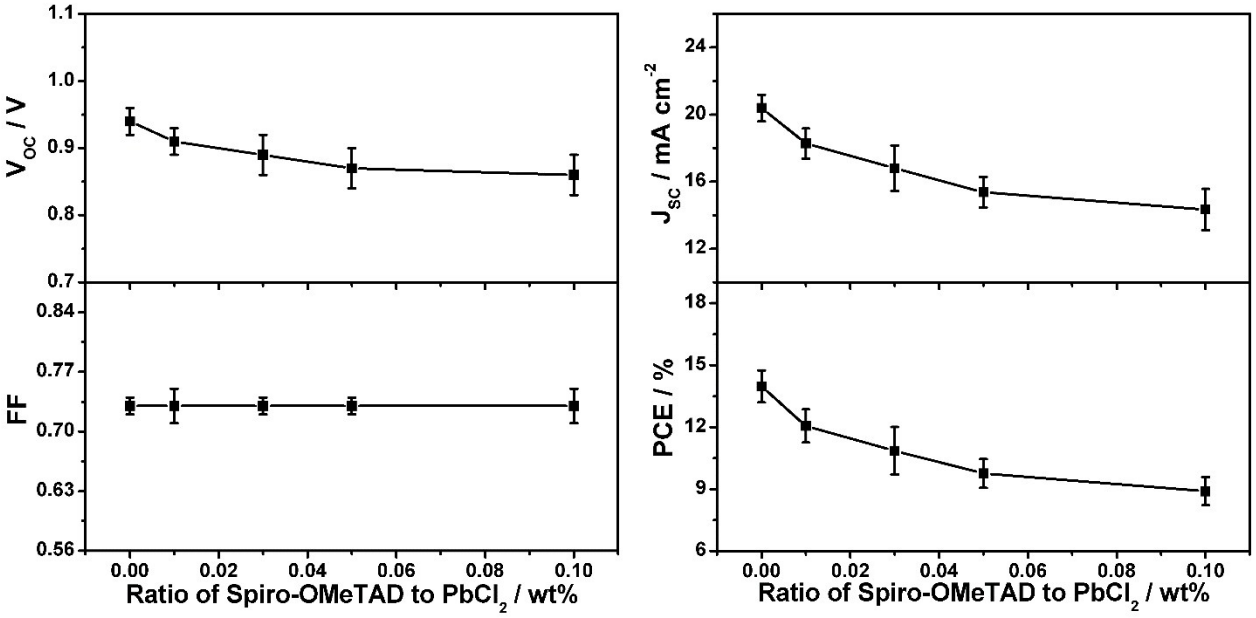

Fig. S7. Jsc, Voc, FF and PCE values versus different percentages by weight of Spiro-OMeTAD additive to mixed halide perovskite ($MAPbI_{3-x}Cl_x$) in planar BHJ-PSCs.

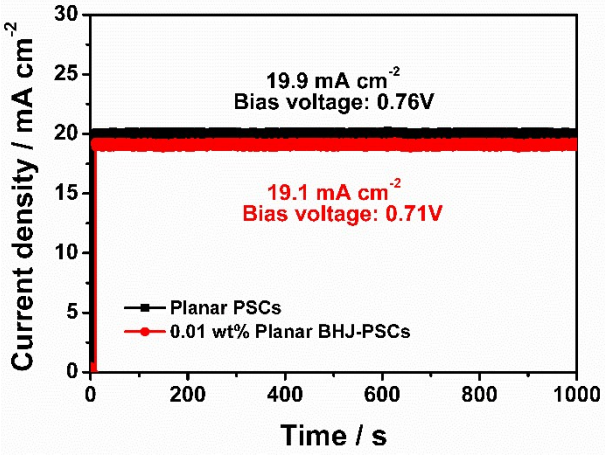

Fig. S8 Steady-state efficiencies measurements in planar PSCs ($MAPbI_{3-x}Cl_x$) and planar BHJ-PSCs ($MAPbI_{3-x}Cl_x$).

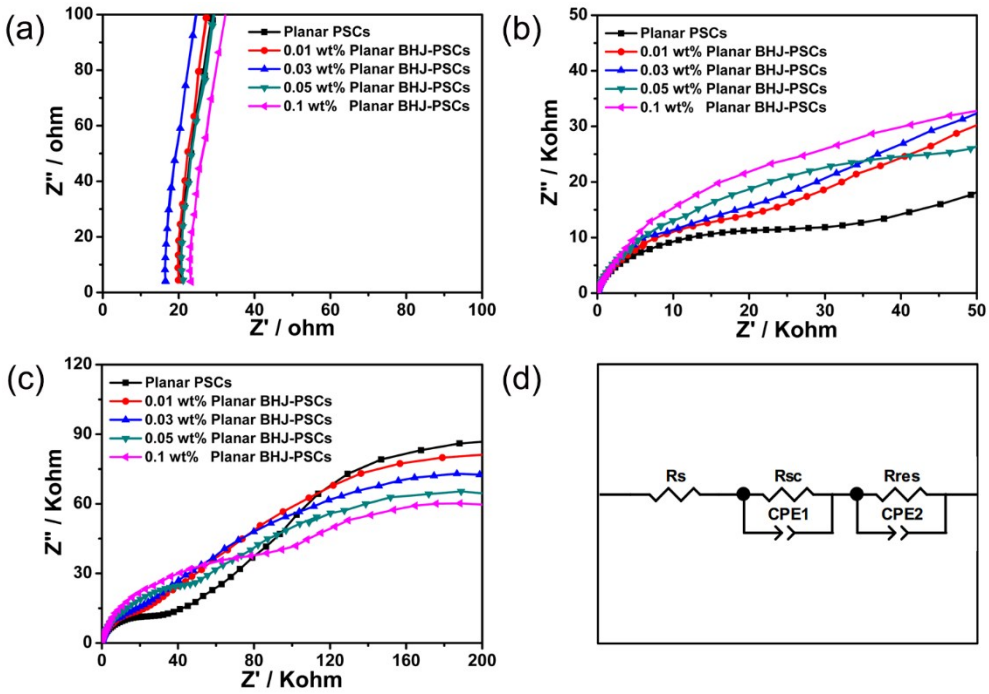

Fig. S9. (a-c) Sequence of Nyquist plot of PSCs based on planar BHJ-PSCs ($MAPbI_{3-x}Cl_x$ light absorber) with different ratio of Spiro-OMeTAD additive in dark at 0.6 V, (d) was the equivalent circuit.