Electronic Supplementary Information

Inverted Planar Solar Cells based on Perovskite/Graphene Oxide Hybrid Composites

Chih-Chun Chung, Sudhakar Narra, Efat Jokar, Hui-Ping Wu and Eric Wei-Guang Diau*

Department of Applied Chemistry and Institute of Molecular Science, National Chiao Tung University, No.1001, Ta-Hsueh Rd., Hsinchu 30010, Taiwan

E-mail:diau@mail.nctu.edu.tw

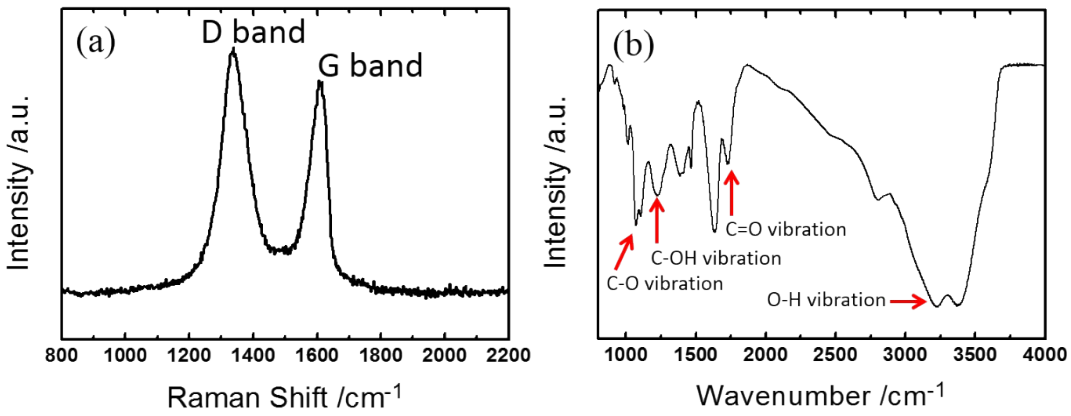

Figure S1. Characteristics of synthesized graphene oxide nanosheets with (a) Raman spectrum and (b) infrared (FTIR) spectrum.

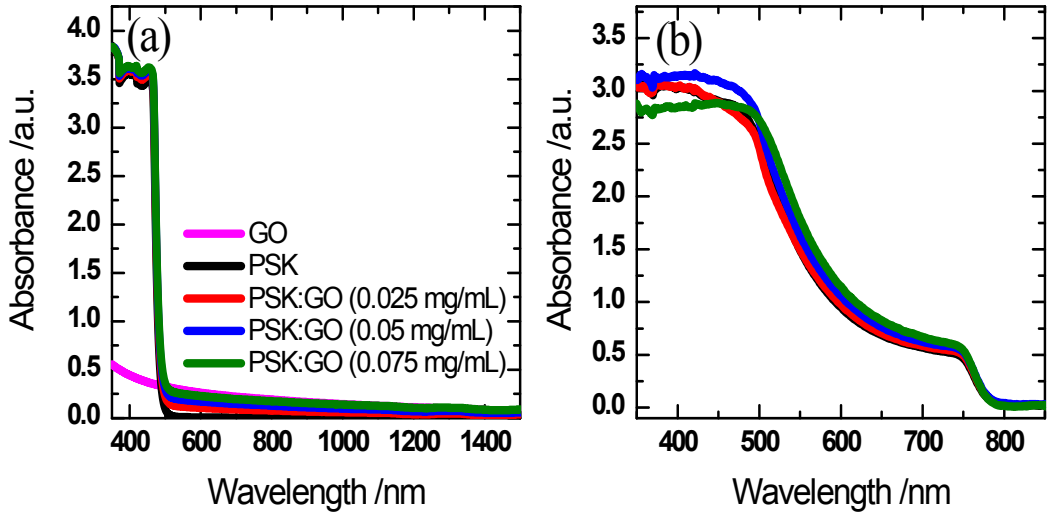

Figure S2. UV-visible absorption spectra of (a) GO and PSK:GO solutions and (b) the corresponding PSK and PSK:GO thin films.

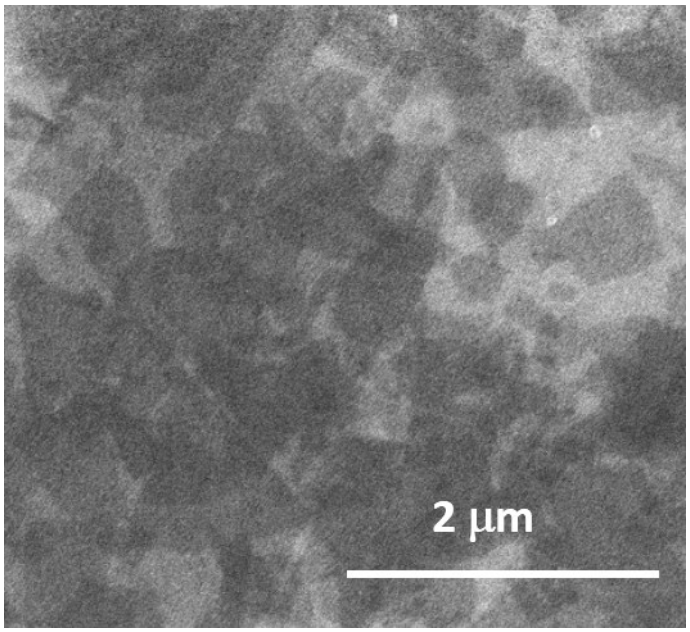

Figure S3. SEM image of GO under layer deposited on ITO substrate showing 1-3 layers of the GO nanosheets.

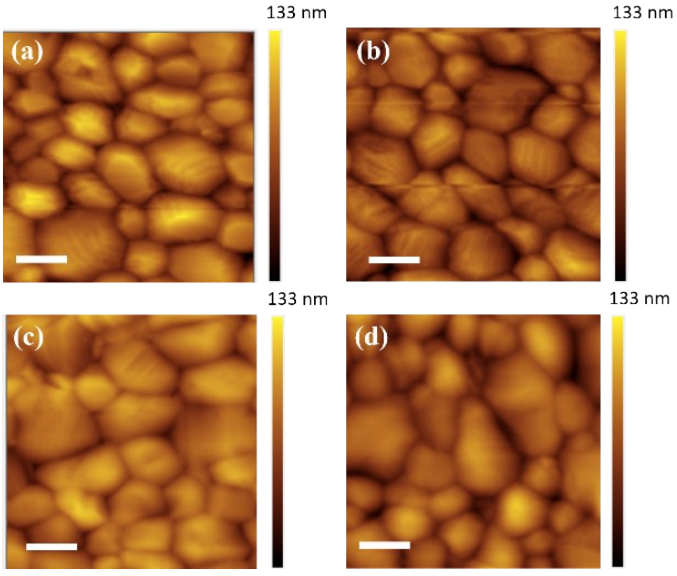

Figure S4. AFM morphology images of (a) pristine PSK layer, (b) PSK:GO (0.025 mg mL^{-1}) layer, (c) PSK:GO (0.05 mg mL^{-1}) layer and (d) PSK:GO (0.075 mg mL^{-1}) layer. Scale bars represent 500 nm.

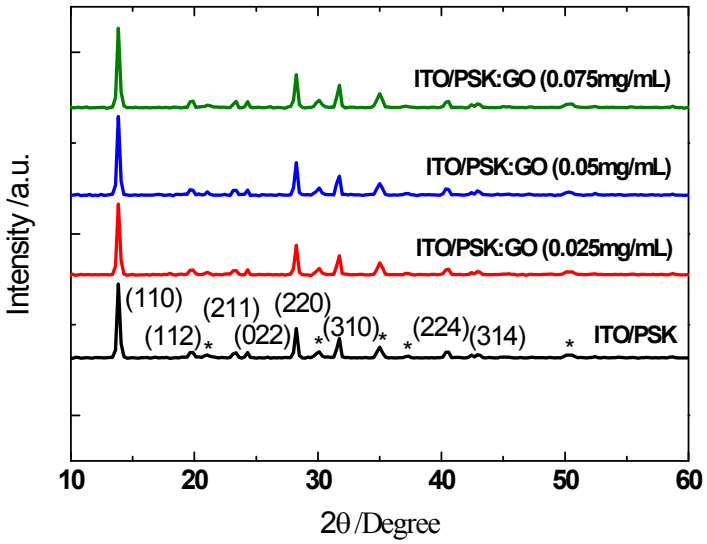

Figure S5. X-ray diffraction patterns of pristine PSK layer and hybrid PSK:GO layer on ITO substrate at varied GO concentrations with XRD facets of PSK as indicated. "*" represents signals of the ITO substrate.

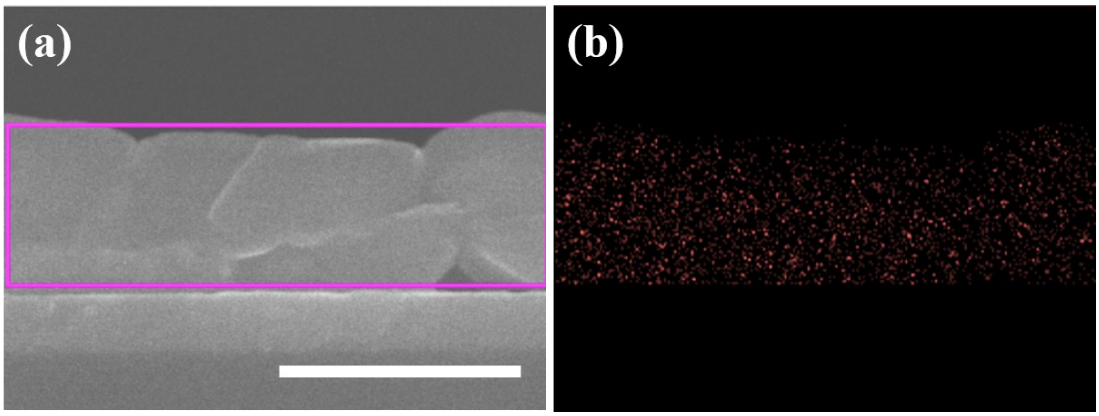

Figure S6. (a) A side-view SEM image and (b) sulfur elemental mapping of EDX of PSK:GO layer blended with $NH_2CH_2CH_2SH$-modified GO nanosheets. Scale bar represents 500 nm.

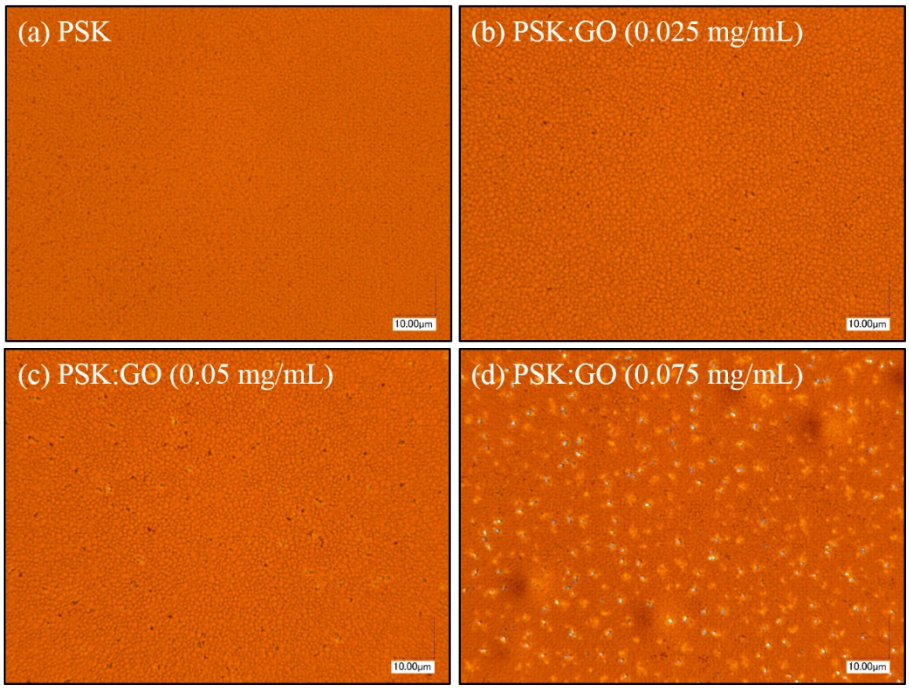

Figure S7. Images (optical microscope) of (a) pristine PSK layer, (b) PSK:GO (0.025 mg mL^{-1}) layer, (c) PSK:GO (0.05 mg mL^{-1}) layer and (d) PSK:GO (0.075 mg mL^{-1}) layer. Scale bars represent 10 µm.

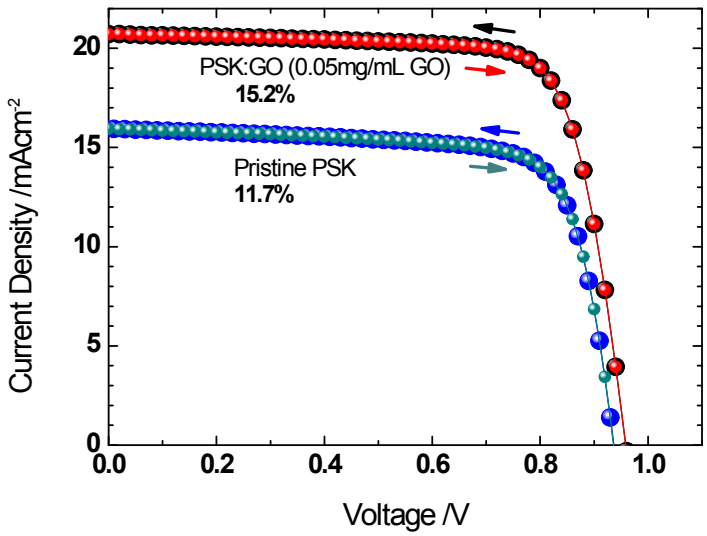

Figure S8. *J*−*V* characteristics of pristine PSK and PSK:GO perovskite solar cells (GO concentration 0.05 mg mL^{-1}) with directions forward (from short circuit to open circuit) and reverse (from open circuit to short circuit).

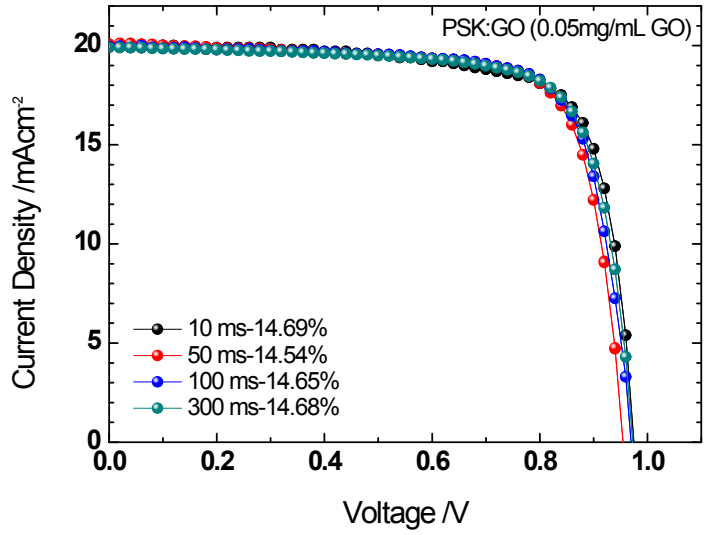

Figure S9. *J*−*V* characteristics of PSK:GO perovskite solar cell (GO concentration 0.05 mg mL^{-1}) with varied delay.

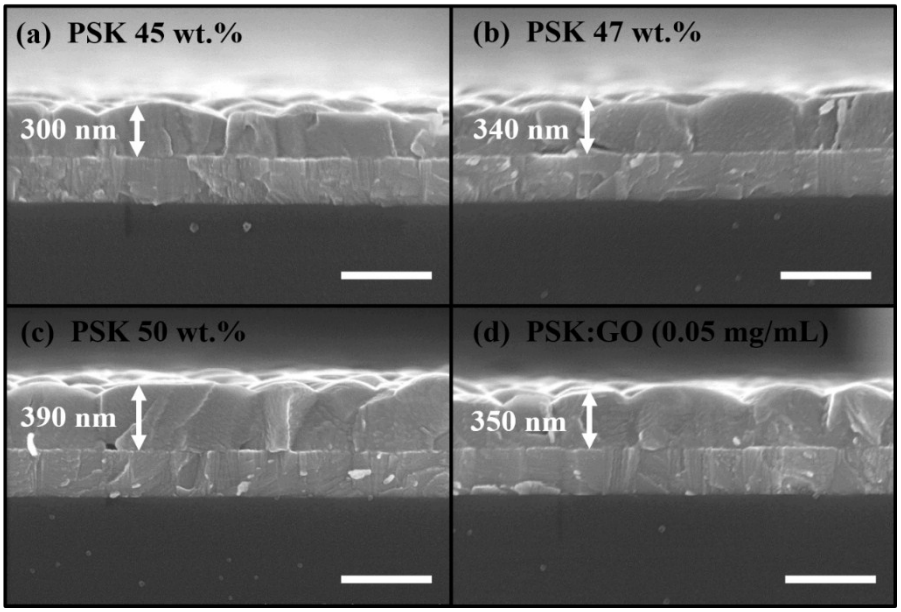

Figure S10. Side-view SEM images of pristine PSK layer fabricated with varied mass proportions in DMF, (a) 45 mass % PSK, (b) 47 mass % PSK, (c) 50 mass % PSK and (d) 45 mass % PSK in the PSK:GO layer with GO concentration 0.05 mg mL^{-1}. Scale bars represent 500 nm.

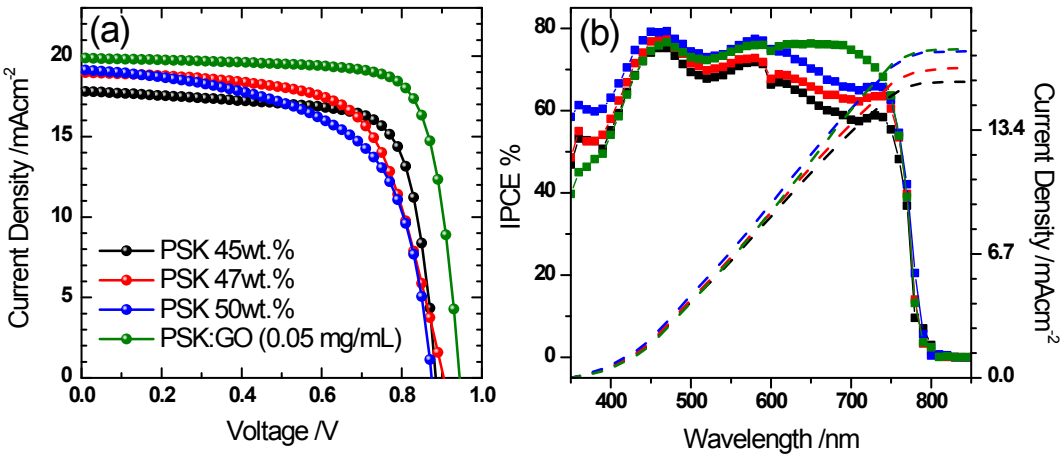

Figure S11. (a) *J–V* characteristics of perovskite solar cells in varied mass proportions of pristine PSK in DMF and hybrid PSK:GO layer with GO concentration 0.05 mg mL^{-1} under AM 1.5 G one-sun illumination. (b) IPCE action spectra and integrated current density of corresponding perovskite solar cells as in (a).

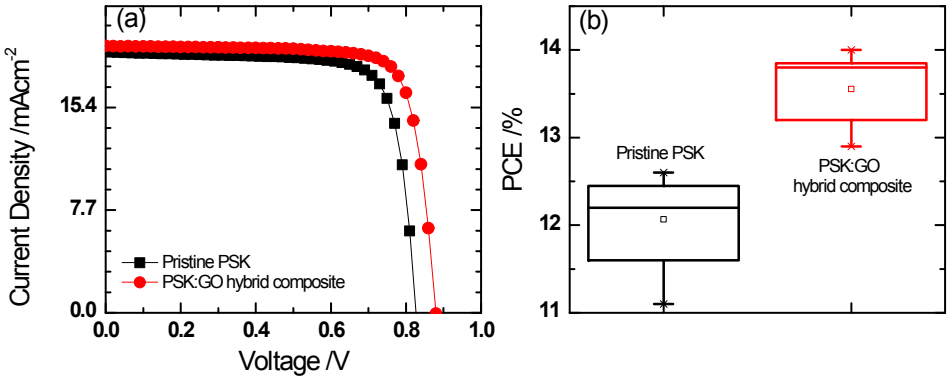

Figure S12. (a) *J–V* characteristics of the best pristine perovskite (no GO; black squares) and PSK:GO hybrid composite (GO concentration 0.05 mg mL^{-1}; red circles) solar cells with PEDOT:PSS as HEL under AM 1.5 G one-sun illumination. (b) The power conversion efficiency (PCE) distribution box plots of the corresponding inverted perovskite solar cells as labelled.

Table S1. Photovoltaic parameters of perovskite solar cells in varied mass proportions of pristine PSK in DMF and hybrid PSK:GO layer with GO concentration 0.05 mg mL^{-1} under AM 1.5 G one-sun illumination.

Device	J_{SC} /mAcm^{-2}	V_{OC} /mV	FF	PCE /%
PSK (45 wt.%)	17.84	900	0.73	11.8
PSK (47 wt.%)	18.96	905	0.65	11.2
PSK (50 wt.%)	19.13	877	0.61	10.2
PSK:GO (0.05 mg mL^{-1})	19.87	949	0.77	14.6

Table S2. Photovoltaic parameters of perovskite solar cells with PEDOT:PSS as HEL under AM 1.5 G one-sun illumination.

Device	J_{SC} /mAcm^{-2}	V_{OC} /mV	FF	PCE /%[a]
Pristine PSK	19.55	835	0.77	12.6 (12.1±0.5)
PSK:GO (0.05 mg mL^{-1})	20.01	886	0.79	14.0 (13.6±0.4)

[a] The average values of PCE are shown in the parentheses (20 devices) with the uncertainties representing one standard deviation.

Table S3. Photovoltaic parameters of twenty PEDOT:PSS based perovskite solar cells with pristine perovskites in the absence of GO under simulated AM 1.5 G illumination (power density 100 mW cm^{-2}) with active area 0.0225 cm^2.

Cell No.	J_{SC} /mAcm^{-2}	V_{OC} /mV	*FF*	*PCE* /%
1	19.55	835	0.775	12.6
2	18.78	842	0.793	12.6
3	18.91	844	0.786	12.6
4	19.67	849	0.746	12.5
5	19.58	847	0754	12.5
6	18.48	846	0.793	12.4
7	18.57	847	0.781	12.3
8	18.63	849	0.776	12.3
9	18.61	869	0.757	12.2
10	19.47	838	0.744	12.2
11	18.61	847	0.774	12.2
12	18.91	844	0.755	12.1
13	19.18	817	0.775	12.1
14	19.70	857	0.719	12.1
15	18.25	844	0.754	11.6
16	19.15	829	0.731	11.6
17	18.70	837	0.740	11.6
18	18.45	839	0.739	11.4
19	18.09	847	0.734	11.3
20	18.21	827	0.737	11.1
mean	18.86±0.51	843±11	0.758±0.022	12.1±0.5

Table S4. Photovoltaic parameters of twenty PEDOT:PSS based perovskite solar cells with perovskite/GO hybrid composite (GO concentration 0.05 mg mL^{-1}) under simulated AM 1.5 G illumination (power density 100 mW cm^{-2}) with active area 0.0225 cm^2.

Cell No.	J_{SC} /mAcm^{-2}	V_{OC} /mV	*FF*	*PCE* /%
1	20.01	886	0.791	14.0
2	20.00	883	0.788	13.9
3	19.92	881	0.793	13.9
4	20.18	853	0.806	13.9
5	19.91	878	0.792	13.9
6	19.94	879	0.786	13.8
7	20.12	839	0.816	13.8
8	19.85	875	0.793	13.8
9	20.27	847	0.806	13.8
10	20.30	842	0.810	13.8
11	19.51	868	0.794	13.5
12	19.57	872	0.790	13.5
13	19.91	850	0.780	13.2
14	19.59	853	0.787	13.2
15	19.86	847	0.786	13.2
16	19.63	875	0.761	13.1
17	19.75	876	0.756	13.1
18	19.67	862	0.768	13.0
19	19.70	863	0.759	12.9
20	19.83	827	0.784	12.8
mean	19.89±0.24	868±14	0.788±0.016	13.5±0.4

Table S5. Parameters obtained from measurements of the Hall effect with perovskite films containing varied amounts of graphene oxide in PSK:GO hybrid composites.

Film	**carrier concentration / 10^{20} cm^{-3}**	**mobility / cm^2 V^{-1} s^{-1}**	**R_{Hs} /m^2 C^{-1}**
pristine PSK	10.8	28.6	-0.0181
PSK:GO (0.025 mg mL^{-1})	8.94	32.5	-0.0205
PSK:GO (0.05 mg mL^{-1})	7.93	35.3	-0.0219
PSK:GO (0.075 mg mL^{-1})	7.68	34.7	-0.0214

Table S6. Time coefficients (relative amplitudes) of corresponding perovskite films obtained from fitting PL transients (Figure 4) (excitation at 635 nm, probe at 770 nm) with the uncertainties representing one standard deviations.

Film	τ_1 /ns (A1)	τ_2 /ns (A2)	τ_3 /ns (A3)	τ_{PL} /ns[b]
PSK	24.3±0.9 (0.23)	429.3±1.8 (0.77)	—	336.1±1.6
PSK:GO (0.025)[a]	3.5±0.1 (0.57)	66.5±3.5 (0.11)	600.6±7.3 (0.32)	201.5±2.8
PSK:GO (0.05)[a]	2.0±0.1 (0.76)	54.2±3.7 (0.08)	946.4±0.2 (0.16)	157.3±0.3
PSK:GO (0.075)[a]	4.0±0.2 (0.35)	71.2±2.3 (0.23)	489.1±4.8 (0.42)	223.2±2.6

[a] Concentration of GO in the PSK:GO hybrid layers in mg mL^{-1}.

[b] The average PL lifetimes calculated with equation, $\tau_{PL} = \frac{\sum A_i \tau_i}{\sum A_i}$, here τ_i are time coefficients and A_i are the corresponding amplitudes of each component.

Table S7. Impedance parameters of corresponding perovskite solar cells obtained from fitting EIS data (Figure 5) (Z-view software) based on the equivalent circuit shown in Figure 5.

Device	R_S/Ω	R_1/Ω	C_1/10^{-8} F	R_2/Ω	C_2/F
PHJ	17.9	75.5	5.0	19.5	0.00137
PSK:GO (0.025 mg mL^{-1})	26.5	79.4	1.5	26.4	1.67E-06
PSK:GO (0.05 mg mL^{-1})	17.1	313.4	12	232.5	0.00144
PSK:GO (0.075 mg mL^{-1})	18.3	176.5	2.3	38.3	0.00186

Table S8. Photovoltaic parameters of forty inverted GO-based perovskite solar cells under simulated AM 1.5 G illumination (power density 100 mW cm^{-2}) with active area 0.0225 cm^2.

Cell No.	J_{SC}/mAcm^{-2}	V_{OC}/mV	*FF*	*PCE* /%
1	18.11	927	0.731	12.3
2	17.38	918	0.767	12.2
3	19.99	885	0.682	12.0
4	17.84	900	0.733	11.8
5	17.94	924	0.706	11.7
6	17.60	979	0.674	11.6
7	16.71	955	0.730	11.6
8	19.00	911	0.669	11.6
9	17.60	979	0.675	11.6
10	19.00	933	0.654	11.6
11	16.98	984	0.688	11.5
12	18.13	926	0.686	11.5
13	18.67	997	0.617	11.5
14	19.28	912	0.654	11.5
15	18.99	971	0.617	11.4
16	18.65	907	0.672	11.4
17	18.72	909	0.653	11.4
18	17.99	993	0.628	11.2
19	17.79	977	0.640	11.1
20	17.95	950	0.650	11.1
21	17.05	975	0.662	11.0
22	17.48	981	0.635	10.9
23	17.50	971	0.634	10.8
24	16.44	914	0.712	10.7
25	17.69	976	0.617	10.6
26	17.36	993	0.614	10.6
27	17.65	989	0.604	10.5
28	18.83	975	0.570	10.5
29	17.80	950	0.558	10.5
30	17.82	981	0.598	10.5
31	17.68	960	0.613	10.4
32	17.76	981	0.594	10.4
33	17.68	972	0.603	10.4

34	17.55	979	0.601	10.3
35	17.77	981	0.591	10.3
36	17.25	985	0.599	10.2
37	16.15	923	0.677	10.1
38	17.58	961	0.589	10.0
39	17.16	983	0.587	9.9
40	15.40	978	0.659	9.9
mean	17.80±0.87	956±32	0.646±0.049	11.0±0.7

Table S9. Photovoltaic parameters of thirty inverted perovskite/GO hybrid composite solar cells (0.025 mg mL^{-1} GO) under simulated AM 1.5 G illumination (power density 100 mW cm^{-2}) with active area 0.0225 cm^2.

Cell No.	J_{SC} /mAcm^{-2}	V_{OC} /mV	*FF*	*PCE* /%
1	19.59	945	0.730	13.5
2	20.17	927	0.723	13.5
3	19.69	931	0.737	13.5
4	19.10	929	0.754	13.4
5	19.19	956	0.728	13.4
6	19.26	920	0.753	13.3
7	19.17	900	0.773	13.3
8	19.85	967	0.695	13.3
9	19.43	950	0.720	13.3
10	19.93	920	0.720	13.2
11	20.14	920	0.703	13.2
12	20.43	940	0.686	13.2
13	20.45	950	0.675	13.1
14	20.69	956	0.663	13.1
15	19.92	984	0.666	13.0
16	21.00	962	0.645	13.0
17	19.18	971	0.691	12.9
18	19.77	945	0.697	12.7
19	18.62	974	0.679	12.3
20	20.56	986	0.604	12.3
21	18.76	959	0.677	12.2
22	20.67	983	0.597	12.1
23	20.00	944	0.636	12.0
24	18.74	955	0.668	12.0
25	18.97	963	0.656	12.0
26	20.96	964	0.594	12.0
27	18.97	966	0.639	11.7
28	19.00	911	0.669	11.6
29	19.00	933	0.654	11.6
30	18.95	966	0.628	11.5
mean	19.67±0.71	950±23	0.682±0.046	12.7±0.7

Table S10. Photovoltaic parameters of forty inverted perovskite/GO hybrid composite solar cells (0.05 mg mL^{-1} GO) under simulated AM 1.5 G illumination (power density 100 mW cm^{-2}) with active area 0.0225 cm^2.

Cell No.	J_{SC} /mAcm^{-2}	V_{OC} /mV	*FF*	*PCE* /%
1	20.71	959	0.763	15.2
2	20.55	921	0.803	15.2
3	21.00	919	0.789	15.2
4	20.12	925	0.811	15.1
5	20.13	967	0.774	15.1
6	20.21	940	0.787	15.0
7	20.55	924	0.790	15.0
8	20.50	940	0.778	15.0
9	20.60	914	0.800	15.0
10	21.44	926	0.744	14.8
11	19.99	987	0.745	14.7
12	19.64	980	0.762	14.7
13	19.53	961	0.784	14.7
14	21.05	945	0.735	14.6
15	20.10	985	0.735	14.5
16	20.19	933	0.770	14.5
17	19.71	949	0.773	14.5
18	20.27	962	0.745	14.5
19	19.72	960	0.756	14.3
20	19.81	988	0.733	14.3
21	20.10	940	0.756	14.3
22	19.36	920	0.805	14.3
23	20.10	946	0.747	14.2
24	19.82	988	0.724	14.2
25	18.95	940	0.796	14.2
26	20.12	984	0.715	14.2
27	19.01	940	0.797	14.2
28	21.75	974	0.663	14.1
29	18.86	940	0.787	14.0
30	18.78	940	0.786	13.9
31	19.32	966	0.737	13.8
32	18.96	960	0.757	13.8
33	19.86	920	0.755	13.8

34	18.71	932	0.790	13.8
35	18.78	962	0.765	13.8
36	20.34	975	0.692	13.7
37	19.90	927	0.736	13.6
38	19.14	955	0.740	13.5
39	19.19	956	0.728	13.4
40	20.95	964	0.646	13.0
mean	19.55±0.75	950±22	0.757±0.037	14.3±0.6

Table S11. Photovoltaic parameters of thirty inverted perovskite/GO hybrid composite solar cells (0.075 mg mL^{-1} GO) under simulated AM 1.5 G illumination (power density 100 mW cm^{-2}) with active area 0.0225 cm^2.

Cell No.	J_{SC} /mAcm^{-2}	V_{OC} /mV	*FF*	*PCE* /%
1	19.02	930	0.651	11.5
2	20.77	993	0.560	11.5
3	18.74	984	0.615	11.4
4	19.89	962	0.596	11.4
5	19.03	971	0.607	11.2
6	18.07	995	0.623	11.2
7	19.08	933	0.627	11.2
8	19.10	932	0.629	11.2
9	18.85	975	0.610	11.2
10	18.84	976	0.611	11.2
11	18.34	970	0.620	11.0
12	20.96	935	0.560	11.0
13	19.92	943	0.578	10.9
14	18.99	973	0.588	10.9
15	20.58	905	0.583	10.9
16	18.71	964	0.596	10.8
17	18.97	965	0.591	10.8
18	18.94	963	0.586	10.7
19	17.36	993	0.614	10.6
20	20.98	930	0.538	10.5
21	18.82	985	0.559	10.4
22	19.79	933	0.559	10.3
23	19.88	927	0.556	10.3
24	18.46	962	0.573	10.2
25	19.05	935	0.575	10.2
26	19.88	913	0.557	10.1
27	18.29	971	0.565	10.0
28	18.43	952	0.566	9.9
29	19.36	979	0.506	9.6
30	18.40	936	0.551	9.5
mean	19.18±0.87	956±25	0.585±0.032	10.7±0.6