## **Supporting Information**

## High Performance PbS Quantum Dot/Graphene Hybrid Solar Cell with Efficient Charge Extraction

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**Fig. S1** (a) X-ray diffraction patterns and HRTEM image (inset) of the PbS nanocrystal. (b) Representative SEM image of SG flakes (average lateral size of ~550 nm) homogeneously deposited on PbS QD film.

The crystallinity of the PbS QD film is examined by the XRD measurement as shown in Figure S1a. Well-defined diffraction peaks were assigned to the (111), (200), (220), and (311) planes, respectively. It clearly implies that the PbS in the solid state has a face-center-cubic (fcc) structure. The HRTEM lattice fringe image is taken from a 1.3 eV PbS QD and the diameter of the QD is approximately 2.5 nm (inset).



Fig. S2 UV-vis absorption of 1.3 eV PbS QDs capped with oleic acid in solution (peak

around 950 nm). A size distribution of the corresponding PbS QD is obtained with a mean diameter of  $\sim 2.5$  nm.



Fig. S3 10 x 10  $\mu$ m AFM images showing the surface morphologies of (a and b) PbS and (c and d) PbS/SG films. The root mean square (RMS) roughness are 13.204 and 11.641 nm, respectively. The image area is 10 x10  $\mu$ m.



Fig. S4 Schematic of devices of (a) TBAI-PbS, (b) TBAI-PbS/SG (5L), and (c) TBAI-

PbS/SG (9L). All devices are fabricated under the same conditions and consist of 12 layers of TBAI-PbS with 0, 5, and 9 layers of SG flake. For the hybrid cell fabrication, 5 and 9 layers of SG flake were symmetrically inserted in the TBAI-PbS layers as seen in Fig. S4 (b) and (c).



**Fig. S5** Schematic of (a) TBAI-PbS/EDT-PbS and (b) TBAI-PbS/SG (9L)/EDT-PbS junction solar cells. They consist of 10 layers of TBAI-PbS and 2 layers of EDT-PbS on ZnO/ITO/glass substrates. For the hybrid cell fabrication, 9 layers of SG flake were inserted in TBAI-PbS CQD layers.

**Table S1** Photovoltaic performance of TBAI-PbS/EDT-PbS and TBAI-PbS/SG (9L)/EDT-PbS junction structure. Average values of each device with standard deviations were collected from four devices.

|   | $V_{\rm oc}({ m V})$ | FF              | $J_{\rm sc}({\rm mA/cm}^2)$ | $R_{\rm s}(\Omega)$ | PCE (%)   |
|---|----------------------|-----------------|-----------------------------|---------------------|-----------|
| TBAI-PbS<br>/EDT-PbS                      | $0.51 \pm 0.01$      | $0.62 \pm 0.02$ | 23.61±0.10                  | 2.10±0.28           | 7.53±0.18 |
| TBAI-PbS/SG (9L)<br>/EDT-PbS              | $0.48 \pm 0$         | 0.61±0.03       | 29.81±0.66                  | $1.47 \pm 0.78$     | 8.82±0.55 |
| TBAI-PbS/SG (9L)<br>/EDT-PbS <sup>a</sup> | 0.48                 | 0.63            | 30.34                       | 1.46                | 9.18      |

<sup>a</sup> Best performance of TBAI-PbS/SG (9L)/EDT-PbS junction device.

Table S2 Photoresponse characteristics of PbS and PbS/SG devices.

|        | off state (pA) | on state (nA) | on-off ratio | Rise time<br>(τ, msec) |
|--------|----------------|---------------|--------------|------------------------|
| PbS    | 310            | 8.91          | 28.74        | 3.3                    |
| PbS/SG | 272            | 10.6          | 38.97        | 2.1                    |