Supporting Information

Surface Photovoltage Predicts Open Circuit Voltage in GaP/PEDOT:PSS and GaP/CuSCN Heterojunction Solar Cells

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Figure S1. Scanning electron micrograph of a crack in an electrodeposited CuSCN film on an unpolished GaP wafer surface. The particles follow the rough contours of the GaP wafer.



Figure S2. Energy dispersive X-ray spectroscopy (EDS) of etched nGaP wafer (a), PEDOT:PSS– n-GaP (b), and CuSCN–n-GaP (c). The beam voltage and current were 5 kV and 1.6 nA, respectively.

Electron Image 3



Figure S3. Scanning electron micrograph of a CuSCN film on GaP with a metallic copper cluster visible. EDS was performed on the cluster area (Spectrum 6) and separate film area (Spectrum 7; values given in **Table S1**), confirming that the cluster primarily contains copper.

Element	Cluster, Spectrum 6 (At%)	Film, Spectrum 7 (At%)
Cu	66.3	11.0
S	9.7	7.5
С	11.8	16.7
Ν	6.2	8.5

Table S1. EDS results showing the atomic percent of elements in the CuSCN film in Figure S3.



Figure S4. Surface photovoltage spectrum of n-type GaP wafer with indium back contact and PEDOT:PSS hole transport layer (In/GaP/PEDOT:PSS) co-plotted with the logarithm of the light power of the monochromated Xe arc lamp at the sample position.



Figure S5. (a) Schematic side view of structure for n-GaP-based devices. Indium (In) is used as the back contact while silver paint is used as the front contact (Ag). Photograph of (b) champion PEDOT:PSS-based device, and of (c) champion CuSCN-based device. Scale bars are 1.00 cm.



Figure S6. *J-V* curves of second PEDOT:PSS–n-GaP (a) and CuSCN–n-GaP (b) devices in the dark (black line) and under simulated AM1.5G illumination (blue and orange lines, respectively).



Figure S7. Full cyclic voltammetry curves for PEDOT:PSS–n-GaP (a) and CuSCN–n-GaP (b). Light source was a Xe lamp adjusted to simulate AM1.5G illumination.



Figure S8. Surface photovoltage spectrum for a GaP wafer with In soldered on the back and annealed in 5% H_2 in N_2 for 10 min at 400 °C.