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Supplementary Materials for

A new metal transfer process for van der Waals contacts to vertical Schottky-junction transition metal dichalcogenide photovoltaics

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This PDF file includes:

Section S1. Detailed metal transfer procedure

Complete details of the metal transfer procedure are described below.

Step 1: Metal contacts on SAM-coated Si/SiO_2. Commercially-available Si wafers coated with 285 nm thermal SiO_2 are diced into chips, cleaned in Nanostrip for 5 minutes, then rinsed 3 times in DI water. The chips are placed in a vacuum desiccator. 5 drops of trichloro(1H,1H,2H,2H-perfluorooctyl)silane (PFOTS, Sigma Aldrich) are placed in a cap in the bottom of the desiccator. The desiccator is evacuated slowly, over the course of 3 minutes, then isolated from the vacuum pump and left evacuated for 1 hour. The chips are removed from the vacuum desiccator. 20 nm of Au is evaporated in an electron-beam evaporator at a speed of 1 Å/s and a base pressure below 5×10^{-7} Torr. Photolithography is performed using positive photoresist and a positive photomask to define the contacts. For photolithography, S1813 is used according to the following recipe: spin at 4000 rpm for 30 seconds, soft bake at 115ºC for 1 minute, expose to 365 nm UV light at 15 mW/cm^2 for 8 seconds, develop in MF 319 for 50 seconds, and then rinse in DI water for 10 seconds. The Au around the photoresist is etched by immersion of the chips in Transene Gold Etchant TFA for 10 seconds, then the sample is rinsed three times in DI water. The photoresist is dissolved in slightly heated acetone (60ºC for 5 minutes).

Step 2: PDMS/PPC stamps. A similar procedure is followed to that developed in Ref. (*25*). PDMS (Sylgard 184) is mixed in a glass petri dish and left in an oven at 60-70ºC overnight. PPC is made by stirring 1.5 g PPC in 10 mL anisole on a hot plate at 60ºC for 1 hour. The PDMS is cut into 1 cm by 1 cm squares with a razor blade. One square is removed from the petri dish, rinsed in IPA for 20 seconds, then dried in nitrogen gas. The PDMS stamp is placed on one end of a glass slide, and the thickest corner of the stamp is identified. The PDMS stamps are plasma-ashed at 300 mTorr and 120 W for 10 minutes. The PDMS stamp is centered on the spinner, and

2 drops of PPC are placed on the PDMS stamp, and then spun at 1500 rpm for 1 minute. The PDMS/PPC stamps are let sit for a few minutes, but not longer than 10 minutes. The edges of the PDMS/PPC stamp are cut away with a fresh razor blade until the stamp is ~2 mm by 2 mm.

Step 3: Transfer of metal contacts. The stage of a 2D transfer setup is heated to 60ºC. A Si/SiO_2 chip containing metal contacts is loaded onto the stage, and the desired contact is centered in the field of view. The PDMS/PPC stamp is loaded onto the top arm of the transfer setup. The thickest corner of the PDMS/PPC stamp is centered in the field of view. When the stamp is lowered, the polymer front should originate from this corner. The PDMS/PPC stamp and the desired contact are aligned so that the PPC completely covers the contact, but so that just the corner of the stamp will make contact with the substrate. The PDMS/PPC stamp is lowered slowly. Once the polymer front progresses just past the contact, the stage temperature is lowered to 40ºC. After the temperature has reached 40ºC, the top arm of the transfer setup is raised slowly. As the polymer front begins to move, but before it reaches the contact, the top arm of the transfer setup and therefore the PDMS/PPC stamp is raised very quickly, picking up the contact with it. The stage of the transfer setup is heated to 60ºC again. The target substrate is loaded onto the stage of the transfer setup and the target flake is centered in the field of view. Once the temperature reaches 60ºC, the contact on the PDMS/PPC stamp is aligned with the flake. The PMDS/PPC stamp is lowered slowly, until the polymer front progresses just past the sample. Immediately, and with the stage still at 60ºC, the PDMS/PPC stamp is raised very slowly. The contact should delaminate from the stamp and stick to the flake. Remaining PPC that occasionally sticks to the flake can be removed by rinsing in chloroform for 5 minutes, then drying in nitrogen gas.

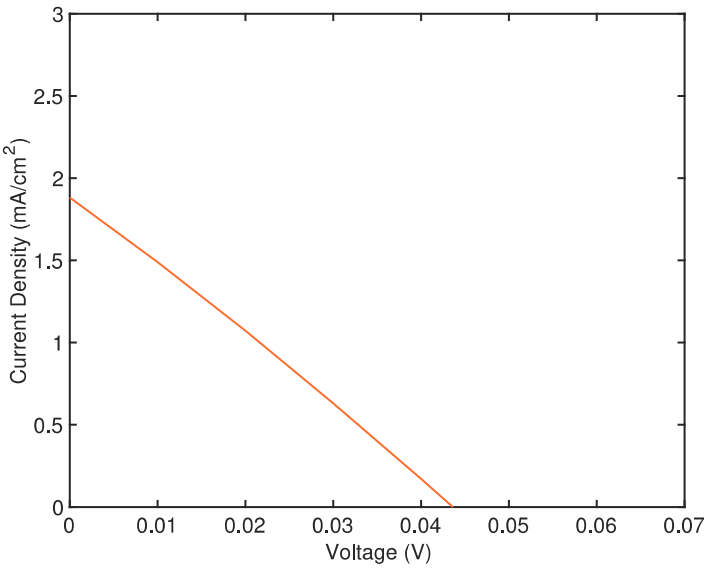
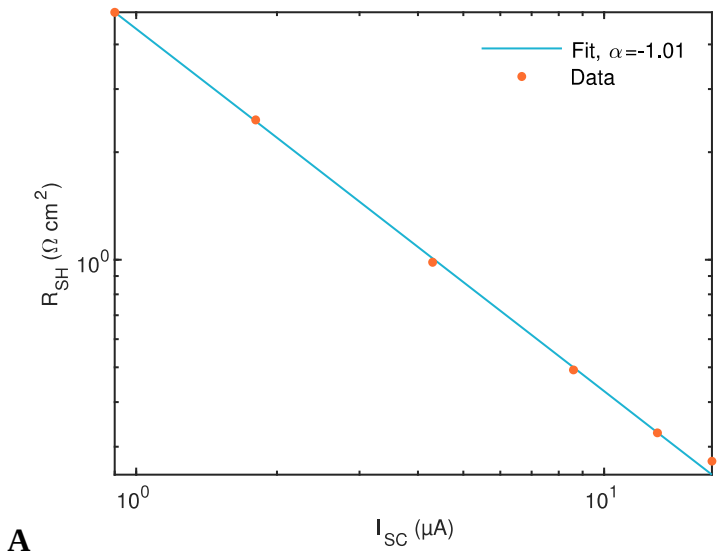
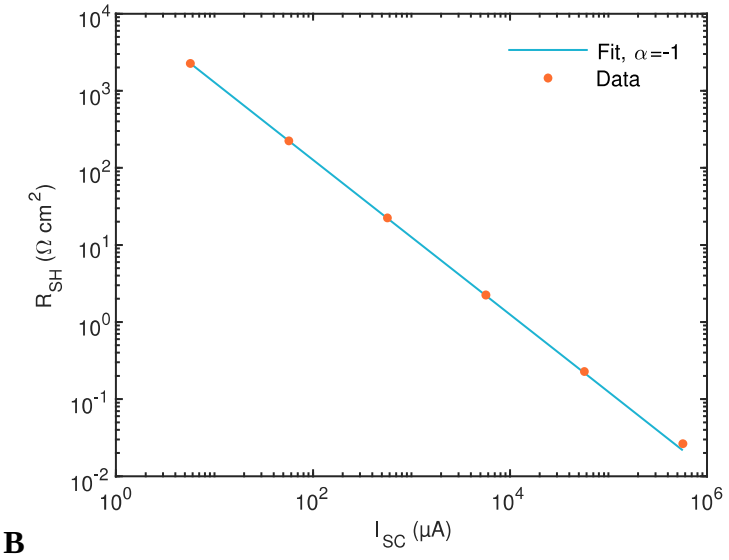


Fig. S1. Simulated I-V curve for evaporated devices. Simulations can replicate the resistive behavior of evaporated-contact devices, assuming the metal contacts have an effective work function difference of 50 meV due to Fermi-level pinning at the Au contact.



A



B

Fig. S2. Photoshunting. (**A**) R_{SH} vs. I_{SC}, experimental. R_{SH} is extracted from the slopes of the power-dependent I-V curves (Fig. 3A of the main text) at short-circuit. A power law fit yields $\alpha \approx -1$. (**B**) R_{SH} vs. I_{SC}, simulated. A power law fit yields $\alpha = -1$, suggesting that the shunt behavior is well-explained by increasing minority carrier conductivity at higher laser powers and higher short-circuit currents.

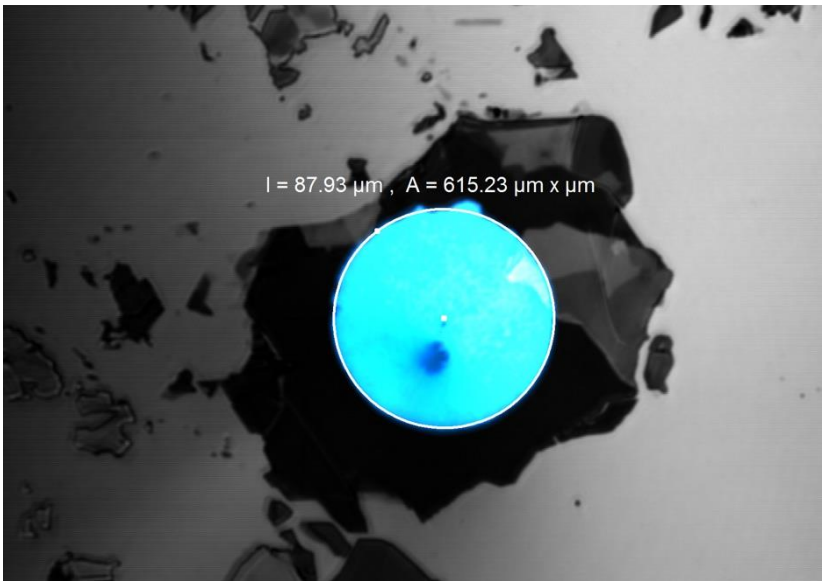
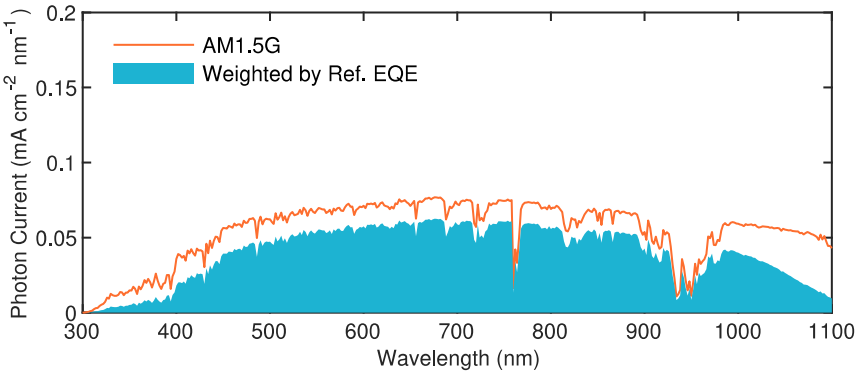
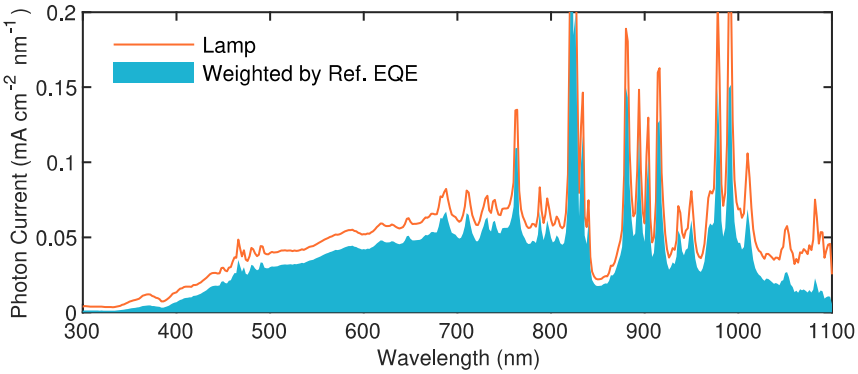


Fig. S3. Active area. The active area of the device presented in the main text is measured to be 615 μm^2.


AM1.5G
Weighted by Ref. EQE
Photon Current (mA cm-2 nm-1)
Wavelength (nm)
0
0.05
0.1
0.15
0.2
300
400
500
600
700
800
900
1000
1100

A


Lamp
Weighted by Ref. EQE
Photon Current (mA cm-2 nm-1)
Wavelength (nm)
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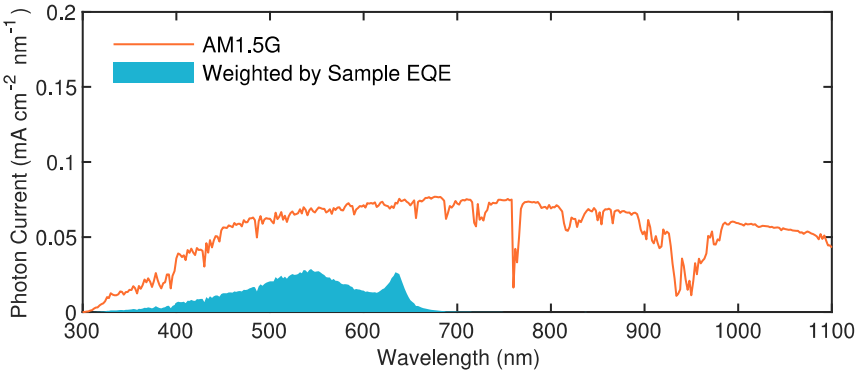
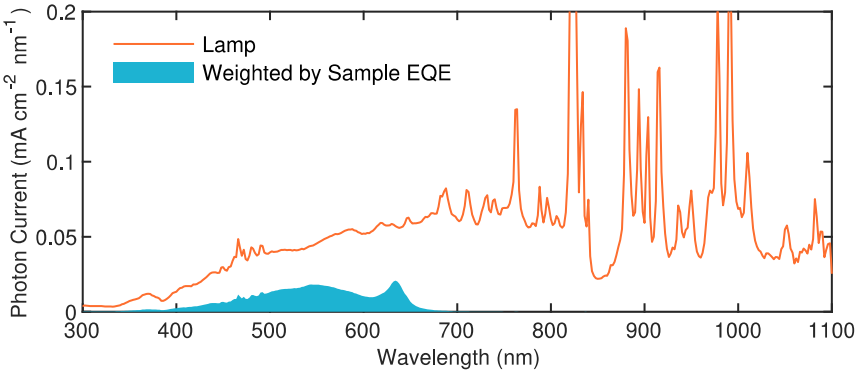
0.1
0.15
0.2
300
400
500
600
700
800
900
1000
1100

B


AM1.5G
Weighted by Sample EQE
Photon Current (mA cm-2 nm-1)
Wavelength (nm)
0
0.05
0.1
0.15
0.2
300
400
500
600
700
800
900
1000
1100

C


Lamp
Weighted by Sample EQE
Photon Current (mA cm-2 nm-1)
Wavelength (nm)
0
0.05
0.1
0.15
0.2
300
400
500
600
700
800
900
1000
1100

D

Fig. S4. Spectral mismatch. (**A**, **B**) Current generated under AM1.5G (A) and solar simulator lamp (B) for our silicon reference solar cell. We adjust the lamp intensity until these integrated currents are equal. (**C**, **D**) Same, but for the sample. We calculate the spectral mismatch factor M = 0.67 by dividing the integrated current in (D) by the integrated current in (C).

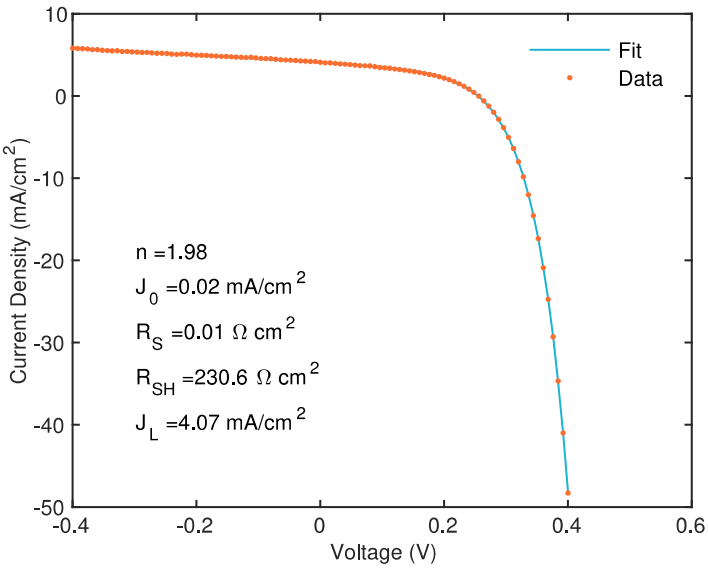
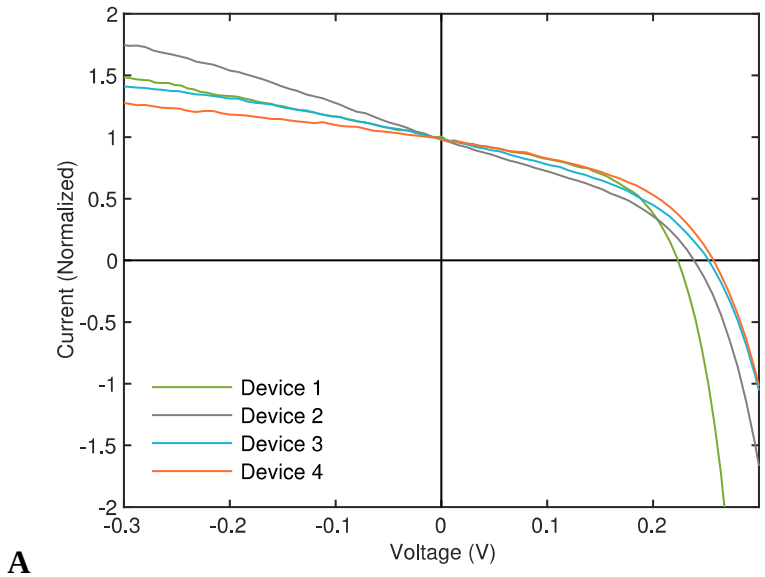
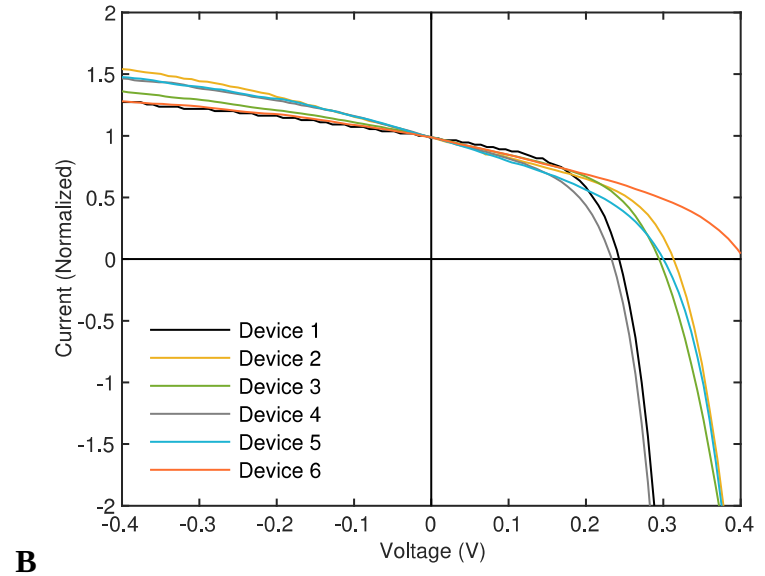


Fig. S5. Fitting for one-sun I-V curve. The diode equation with shunt and series resistances is used to extract the parasitic resistances and diode parameters.



A



B

Fig. S6. Reproducibility. (**A**) I-V curves taken from 4 different devices under one-sun illumination. V_{OC} is between 220 and 260 mV across all devices studied. (**B**) I-V curves taken from 6 different devices under illumination with a halogen lamp (~20 suns power density). V_{OC} is greater than 220 mV across all devices studied. In both one-sun and halogen lamp I-V curves, I_{SC} varies due to the different thicknesses and active areas across different devices, and therefore current is normalized by the short-circuit current value for easier comparison.

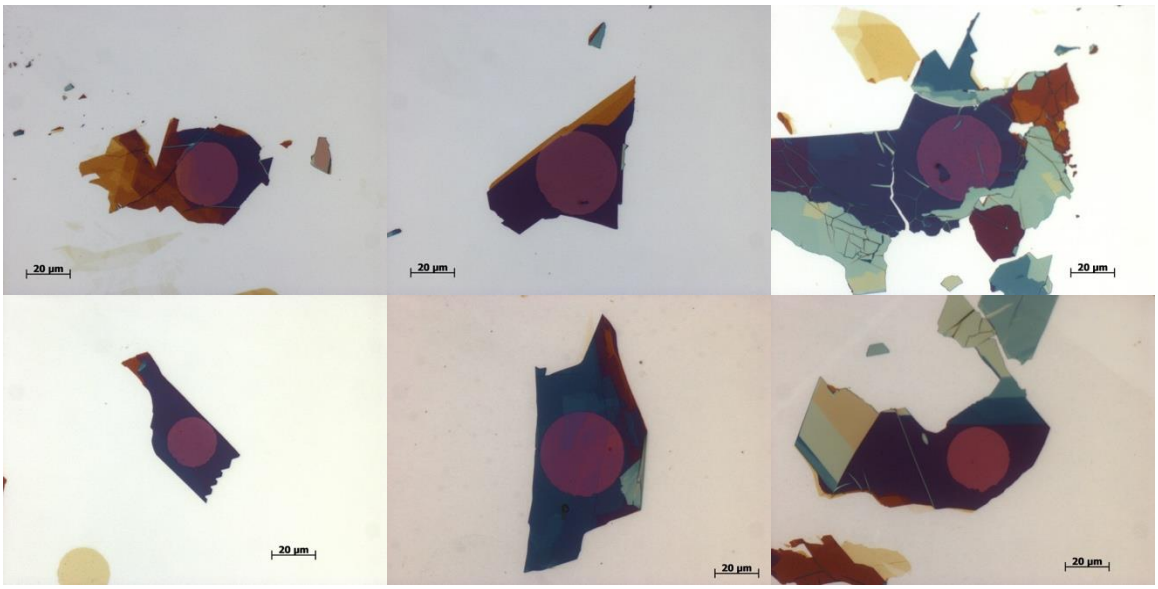


Fig. S7. Microscope images of other fabricated devices. The first 5 images are Devices 1-5 in fig. S6B.

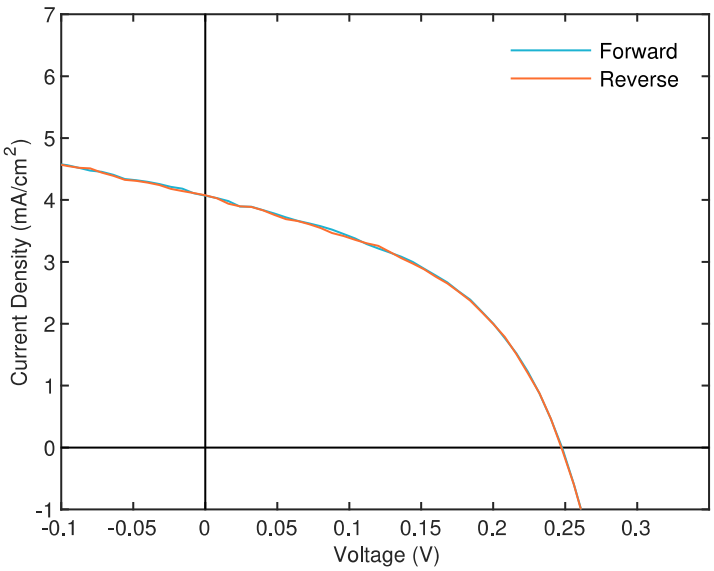
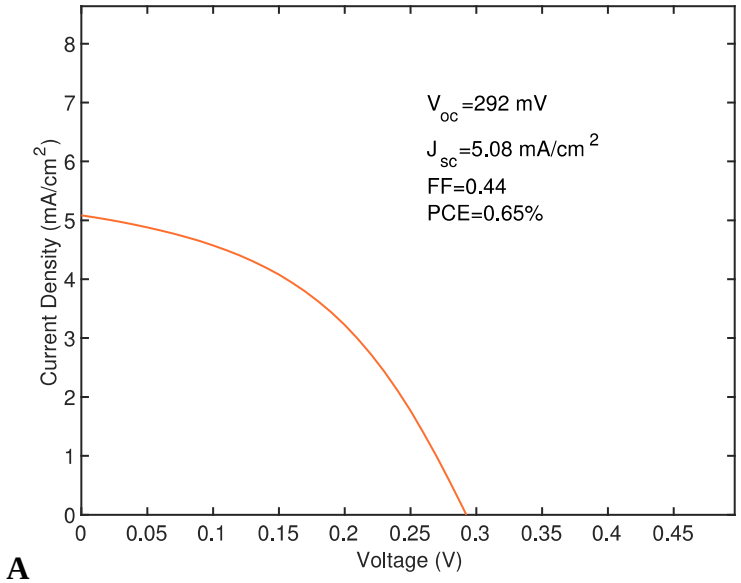
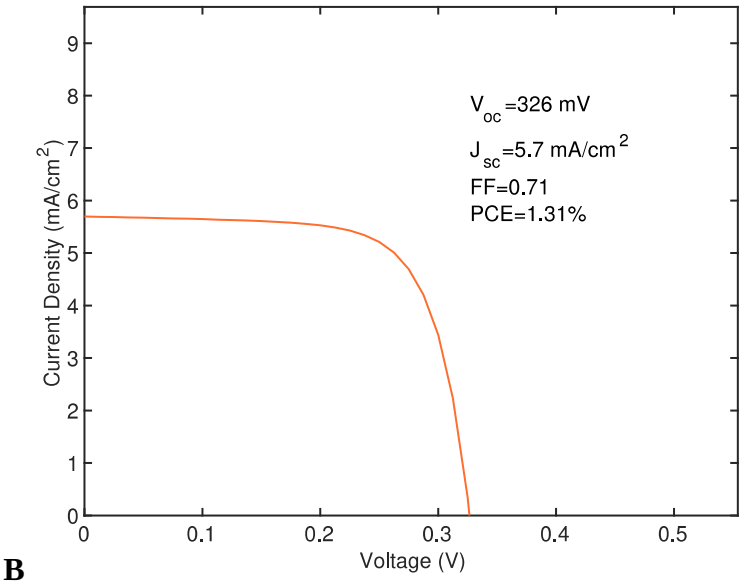


Fig. S8. Forward/backward scans. One-sun I-V curves swept in forward and reverse directions show no hysteresis.



A



B

Fig. S9. Matching simulations to experimental device. (**A**) An I-V curve with parameters close to the experimentally-observed I-V curve can be simulated if the Au work function is set to 4.6 eV, while all other parameters are kept the same. R_{SH} for this simulation, approximated by the inverse slope of the I-V curve at short-circuit, is 275 Ω cm^2. Au could have a lower effective work function due to contamination or thiol bonding at the Au/WS_2 interface. However, this would also limit the V_{OC} observed under laser illumination to ~400 mV and thus is not a complete explanation. (**B**) Increasing mobility reduces the V_{OC}, but does not change the shunt

resistance. Here, mobility is set to 1000 $cm^2 V^{-1} s^{-1}$ for both electrons and holes. R_{SH} for this simulation, approximated by the inverse slope of the I-V curve at short-circuit, is 2238 Ω cm^2.

Table S1. WS_2 parameters for device simulations.

Parameter	**Value**	**Source**
Bandgap	1.35 eV	Ref. (*36*)
Work function	4.62 eV	Ref. (*37*)
Electron effective mass	$0.63m_e$	Ref. (*39*)
Hole effective mass	$0.84m_e$	Ref. (*39*)
Doping	10^{14} cm^{-3}	HQ Graphene
Out-of-plane mobility (for holes & electrons)	0.01 cm^2/Vs	Ref. (*40–42*)
DC permittivity	6.7	Ref. (*38*)
Radiative recombination coefficient	1.64×10^{-14} cm^3/s	Calculated using optical constants in Ref. (*35*)
Shockley-Read-Hall Lifetime	611 ns	Calculated using PLQY from Ref. (*43–45*)