

Supporting Information

**All Inorganic Large-area Low-Cost and Durable
Flexible Perovskite Solar Cells Using Copper Foil as a
Substrate**

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Experimental

Preparing substrate and CuI layer growth: A 10-micron copper foil (99.999% purity) was cleaned by soap-deionized water solution, followed by ultrasonication at 50°C deionized water, ethanol, and isopropanol, and then immersed into 0.5M HCl solution in DI water. The washed foil dried by pure nitrogen air-gun followed by exposing with iodine gas in the vacuumed close vessel. In this regard, the clean foil placed in the glass chamber which connected to a small glass containing 400 mg iodine solid particles. At first, the connector between iodine chamber and the reaction chamber is kept close. By evacuating the reaction chamber up to 2 mTorr, the connector is opened. The iodination conducted at room temperature for 4 minutes followed by annealing the substrate at 100°C for 10 minutes

Synthesis of CH₃NH₃I: The CH₃NH₃I was synthesized by reacting 24 mL of CH₃NH₃I and 10 mL of HI in a 250 mL round-bottom flask at 0°C for 2 h with stirring. The precipitate was collected using a rotary evaporator through the careful removal of the solvents at 50°C. The as-obtained product was re-dissolved in 100 mL absolute ethanol and precipitated with the addition of 300 mL diethyl ether. After repeating this procedure for three times, the final CH₃NH₃I was collected and dried at 60°C in a vacuum oven for 24 h.

Deposition of pinhole-free perovskite layer: To deposit the uniform layer of perovskite layer on CuI compact layer, we employed spin coating of a solution of 460 mg/mL PbI₂ in DMF which previously filtered through a 0.2 μm syringe filter. Spin coating was carried out at 3000 rpm for 30 seconds followed by annealing at 75°C for 15 minutes. The perovskite layer was synthesized by spin coating of 40

mg/mL MAI in 2-propanol at 4000 rpm for 20 seconds with a retention time of 20 seconds. The perovskite layer was washed by spin coating of the layer by 200 μ L pure 2-propanol at final 10 seconds of spin coating. The final perovskite layer was annealed at 100°C for 5 minutes.

Synthesizing ZnO nanoparticles and thin film: For deposition of zinc oxide electron transport layer on the as prepared perovskite layer, a 5 nm crystalline zinc oxide nanoparticles solution in chloroform was used. ZnO nanoparticles were synthesized by the previous reported method ¹. In short, zinc acetate was dissolved in methanol with a small trace amount of deionized water at 60°C. The dissolved KOH in methanol was added into the previous solution while stirring. After reacting for 90 minutes, the solution cooled down to room temperature and kept overnight to precipitate ZnO nanoparticles. After decantation, the ZnO nanoparticles was washed by methanol twice to remove potassium and unwanted impurities. The washed ZnO nanoparticles dried at room temperature and redispersed in chloroform to form 40 mg/mL ZnO solution. The prepared solution was spin coated on the perovskite at 2000 rpm for 30 seconds followed by annealing at 70°C for 10 minutes.

Synthesizing silver nanowire: For rapid and massive synthesizing the long silver nanowires, as reported, a pattern free one-pot process was used ² with some modification. Briefly, 120 mg polyvinylpyridine (PVP 360000) was dissolved in 10 mL ethylene glycol (EG) at room temperature followed by adding 100 mg silver nitrate (AgNO_3) and stirring up to complete dissolving. The prepared solution was loaded by 240 μ M CuCl_2 in EG in two minutes. The prepared mixture was transferred into the preheated close vessel up to 130°C and reaction was conducted under a nitrogen atmosphere for 6 hours. Afterward, the acetone and 2-propanol was used to wash the precipitate by 6000 rpm for 10 min. This repeated twice for better

removal of colloidal Ag nanoparticles and PVP from Ag NWs. The final product was redispersed into 2-propanol.

Silver nanowire electrode: Silver nanowire electrode was created by direct spray coating of dispersed silver NWs in 2-propanol on the ZnO thin film through a 0.1 cm² stainless steel shadow mask followed by rod rolling for better surface contact of silver NWs with each other and ZnO layer.

Thin film and device characterization: To study the microstructure field emission scanning electron microscopy (FE-SEM, S4160 Hitachi Japan) was used. The phase structure and crystal size of films were also investigated by X-ray diffraction (XRD, Philips Expert- MPD). XRD was performed in θ - 2θ mode using Cu-K α with wavelength of 1.5439 Å radiation. All the XRD experiments were performed at grazing incident angle of 2°. The optical characteristics of deposited films were analyzed by UV–vis spectroscopy using the wavelength range of 190–900 nm. The photocurrent-voltage (I–V) characteristics of solar cells were measured under one sun (AM1.5G, 100mW/cm²) illumination with a solar simulator (Sharif solar simulator). Steady-state PL measurements were acquired using a Varian Cary Eclipse (USA) fluorescence spectrometer.

Table S1. J-V characteristics of the prepared device in different days of working

Working time (days)	V_{oc}(V)	J_{sc} (mA/cm²)	FF	PCE (%)
0	0.958	22.50	0.594	12.80
5	0.95	22.6	0.57	12.36
10	0.95	22	0.56	11.79
15	0.94	21.8	0.56	11.47
20	0.940	21.6	0.55	11.16
25	0.93	21.2	0.53	10.44
30	0.93	20.6	0.54	10.34
35	0.93	20.1	0.54	10.09
40	0.92	19.9	0.53	9.70
45	0.92	19.74	0.52	9.44
50	0.92	19.68	0.54	9.77
55	0.92	19.53	0.53	9.52
60	0.92	19.49	0.52	9.32

Table S2. J-V characteristics of the prepared device in different bending cycles

Bending Cycles (times)	V_{oc}(V)	J_{sc} (mA/cm²)	FF	PCE (%)
0	0.958	22.50	0.594	12.80
100	0.958	21.2	0.57	11.6
200	0.95	20.3	0.57	11
300	0.947	20	0.57	10.8
400	0.94	19.6	0.56	10.5
500	0.935	19.2	0.55	10
600	0.935	18.8	0.55	9.8
700	0.93	18.7	0.54	9.5
800	0.928	18.5	0.54	9.4
900	0.924	18.3	0.53	9
1000	0.92	18.2	0.53	8.9

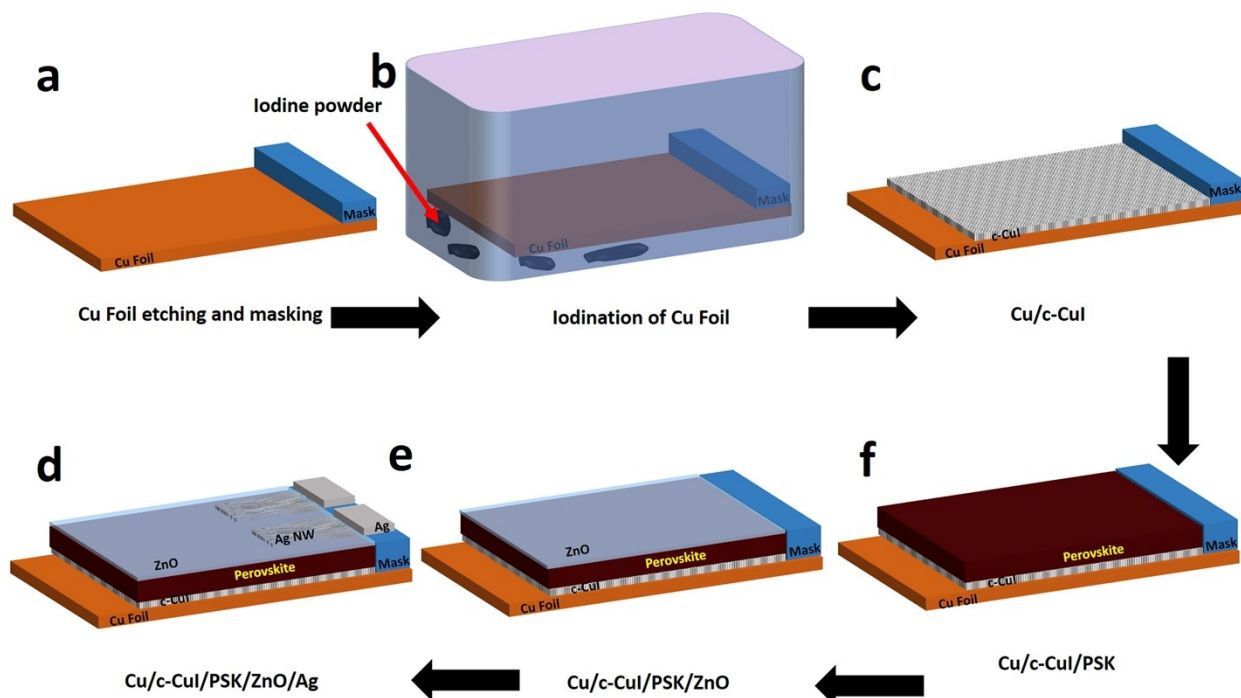


Fig. S1. Fabrication schematic of all inorganic perovskite solar cells low temperature.

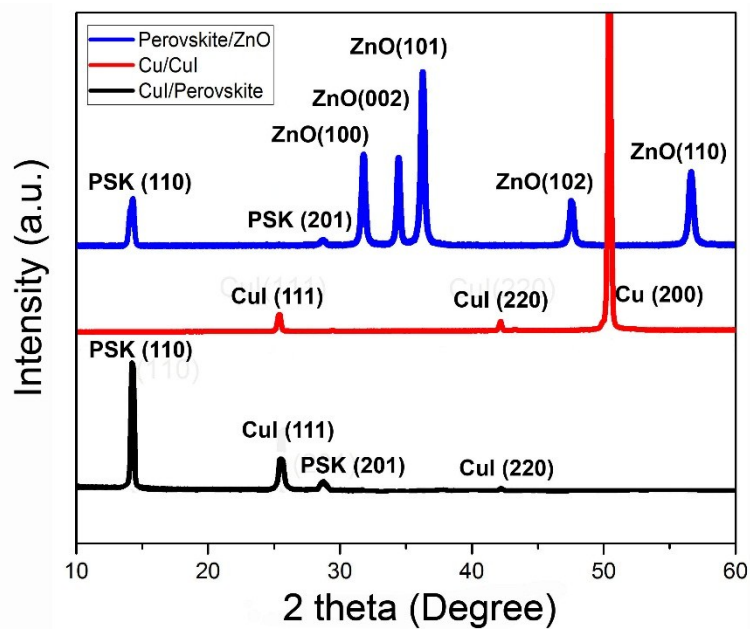


Fig. S2. Grazing incident XRD patterns of grown CuI on Cu foil, deposited perovskite layer

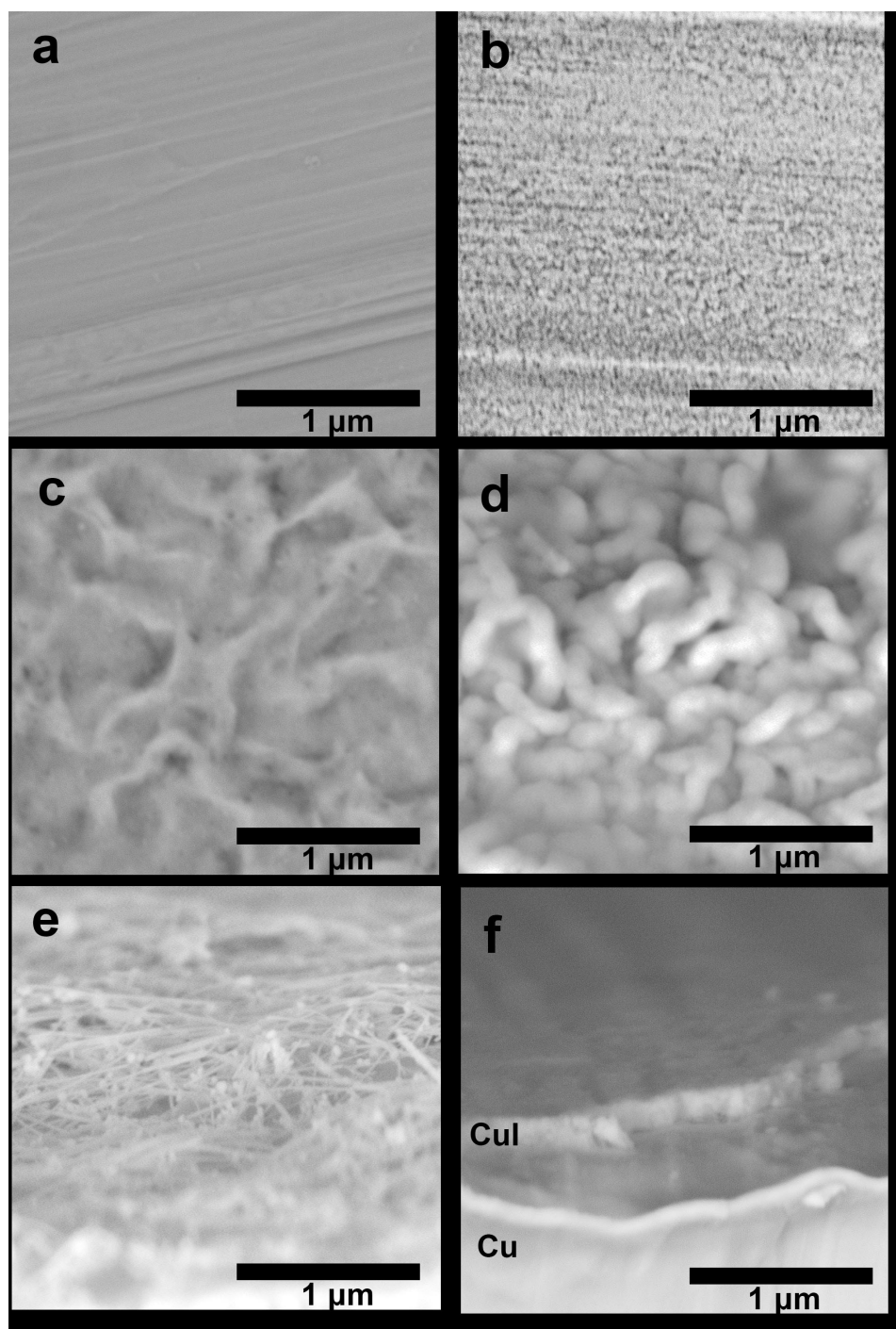


Fig. S3. FE-SEM images of copper foil (a), grown CuI on the Cu foil (b), deposited perovskite layer by 40 mg/mL MAI solution in 2-propanol (c), deposited ZnO thin film on perovskite by spin coating of

40mg mL⁻¹ dispersed ZnO nanoparticles in chloroform (d), tilted image of spray coated and compressed Ag NWs on the ZnO thin film (e), and cross section of grown CuI on the Cu foil (f).

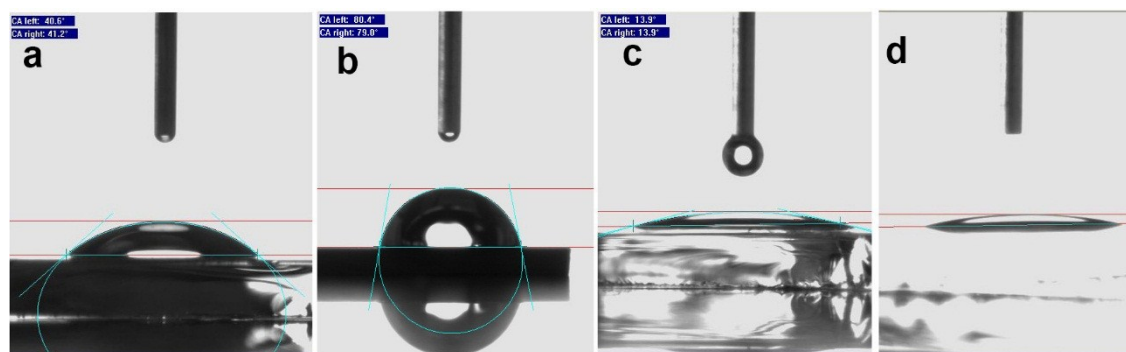


Fig. S4. Contacting angle of 460 mg/mL PbI₂ in DMF with glass (a), NiO (b), TiO₂ anatase (c), and CuI (d) substrates.

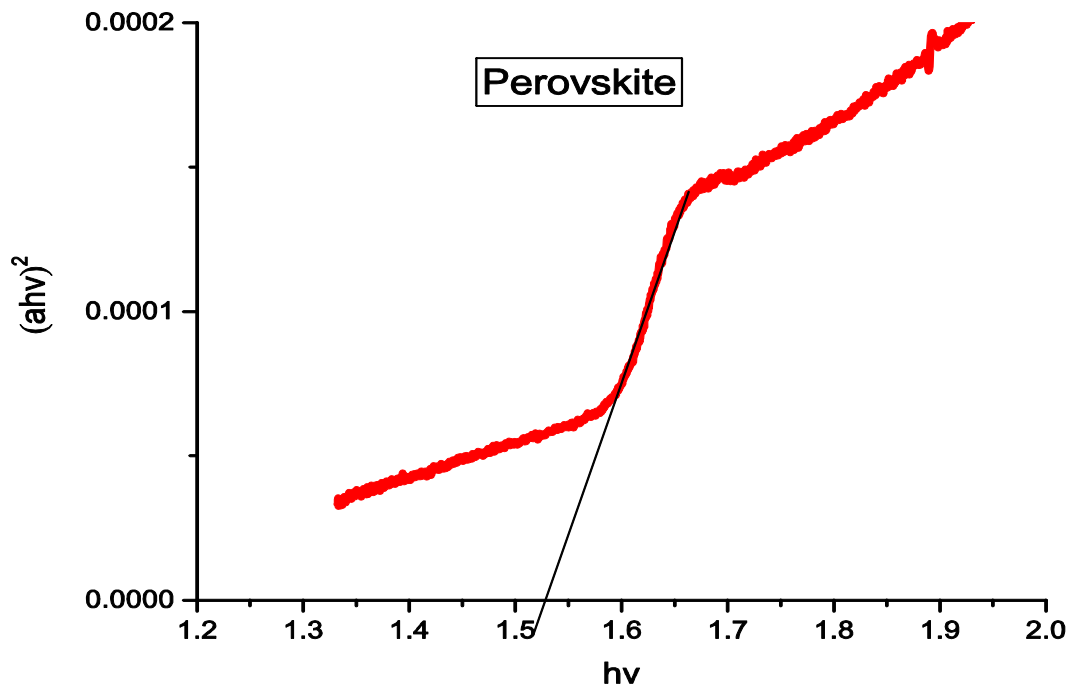


Fig. S5. $h\nu$ - $(ah\nu)^2$ curve of deposited Perovskite layer

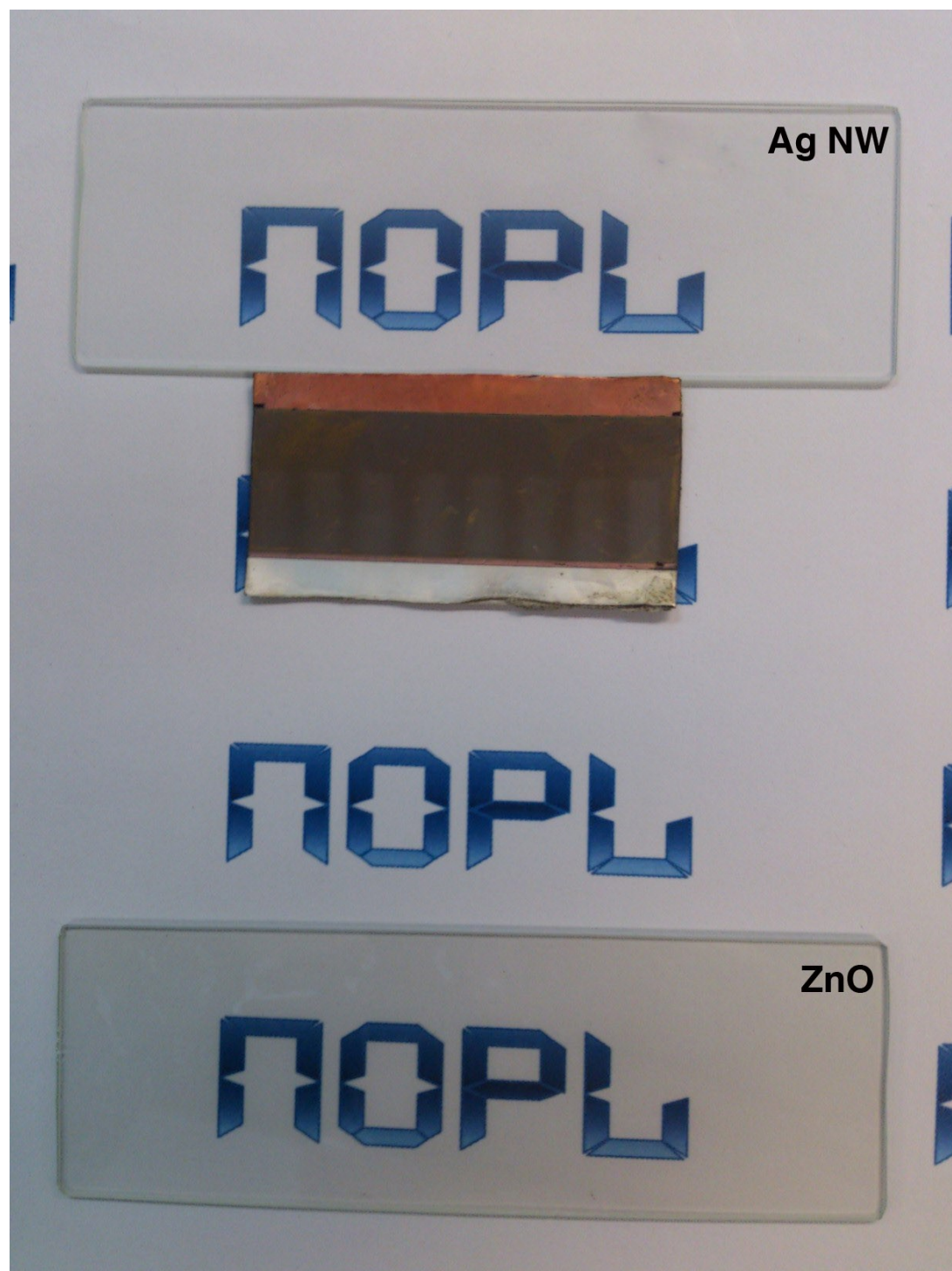


Fig. S6. Photograph of deposited silver NWs by spray coating (up), Cu/CuI/Perovskite/ZnO/Ag NW/Ag device (middle), and deposited ZnO thin film on glass substrate (down).

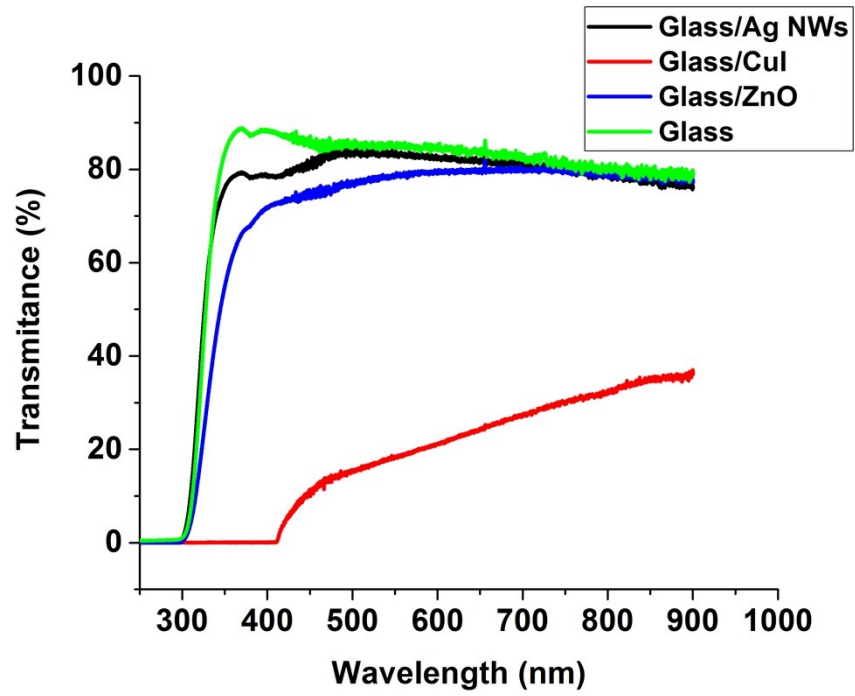


Fig. S7. Transmittance spectra of deposited thin films on glass substrate

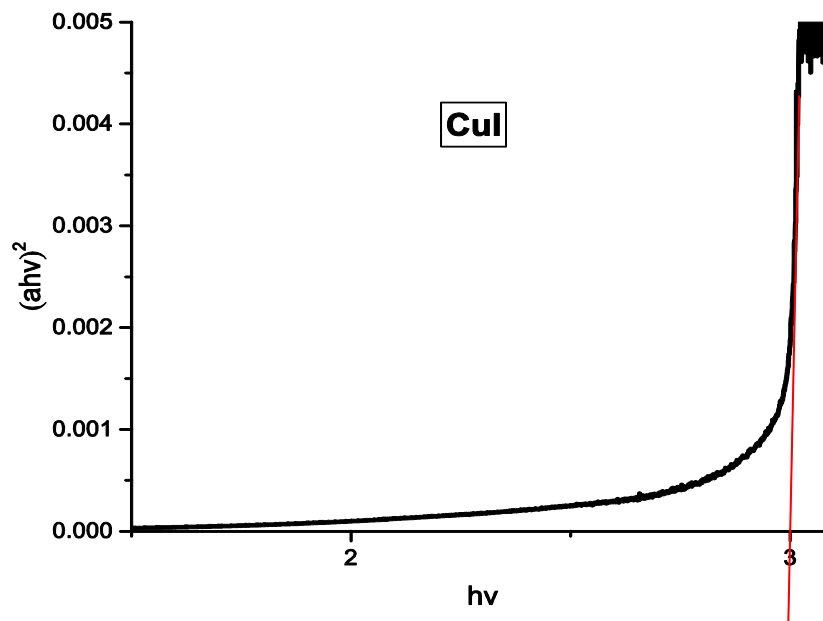


Fig. S8. $h\nu - (\alpha h\nu)^2$ curve of CuI thin film prepared by gas-solid formation of CuI on Cu foil.

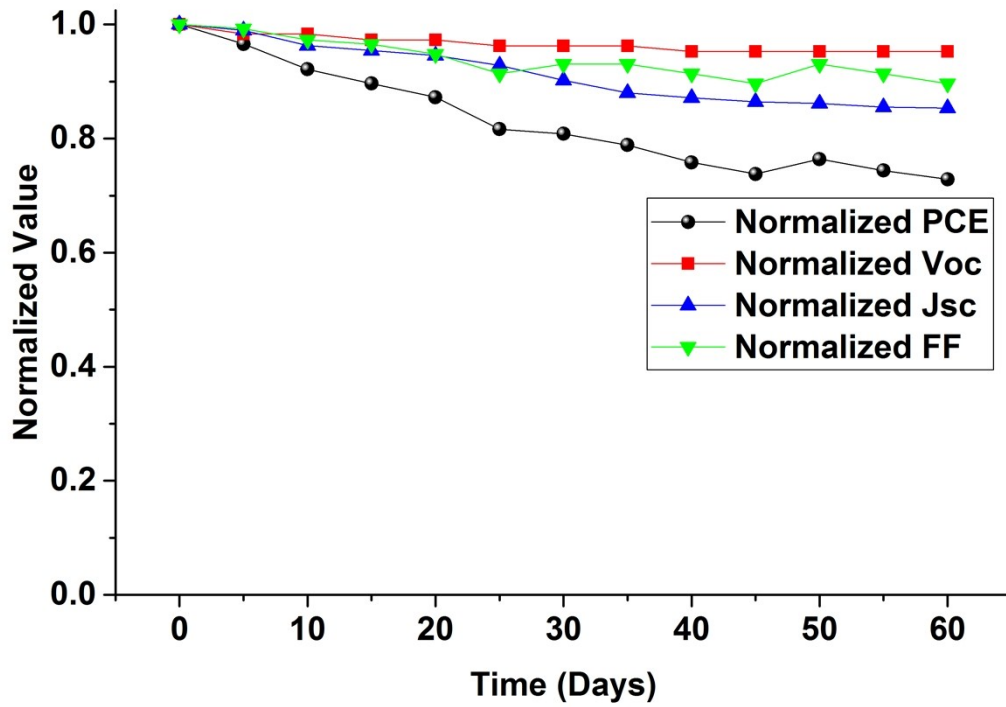


Fig. S9. Device performance durability in 60 days at 25°C and $28 \pm 2\%$ moisture under dark retention and analyzing under one sun (AM1.5) illumination.

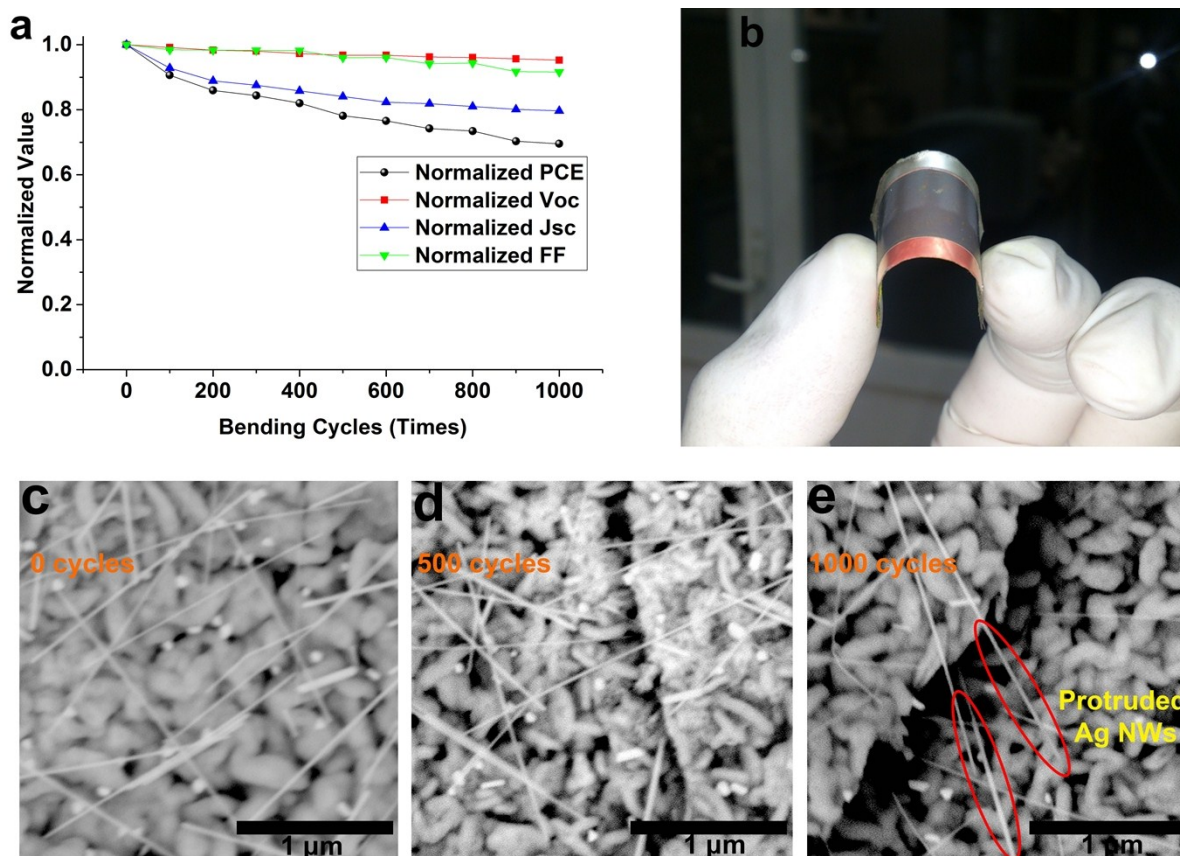


Fig. S10. Durability of device at different bending cycles (a), the photograph of bent device at 180°(b), and contacting of silver NWs with ZnO surface at different bending cycles of zero (c), 500 (d), and 1000 (e).

1. J. You, L. Meng, T.-B. Song, T.-F. Guo, Y. Yang, W.-H. Chang, Z. Hong, H. Chen, H. Zhou, Q. Chen, Y. Liu, N. De Marco and Y. Yang, *Nat Nano*, 2016, **11**, 75-81.
2. J. Jiu, T. Araki, J. Wang, M. Nogi, T. Sugahara, S. Nagao, H. Koga, K. Suganuma, E. Nakazawa, M. Hara, H. Uchida and K. Shinozaki, *J. Mater. Chem. A.*, 2014, **2**, 6326-6330.