Supporting Information

High-Performance Deformable Photoswitches with p-Doped Graphene as Top Window Electrode

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Experimental Section

Materials: Cu foils are from Alfa Aesar, item No.13382. PEN substrates are from DuPont (Melinex® 506). PET substrates are from chemplex (SpectroMembrane®). PVDF-HFP is from Sigma-aldrich (average M_n ~130,000). PMMA (solution) is from micro resist technology GmbH (product number: 950 PMMA C2). P3HT is from Sigma-aldrich and PCBM is from American Dye Source, Inc.

CVD growth of SLG: SLG were grown on 25-μm thick Cu foils (cut into 3.5 by 3.5 cm squares) in a hot wall furnace. A typical growth process is: (1) Load the quartz tube with the Cu foil, evacuate the tube down below a pressure of 5×10^{-3} mTorr. (2) H_2 (purity 99.999%) was supplied at 100 sccm and the pressure was maintained at ~1 mTorr. (3) The samples were heated to 1020°C within 40 min and then annealed at 1020 °C for another 40 min. (4) 20 sccm of CH_4 (purity>99.9%) was introduced for 20 min. (5) The furnace was cooled down to room temperature (CH_4 was turned off when the temperature was below 600 °C and H_2 was turned off when the temperature was below 200 °C).

Measurements: Raman spectra were obtained by a SENTERRA dispersive Raman microscope (Bruker Corporation). The laser wavelength was 532 nm and the power was 2mW. The R_S of the graphene films were measured by a standard four-point-probe system using a Keithley 2700 Multimeter (probe spacing: 0.635 mm, R_S =4.532 V/I). XPS and UPS were performed in a Kratos AXIS ULTRA DLD ultrahigh vacuum

photoemission spectroscopy system with a base pressure better than 2×10^{-9} Torr, and He I (21.22 eV) as the excitation source. Samples were -9 V biased for the observation of secondary electron cutoff. The Fermi level of the system was referenced as the zero binding energy in all the UPS spectra, and was calibrated by determining the Fermi level edge of a sputtered clean gold film. The photoresponse of the photoswitch was measured in a glove box under nitrogen atmosphere using a Keithley 4200 SCS (with probe station). The light of a Nikon microscope (SMZ1000, ca. 20 mW/ cm^2) was used as the light source. All photos of the devices were taken outside the glove box.

Detailed procedures for the fabrication of photoswitch arrays. (1) Aluminum (60 nm) was deposited on the SLG/Cu foil through a shadow mask to partially protect the graphene film. (2) The substrate was then exposed to an O_2 plasma for ~5 seconds at 100 W rf power to remove the unprotected areas. (3) The patterned SLG/Cu foil was immersed in 10 % HNO_3 solution to remove the sacrificial Al. The sample was then rinsed with DI water for 3 times and blow dried. (4) PMMA or PVDF-HFP (1g PVDF-HFP in 40 ml acetone) solution was spin coted on the patterned SLG/Cu foil. PMMA was spin coated at 2K RPM for 90 s and PVDF-HFP was spin coated at 10K RPM for 60 s. Samples were baked at 120 ^oC for 60 s after spin coating. (5) Cu foil was etched away by $Fe(NO_3)_3$ solution. After that, the etchant solution was replaced by DI water for several times, leaving the polymer supported SLG pattern floating on water surface. (6) Al (60 nm) was deposited on clean PEN or PET substrates through the same shadow mask. (7) Blend of P3HT:PCBM (10 mg P3HT and 8 mg PCBM dissolved in 1 ml dry chlorobenzene and stirred overnight at 80^oC in glove box) was spin coated on the substrate at 800 RPM for 60 s. (8) The polymer supported SLG patterns were vertically

aligned with the bottom Al electrodes and transferred to the substrate. (8) The devices were blow dried quickly and moved into a glove box, where annealing at 150 ^{o}C for 20 min was carried out on a hot plate before test.

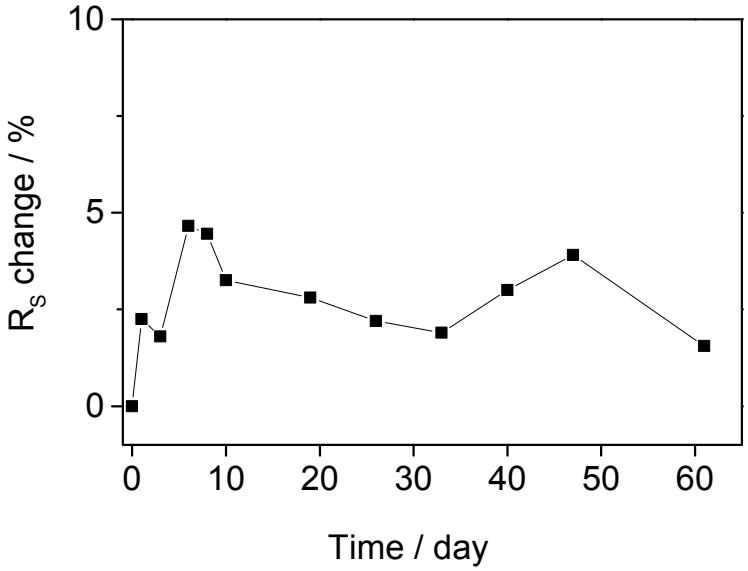

Figure S1. Change of sheet resistance of FG film as a function of time. Each R_s value was averaged on 9 random positions of the sample. The FG film was stored in ambient air during the measurements.

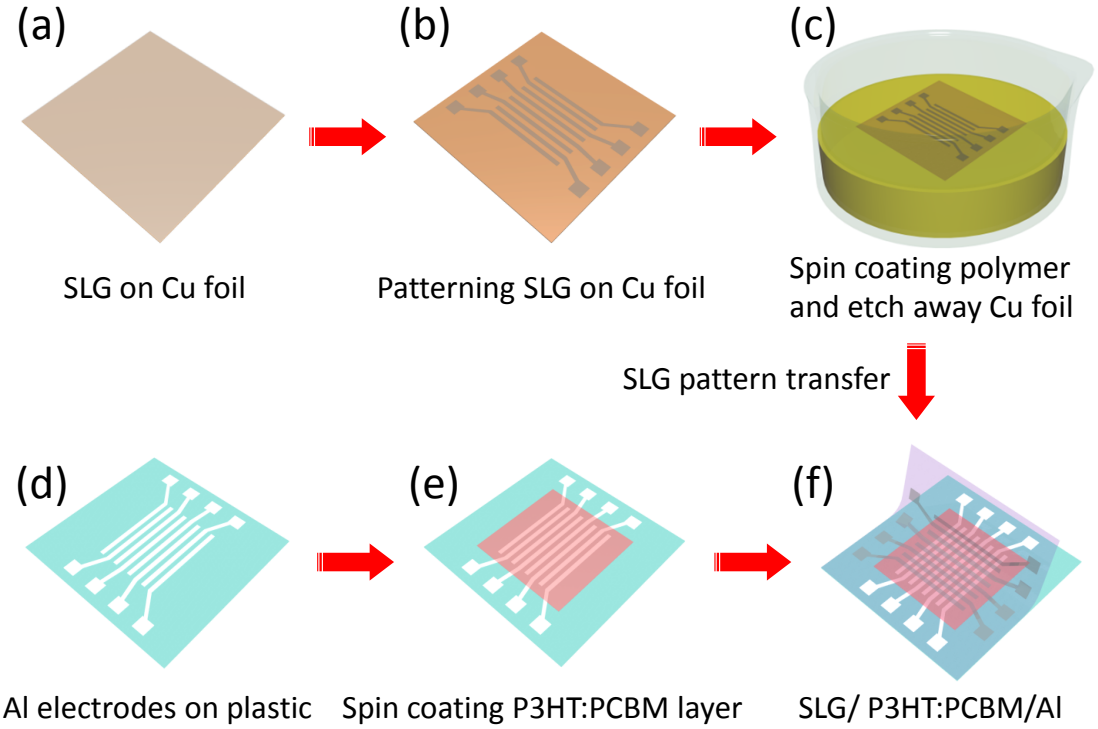

Figure S2. The main processes for the fabrication of the photoswitch array.

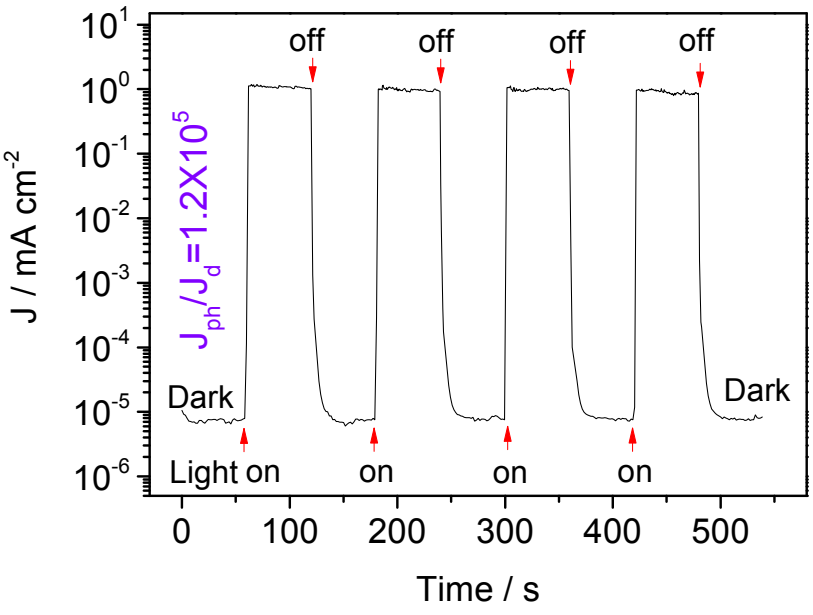

Figure S3. Photoswitch using PMMA transferred SLG as the window electrode.

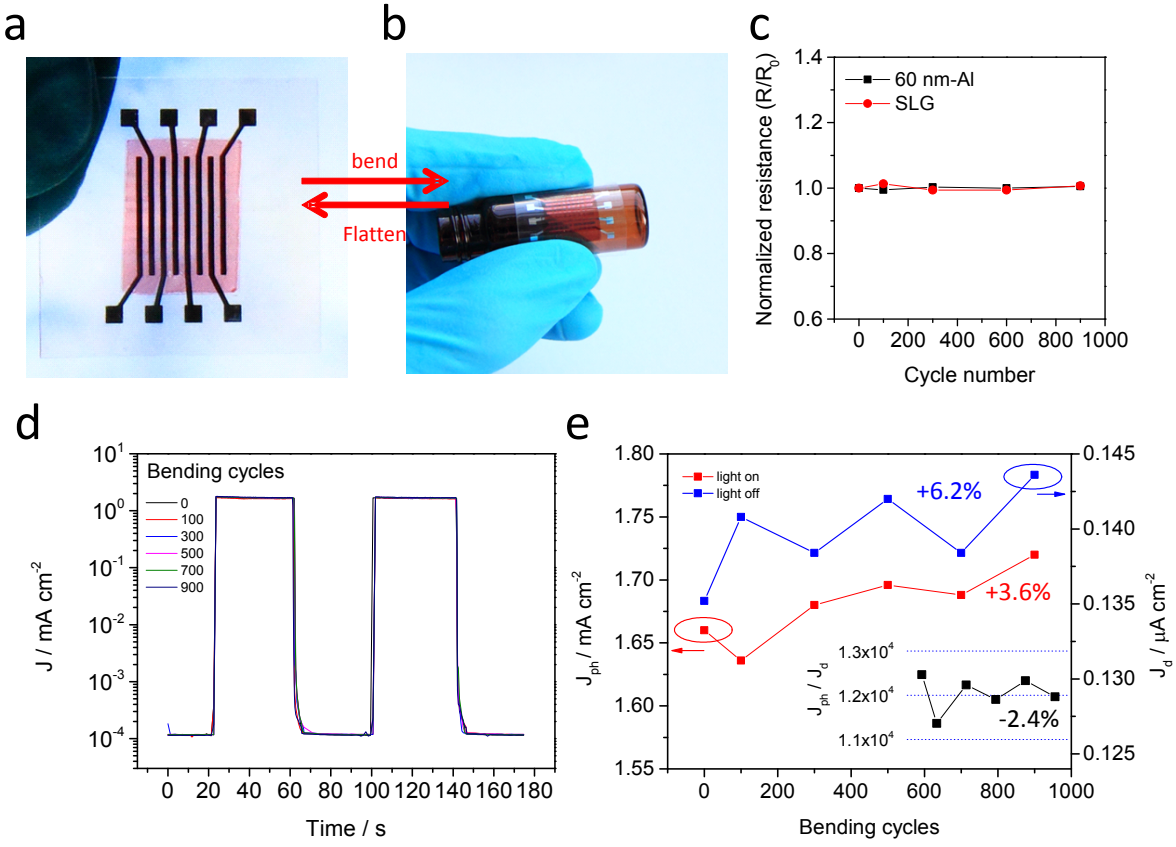

Figure S4. Flexibility of the photoswitches. (a, b) Photographs of photoswitches in the flat state and under bending. (c) Normalized sheet resistance change of the SLG and Al electrode in the bending and flattening cycles. (d) Photoresponse of FG- photoswitch after bending to a radius of ~6 mm for 100, 300, 500, 700 and 900 cycles. (e) The change of photo current and dark current. The inset shows the change of on/off ratio. A bias voltage of 20 mV was applied during this measurement.

a

Flat ×10⁴	after deformation ×10⁴
6.9	6.6
7.8	5.6
6.8	5.7
1.6	5.4
6.4	5.3
6.2	4.9
9.1	6.8
7.1	6.3
6.9	7

b

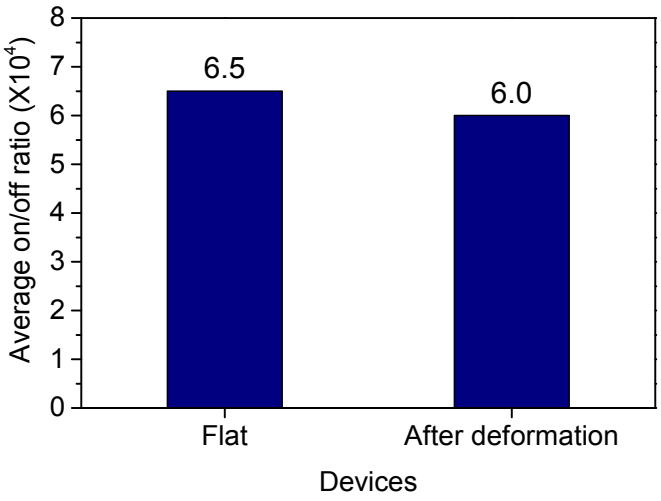

Figure S5. (a) On/off ratios of 9 randomly selected devices before and after deformation. (b) The average on/off ratio change.

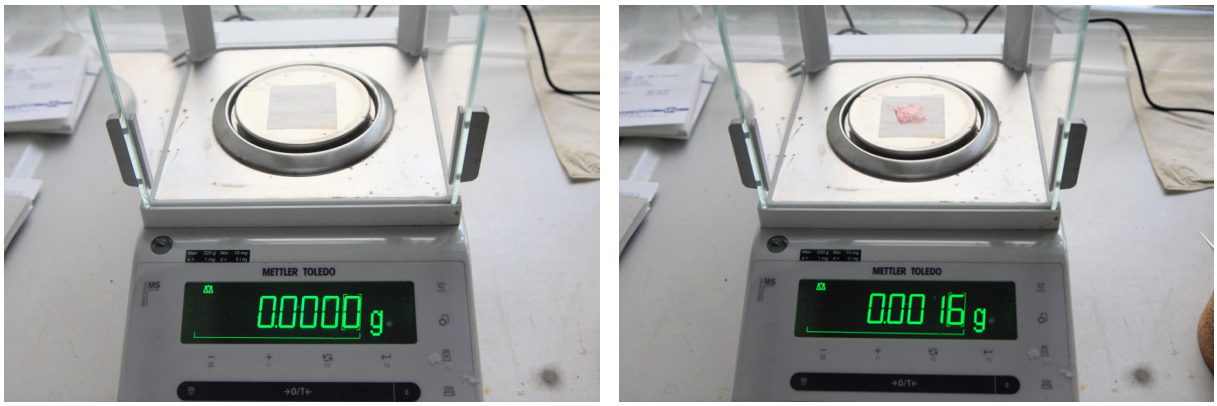

Figure S6. Weight of the 8×8 photoswitch arrays on PET substrate.

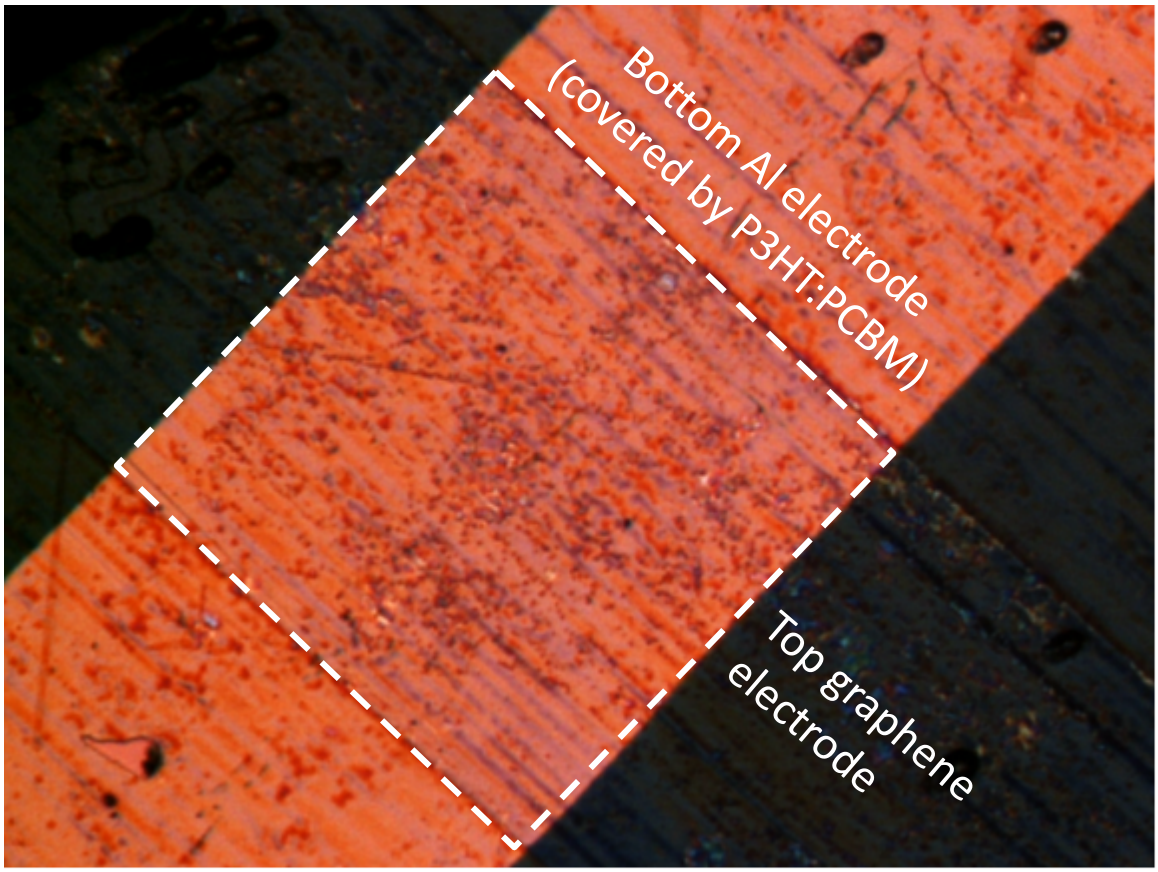

Figure S7. Optical microscopic image of one device. The white square in the centre indicates the active area of the photoswitch (250 by 250 μm).