Supporting information

High-Sensitivity, Highly Transparent, Gel-Gated MoS₂ Phototransistor on Biodegradable Nanopaper

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S1. Fabrication process of photodetector

The whole process of fabrication of multilayer MoS_2 based photodetector is conducted in clean room. The fabrication procedure includes five major steps: prepare multi-layer MoS_2 , transfer material, deposit electrodes, drop electrolyte and seal top nanopaper.

- In the first step, we are about to prepare multilayer MoS₂ nanosheets. Scotch type is used for the micromechanical cleavage technique to exfoliate the MoS₂ nanosheets.
- After the exfoliation process, we need to transfer the few layers MoS₂ nanosheets from the scotch type onto the nanopaper substrate. The smooth surface of nanopaper is selected, and we put scotch type with MoS₂ onto the surface of nanopaper. By hands pressing several times, the MoS₂ nanosheets will adhere on the surface of nanopaper through Van de Waals interaction.
- Then, we put nanopaper with MoS₂ nanosheets under optical microscope, and locate the ideal piece of MoS₂ nanosheets, which has the perfect size and thickness for making device. Then we align the ideal piece of MoS₂ nanosheets with shadow mask. 50 nm gold electrodes are deposited onto the MoS₂ through e-beam evaporation system.
- After that, 0.1 µL~0.15 µL ion liquid gel-electrolyte is dropped onto the as fabricated device. The liquid electrolyte is supposed to cover the whole MoS₂ nanosheets and part of source, drain and gate electrodes. The electrolyte in this phototransistor has another function: adhesive layer. It adhere the bottom and top nanopaper.
- Before the liquid electrolyte becoming dry, another layer of nanopaper is coated onto the as-fabricated device. This top layer nanopaper is supposed to be large enough to cover the entire MoS₂ nanosheets and part of the electrodes in direct contact with MoS₂, while small enough at the same time to expose the bonding pads.

S2. Detail of electrolyte

The recipe of electrolyte we used is a 1:10 ratio in weight of $LiClO_4$:PEO. We learned the electrolyte formula from reference 35 (Nano Lett. 2015, 15, 4295–4302), which worked nicely for our devices. $LiClO_4$ and PEO are purchased from Sigma Aldrich. The average molecular weight of the PEO in electrolyte is 200,000.

- Firstly, we weight 0.1 g LiClO₄ and 1 g PEO through high precision balance.
- Then, mix the LiClO₄ and PEO powder together, and put them into a bottle.
- Add 20 mL methanol into the bottle and keep 50 °C about 10 minutes for the power dissolved.
- Cool down the liquid to room temperature. Then seal the bottle and keep stirring for more than 24 hours to get the uniformly dispersed ion liquid gel-electrolyte.

The volume of liquid electrolyte we drop on the device is around 0.10μ L. The drop spreads into an area of approximately 3 mm², the thickness of the electrolyte is therefore estimated to be ~30nm for a typical device.

S3. IV curve of conductance of the nanopaper, and nanopaper sheet resistivity

The IV curve of the nanopaper is shown below. The resistance of nanopaper is tested as 5700 M Ω . Through the experiment data, we can estimate the sheet resistance of nanopaper based on the following function:

$$\rho = \frac{SR}{L} = \frac{hWR}{L}$$

For the nanopaper we use in this paper, R is resistance (5700 M Ω in resistance test), h is thickness (1µm), W is the width (1mm), L is the length (2.5mm). The sheet resistance of nanopaper is around 2.3E13 Ω ·cm, which can be considered as an insulator (sheet resistance higher than 1E10 Ω ·cm). This sheet resistance of nanopaper is slightly lower than SiO₂ (1E16 Ω ·cm).



Figure S3. *I-V* characteristic of pure nanopaper

S4. The absorbance/transmittance spectrum of the electrolyte

We were not able to make a direct transmittance measurement on the liquid electrolyte with our experimental setup. Nevertheless, the number (t_e) can be derived from the transmittance of a single sheet of nanopaper (t_p) and the nanopaper-electrolyte-nanopaper sandwich structure (t_s) : $t_e = t_s/t_p^2$. We have plotted all the three spectra, t_s , t_p and t_e in the figure below. According to our measurements, the transmittance of the electrolyte itself is greater than 95% across the entire spectral range from 400 to 1100 nm.



Figure S4. The transmittance spectrum of the electrolyte

S5. Electrical characterization of the device of figure 5 before and after bending

According to our test results, the electrical characteristics of the device are almost the same before and after bending. We have included the measurements shown below. The device was bended 135 degree in all measurements. The performance ((A). on-off ratio, (B) mobility, (C) photo responsivity, and (D) photo current) were essentially the same as flat devices. The numbers were collectively within 3.6%.



Figure S5 I_{ds} - V_{ds} character of photodetector before and after bending in Fig. 5D

S6. Detail of fitting the parameter in Fig. 4A

The fitting function and parameter are shown in table S6. The relationship between R and E can be modeled by a power law as:

 $R \sim E^{\beta - 1}$

In the table below, the parameter b is β -1 in the equation above. So the parameter β can be estimated as 0.26.

Model	Allometric1		
Equation	y = a*x^b		
Reduced Chi-Sqr	0.07492		
Adj. R-Square	0.96837		
		Value	Standard Error
Responsivity	а	4.3207E-7	2.1574E-7
	b	-0.98712	0.03119

Table S6. The calculation detail for parameter β

S7. Calculation of the mobility.

The transfer curve below exhibits a linear triode regime. From the data presented, the field-effect mobility μ of the few layers MoS₂ phototransistor can be estimated based on the following equation:

$$\mu = \frac{L}{W} \times \frac{\varepsilon_0 \varepsilon_r}{d} \times \frac{1}{V_{sd}} \times \frac{dI_{sd}}{dV_g}$$

Where *L* and *W* are the channel length and width, respectively (here $L/W = 1.6 \ \mu m/8 \ \mu m$), ε_0 is 8.854E-12 Fm⁻¹, ε_r for electrolyte is estimated as 4, and *d* is the thickness of top gate dielectric (30nm). The value of the charge carrier mobility μ is estimated to be 150 cm²V⁻¹s⁻¹, which is comparable with the previous report about multilayer MoS₂ electrolyte FET devices. (Reference 44 in manuscript: ACS nano 2013, 7(5): 4449-4458)



Figure S7. Transfer curve of mobility

S8. Response time of phototransistor

We have tested the response time of our phototransistor (shown below). It is estimated to be 25s, longer than the state of the art phototransistor (usually within 10s). (reference 31 in manuscript: *Nat. Nanotech.*, 2013, **8**, 497-501). In general, the response speeds of electrolyte gated devices are averagely lower compared to those with solid gates (ref. 43: ACS nano 2013, 7(5): 4449-4458). The reason is likely associated with redistribution of mobile ions in the electrolyte that limits the rate of current increase.

