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

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23 **ABSTRACT:**

24 Transition metal dichalcogenides hold great promise for a variety of novel electrical, optical and
25 mechanical devices and applications. Among them, molybdenum disulphide (MoS₂) is gaining
26 increasing attention as the gate dielectric and semiconductive channel for high-performance field
27 effect transistors. Here we report on the first MoS₂ phototransistor built on flexible, transparent
28 and biodegradable substrate with electrolyte gate dielectric. We have carried out systematic
29 studies on its electrical and optoelectronic properties. The MoS₂ phototransistor exhibited
30 excellent photo responsivity of ~1.5 kA/W, about two times higher compared to typical back-
31 gated devices reported in previous studies. The device is highly transparent at the same time with
32 an average optical transmittance of 82%. Successful fabrication of phototransistors on flexible
33 cellulose nanopaper with excellent performance and transparency suggests that it is feasible to
34 achieve an ecofriendly, biodegradable phototransistor with great photoresponsivity, broad
35 spectral range and durable flexibility.

36

37 **KEYWORDS:** Molybdenum Disulfide phototransistor, Nanopaper substrate, Flexible
38 electronics, Biodegradable electronics, Gel-electrolyte gating

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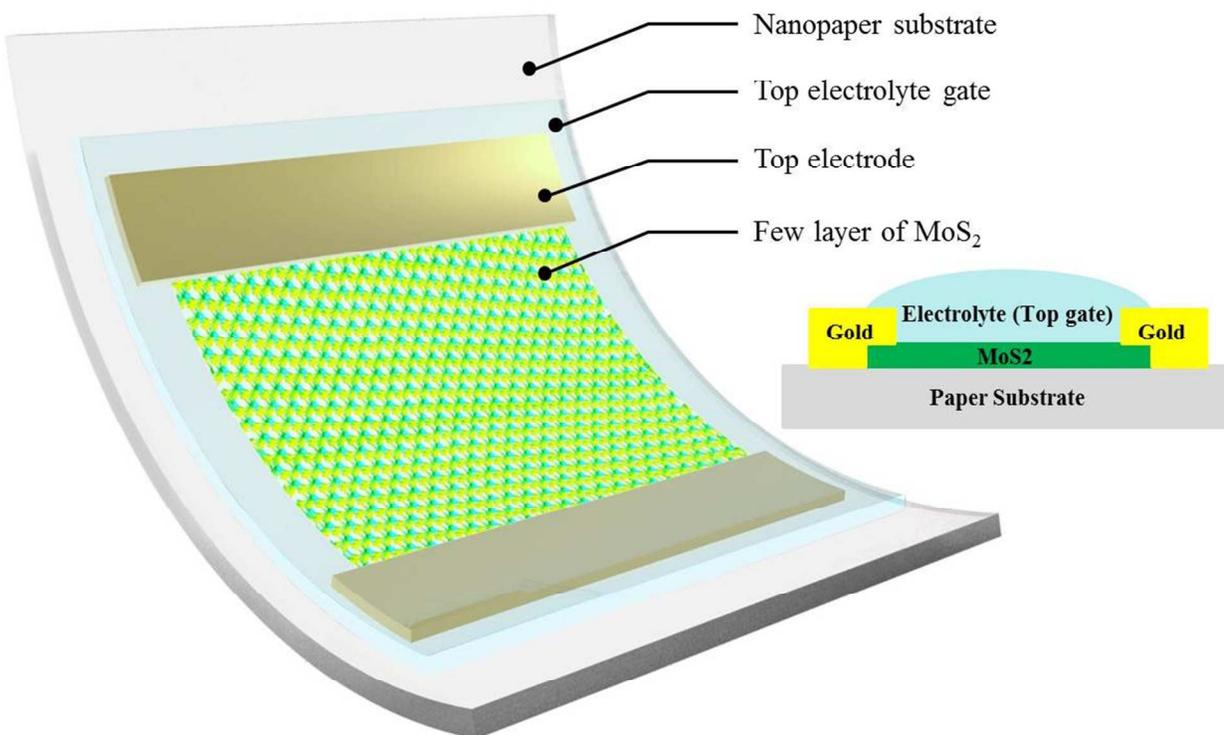
41 **Main text:**

42 Printed electronics with flexibility has attracted tremendous interests in recent years.¹⁻⁴

43 Conventional electronics are made with a glass or plastic substrate. Glass substrates can endure
44 very high handling temperature² but provide poor flexibility. Plastic substrates are transparent
45 and flexible,⁵ but they are not environmentally friendly and may take hundreds of years to
46 decompose. Recently, ultra-smooth transparent nanopaper with optical transmittance over 90%
47 has been developed for flexible electronics.⁶⁻⁹ Nanopaper is made from the same natural wood
48 pulp material as in paper, while consisting of much thinner nanofibrillated cellulose (NFC) fibers
49 of 5 to 10 nm in diameter.¹⁰⁻¹² The wood fibers are treated with (2,2,6,6-tetramethylpiperidin-1-
50 yl)oxyl (TEMPO)^{13, 14} to convert the hydroxyl groups to sodium carboxylate groups for
51 improved packing density.¹⁵⁻¹⁸ The fibers are then disintegrated by high pressure mechanical
52 homogenizer.⁹ The densely packed nanofibers leave minimal amount of air trapped in the paper
53 and give rise to very high transparency. Their ultra-small diameter also makes the paper surface
54 sufficiently smooth to be used as the supporting substrate for a variety of functional nano-
55 devices. Furthermore, the use of biodegradable natural wood pulp ensures the device is
56 environmentally friendly at the same time.¹ In a typical CMOS chip, the functional section is
57 formed by only a small portion of the chip, whereas the supporting substrate comprises more
58 than 99% of the semiconductor materials.¹⁹ These ecofriendly transistors can help conserve non-
59 renewable natural resources by replacing toxic semiconductor materials with biodegradable
60 nanopapers.²

61 Phototransistor is essentially a light-sensitive field effect transistor (FET) that transduces
62 incoming photo energy to electrical current. Phototransistors based on two dimensional (2D)
63 materials have become increasingly popular in recent years.²⁰⁻²² Among them, MoS₂ has received
64 particular interest due to its unique electrical and optical properties.²³⁻²⁶ In fact, devices made of
65 bulk MoS₂ have emerged decades before.²⁷⁻²⁹ Its special band structure, mechanical flexibility

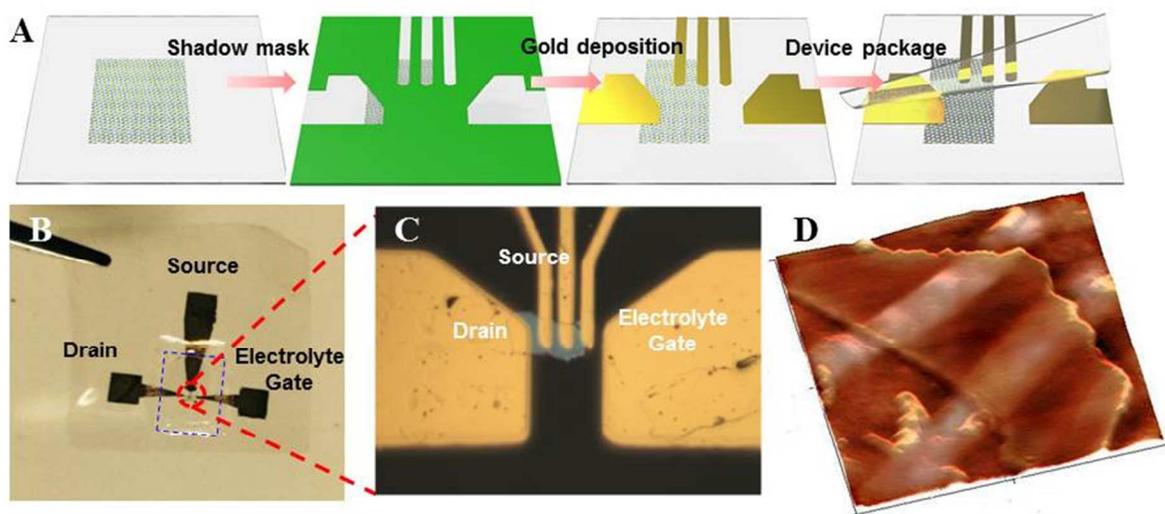
66 and ease of processing make MoS₂ an ideal candidate material for optoelectronics applications.³⁰
67 Many groups have reported on MoS₂-based phototransistors fabricated on Si/SiO₂ substrates,
68 which were neither flexible nor environmental friendly.^{31, 32} In this work, we demonstrate a
69 flexible, transparent and biodegradable phototransistor gated through gel-electrolyte. Two sheets
70 of nanopaper are used as the supporting substrate and the top passivation layer to sandwich the
71 MoS₂ channel and the gel-electrolyte in between. We used multilayer MoS₂ as they possess
72 higher density of states in conduction band and yield higher photo current compared to single
73 layer crystals according to theoretical predictions.³⁴ We have systematically characterized the
74 electrical and optoelectronic properties as well as the optical transmittance of the phototransistor.
75 The device exhibits exceptionally high photoresponsivity (~1.5 kA/W) and excellent optical
76 transmittance (~82%).



77

78 **Figure 1.** Three-dimensional schematic and cross-sectional view of the MoS₂ phototransistor on
 79 transparent and flexible nanopaper (The top passivation layer is not included in this illustration).

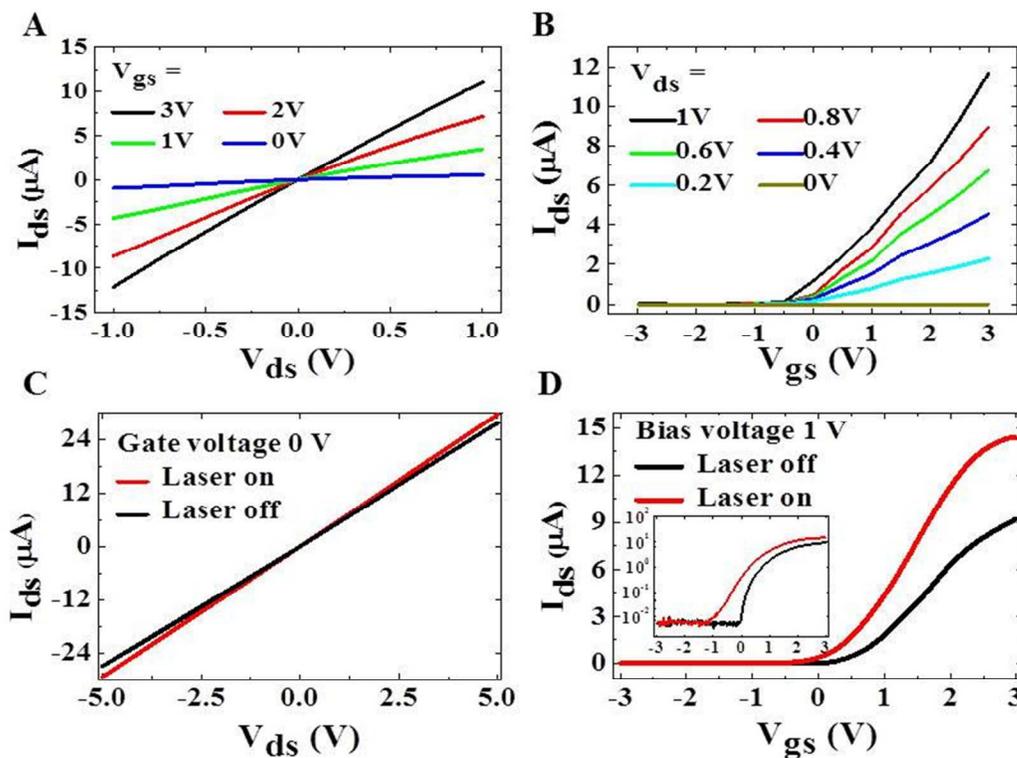
80 Figure 1 illustrates the device structure prior to passivation. 50 nm thick gold electrodes were
 81 deposited on top of a mechanically exfoliated MoS₂ flake through shadow mask. We chose
 82 shadow mask over standard photolithography to avoid dissolving the nanopaper with wet
 83 processes. The shadow mask also minimizes process-induced contaminations and ensures good
 84 contact quality between the MoS₂ and metal electrodes. We then coated a thin layer of gel-
 85 electrolyte (1 M LiClO₄ in w/w = 1:10³³ polyethylene oxide, see SI file) on top of the MoS₂ flake
 86 as the gate dielectric³⁴⁻³⁶. To immobilize the gel-electrolyte and protect the active area, we
 87 covered the surface with a separate sheet of nanopaper to form a conformal seal. The entire
 88 process flow of the sandwich structure is summarized in Fig. 2A and in SI file.



89

90 **Figure 2.** A. Fabrication process of the phototransistor. B. Photograph of a sealed
 91 phototransistor. C. Optical image of zoom-in area of the phototransistor with MoS₂. D. An
 92 atomic force microscope image of the phototransistor.

93 The photograph in Fig. 2B shows a sealed device. The blue dashed line identifies the edge of the
 94 top sealing nanopaper. The size is large enough to cover the entire MoS₂ flake and the electrodes,
 95 while small enough to expose the metal pads for probing or wire bonding. The microscope image
 96 of Fig. 2C zooms into the active area of the device. The MoS₂ flake at the center serves as the
 97 photo-sensitive channel. An atomic force microscopy image is shown in Fig. 2D. Thickness of
 98 the MoS₂ flake was measured to be 25 nm.



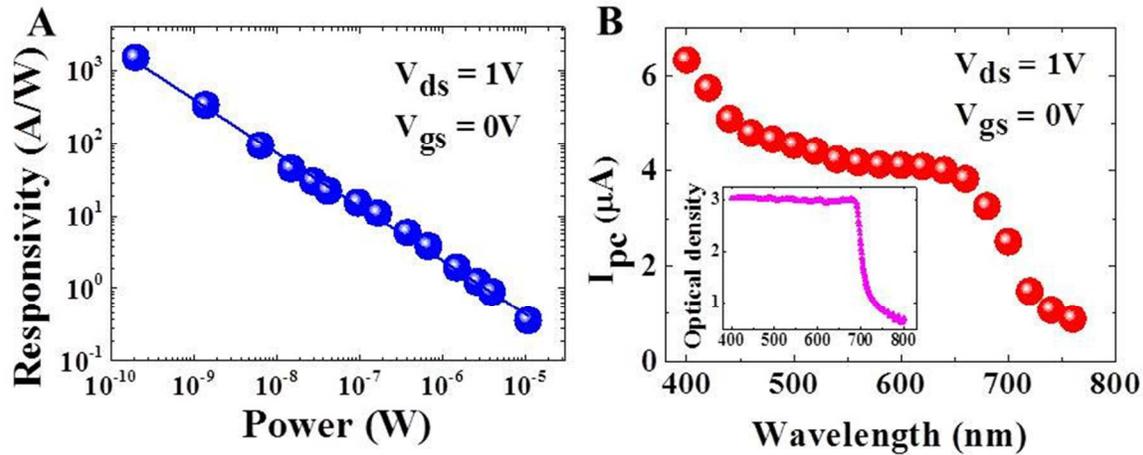
99

100 **Figure 3. Room temperature electrical and optoelectrical properties of the MoS₂**
 101 **phototransistor. A.** Output ($I_{ds} - V_{ds}$) characteristics of the phototransistor under different gate
 102 voltages. **B.** Room temperature transfer characteristics ($I_{ds} - V_{gs}$) of the same phototransistor at
 103 different source-drain biases. **C.** $I_{ds} - V_{ds}$ at $V_{gs} = 0V$ before (black) and after (red) exposure to
 104 laser illumination. The power of the incident laser is 15 μW , and the wavelength is 532 nm. **D.**

105 Transfer characteristics ($I_{ds} - V_{gs}$) before (black) and after (red) exposure to laser. The inset shows
106 the same data in log scale.

107 The electron transport properties of the phototransistor are characterized at room temperature in
108 atmosphere. Before the characterization, we took an I-V measurement on a blank nanopaper and
109 estimated the leakage current to be $< 2\text{pA}$ under 5V (SI file). Fig. 3A shows the $I_{ds} - V_{ds}$ curves
110 under four different gate biases, which are linear and symmetric, indicating good contact quality
111 between the MoS_2 channel and Au electrodes. The lithography-free fabrication eliminates the
112 need of adhesion metals and prevents contaminations associated with wet processes, resulting in
113 highly stable conductance and superior carrier mobility compared to previous work (see SI
114 file).³⁷

115 The transfer curves ($I_{ds} - V_{gs}$) of the MoS_2 phototransistor are presented in Fig. 3B, revealing a
116 typical n-type semiconducting behavior, which is consistent with previous reports.^{4, 6, 8, 9} Fig. 3C
117 shows the $I_{ds} - V_{ds}$ curves of the phototransistor when illumination is on (red) and off (black),
118 respectively. Under stable and continuous illumination (532 nm , $15\mu\text{W}$), the incident laser can
119 generate a significant photocurrent in the phototransistor. We compare the transfer characteristics
120 with (red curve) and without (black curve) the illumination in Fig. 3D. In the dark state, the
121 phototransistor shows a threshold voltage of $V_t = 0.4\text{ V}$, which shifts to $V_t = 0.1\text{ V}$ when the
122 illumination is on. .



123

124 **Figure 4. Photo response of the MoS₂ phototransistor.** A. Photoresponsivity of the device as a
 125 function of illumination power. The fitting uses $\sim E^{\beta-1}$, where E is the illumination power; β is
 126 the constant B. Photocurrent of the same MoS₂ device at different wavelengths under $V_{ds} = 1$ V
 127 and $V_{gs} = 0$ V. The inset shows the absorption curve of the MoS₂.

128 We further explored the photo detection performance of the device, characterized by its external
 129 photoresponsivity R , defined as the ratio of the photocurrent (I_{pc}) and the incident illumination
 130 power (P_{in}):

$$R = \frac{I_{pc}}{P_{in}}$$

131 As shown in Fig. 4A, the responsivity decreases with increasing illumination power (E), which is
 132 consistent with the observation in previous studies.^{Error! Bookmark not defined.} A quantitative
 133 correlation between R and E can be written³⁸ as:

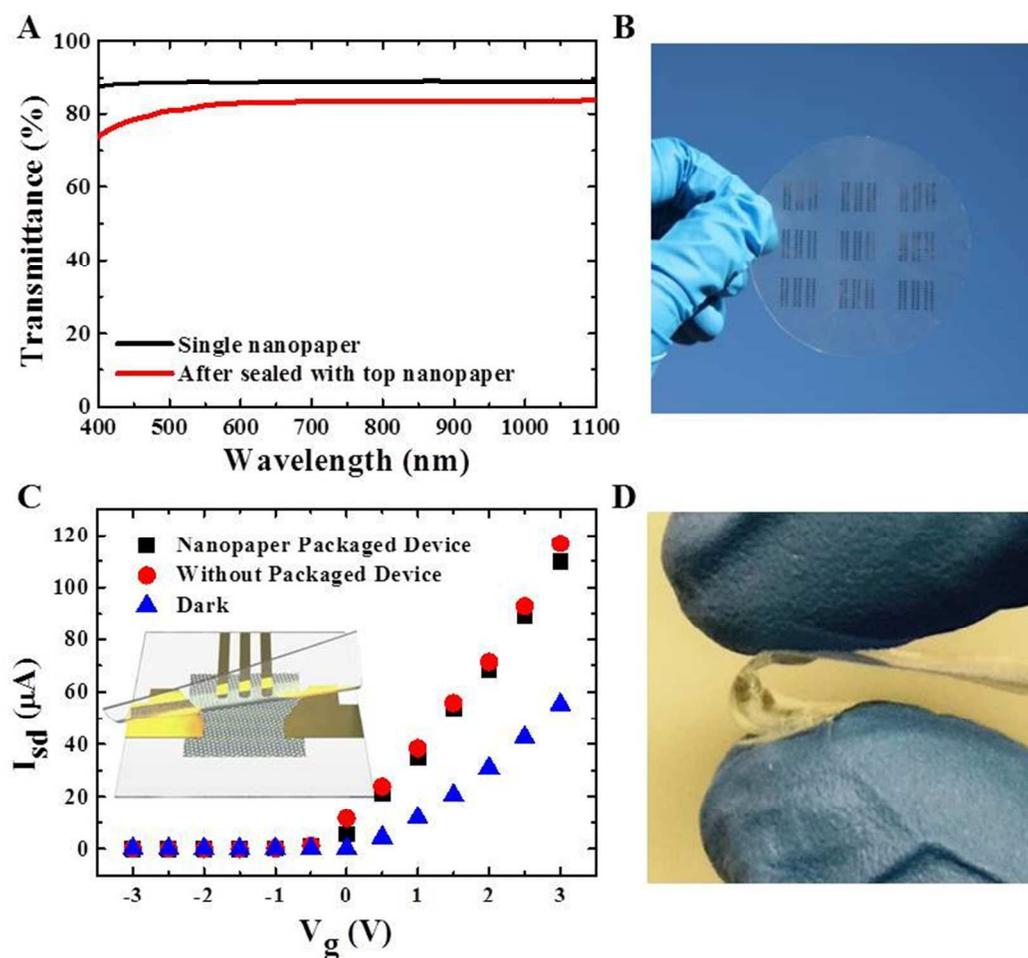
$$R \sim E^{\beta-1}$$

134 The equation has been successfully applied to other phototransistors based on single-layer MoS₂,

135 graphene and PbS.³⁷ By fitting the data (in SI file), we derived to be 0.26. The parameter reflects
136 the recombination dynamics of photo induced carriers ^[39] and ranges between 0.2 to 0.7 in
137 previous studies.^{39, 40} The decrease in responsivity with incident optical power is commonly
138 observed in phototransistors according to other reports.^{20, Error! Bookmark not defined., 41} This effect is
139 presumably associated with a reduction of the number of photo generated carriers available for
140 extraction under high photon flux due to Auger processes or the saturation of recombination and
141 trap states that influence the lifetime of the generated carriers.³⁹ In addition, we note that the
142 responsivity of our device is considerably higher than the average value reported in literatures.
143 The maximum value reached 1.5 kA/W at 10nW illumination power (see SI file for more
144 information), while the highest responsivity measured with back gated MoS₂ phototransistors are
145 typically below 1kA/W.^{26,30} We believe both the lithography-free fabrication and the electrolyte
146 gating contribute to the responsivity enhancement, and the latter plays a more dominant role. The
147 electrolyte we used is a liquidized salt that contains cations (Li⁺).³³ The cations accumulate on
148 MoS₂ surface under an externally applied voltage, forming an electric double layer (EDL). The
149 EDL is able to effectively screen charge impurities and reduce electron scattering, thereby
150 largely boosting the carrier mobility by 1 to 2 orders of magnitude.⁴² At the same time, the
151 electrolyte can also induce strong band bending at the MoS₂/Au interface and significantly lower
152 the Schottky barrier.^{43, 44} The large enhancement in both intrinsic and extrinsic mobility is
153 presumably the reason for the exceedingly high responsivity observed in our phototransistors.

154 We have also measured the dark/light current across a wide range of wavelengths (Fig. 4B).
155 When the incident laser is above 685 nm, the photocurrent drops sharply with increasing
156 wavelength. The transition around 685 nm corresponds to the excitation of electrons across the
157 bandgap (1.81eV) of multilayer MoS₂. The same transition is also evidenced in the absorption

158 curve in the inset. Below 685 nm, the slow and monotonic increase in photocurrent with
 159 decreasing wavelength (or increasing photon energy) is presumably due to the fact that higher
 160 energy photons can excite electrons to higher energy states, resulting in a larger number of
 161 photocarriers that can overcome local energy barriers in the conduction channel and/or at the
 162 MoS₂/metal interface.



163

164 **Figure 5. Optical transmittance, transparency and flexibility of the MoS₂ phototransistor.**

165 **A.** Optical transmittance of a single sheet of nanopaper (black line) and a MoS₂ phototransistor

166 sandwiched between two layers of nanopaper (red line). **B.** Photograph of an array of

167 phototransistors showing high transparency. **C.** Transfer characteristics of the phototransistor
168 without illumination (blue), under illumination but without passivation (red), and with both
169 illumination and passivation (black). **D.** The device can be largely bended, exhibiting great
170 flexibility.

171 Besides excellent photoresponsivity, the phototransistor is highly transparent and flexible at the
172 same time. In Fig. 5A, we compare the optical transmittance of the device (red curve) with a
173 sheet of bare nanopaper (black curve) across a broad spectral range from 400 nm to 1100 nm.
174 When compared at 550 nm, the optical transmittance of the phototransistor (82%) is slightly
175 lower than that of the bare nanopaper (85%) due to the addition of gel-electrolyte and passivation
176 layer. Fig. 5B and 5D demonstrate the excellent flexibility of the device and its compatibility
177 with large-scale integration. The electrical and photodetection properties remain nearly the same
178 before and after bending (in SI file). To verify that the passivation layer induces negligible
179 perturbation to the phototransistor, we have recorded the transfer curves before and after
180 passivation (Fig. 5C). In the dark state, the two devices behave the same as indicated by the data
181 points in blue triangle. Under illumination (532 nm), the bare device shows slightly higher
182 current compared at the same gate bias. The difference at $V_{gs} = 3$ V is approximately 6 μ A
183 between the two devices. The result is in good agreement with the optical transmittance response
184 in Fig. 5A, in which the sealed phototransistor has a slightly lower optical transmittance
185 therefore less sensitive to the incident laser.

186 **Summary**

187 We have demonstrated an electrolyte gated phototransistor based on multilayer MoS₂. The device
188 uses highly transparent, flexible and biodegradable nanopaper as both the supporting substrate

189 and passivation layer. It exhibits excellent photoresponsivity of approximately 1.5 kA/W under
190 10nW illumination power, considerably higher than typical back gated phototransistors reported
191 in other studies. The device functions across a broad spectral range with excellent stability under
192 sustained voltage bias and illumination. These properties make the device highly attractive for
193 various industrial applications including touch sensor panels, image sensors, solar cells, and
194 intelligent displays.

195

196 **EXPERIMENTAL SECTION**

197 **Fabrication of Transparent Nanopaper.** The nanopaper was fabricated by previously
198 established method published by Zhu. et al.⁸ The experimental procedure includes three major
199 steps: (2,2,6,6-tetramethylpiperidin-1-yl)oxyl (TEMPO) oxidation reaction, Büchner filtration,
200 and disintegration through a microfluidizer (M110 EH, Microfluidics Inc., USA). TEMPO-
201 mediated oxidation started by adding and dissolving 78 mg TEMPO into 25 mL buffer solution.
202 The buffer consists of 0.08 M Sodium carbonate (Na_2CO_3), and 0.02 M Sodium bicarbonate
203 (NaHCO_3). Combine three solutions, that is, 78 mg TEMPO in 25 mL buffer, 514 mg of sodium
204 bromide (NaBr) in 50 mL buffer and 5 g dry weight of Kraft bleached softwood pulp in 115 mL
205 buffer solution. The solution was stirred at a speed of 700 rpm with IKA RW20 digital mixer for
206 10 min followed by drop-wise addition of 35 mL sodium hypochlorite (NaClO). The pH value
207 of the solution was measured every 20 mins and kept at pH=10.5 by adding 3M sodium
208 hydroxide (NaOH) for 2 hours. Reaction with continuous stirring 700 rpm lasted overnight at
209 room temperature. Then, the TEMPO-oxidized fibers were transferred to a vacuumed flask and
210 funnel for Büchner filtration using 0.65 μm nitrocellulose ester filter (Millipore DAWP29325) to

211 wash away the reagents. Fiber was washed by 1L deionized (DI) water by stirring at 800 rpm for
212 30 min and passing through Büchner funnel into a flask twice. The resulting fiber cake was
213 dissolved in DI water to form a 1 wt% solution and passed through a microfluidizer with thin z-
214 shaped chambers. The channel dimension was 200 μm and the process pressure was 25,000 psi.
215 After passing through the microfluidizer the fibers were dispersed in water to create a 1 wt%
216 nanofibrillated cellulose (NFC) solution. The NFC solution was further diluted with DI water to
217 0.2 wt% and mixed at 500 rpm for 10 min with IKA RW20 digital mixer. After that, the
218 dispersion was degassed in a bath sonicator for 20 min, and passed through a 0.65 μm pore size
219 nitrocellulose eater filter (Millipore DAWP29325). The cake formed was compressed between a
220 PET film (on top) and papers supported by iron plate (below). Then, it was transferred to a hot
221 pressing machine in 105 $^{\circ}\text{C}$ for 4-5 days to form nanopaper with a fiber diameter of about 5 nm.

222

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