

## Supplementary Information

### Interfacial Charge Phonon Coupling in Bi<sub>2</sub>Se<sub>3</sub>/WSe<sub>2</sub> Nanohybrids for Bifunctional Near-Infrared Devices

Alka Rani<sup>ab</sup>, Priyanka Chaudhary<sup>a</sup>, Wei-Cheng Jhao<sup>a</sup>, Bal Chandra Yadav<sup>\*b</sup>, Meng-Fang Lin<sup>\*a c</sup>

<sup>a</sup>Department of Materials Engineering, Ming Chi University of Technology, New Taipei City, Taiwan

<sup>b</sup>Nanomaterials and Sensors Research Laboratory, Department of Physics, Babasaheb Bhimrao Ambedkar University, Lucknow, U.P., India

<sup>c</sup>Center for Plasma and Thin Film Technologies, Ming Chi University of Technology, New Taipei City, Taiwan

**Corresponding Author:** balchandra\_yadav@rediffmail.com, mflin@mail.mcut.edu.tw

#### S1. Experimental details

##### S1.1. Materials and Methods

Selenium dioxide (SeO<sub>2</sub>, 99.8%), bismuth nitrate pentahydrate (Bi(NO<sub>3</sub>)<sub>3</sub>·5H<sub>2</sub>O, 98%, Thermo Scientific), sodium hydroxide (NaOH), gallic acid monohydrate (99.8%, Thermo Scientific), Triton X-100 (Sigma Aldrich), bulk WSe<sub>2</sub> (99.8%, Thermo Scientific), isopropanol (IPA), ethanol, acetone, deionized (DI) water, polycarbonate filter membrane (0.4 μm pore size, 47 mm diameter, Merck Isopore), Kapton tape (polyimide), and Aluminum tape were used in this study.

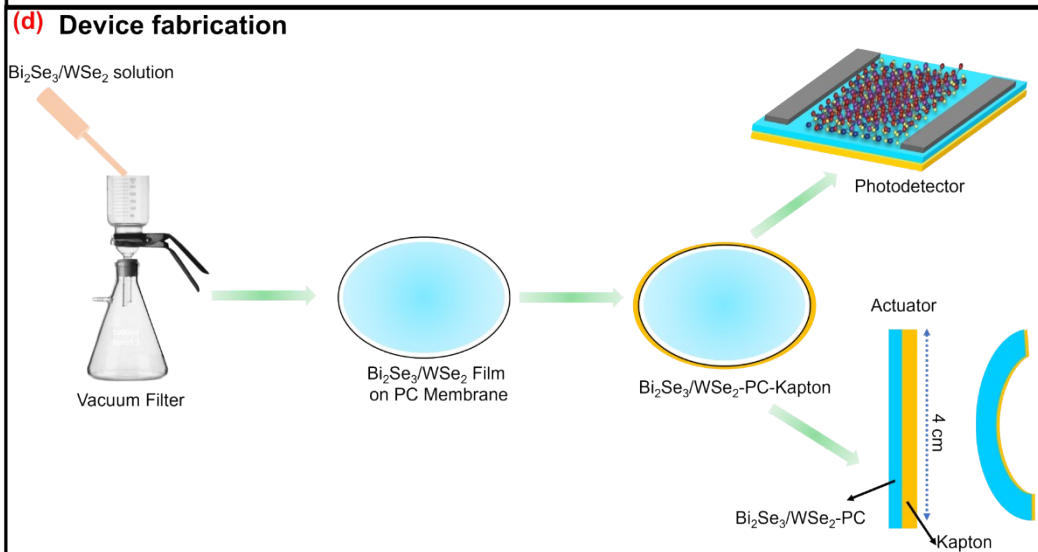
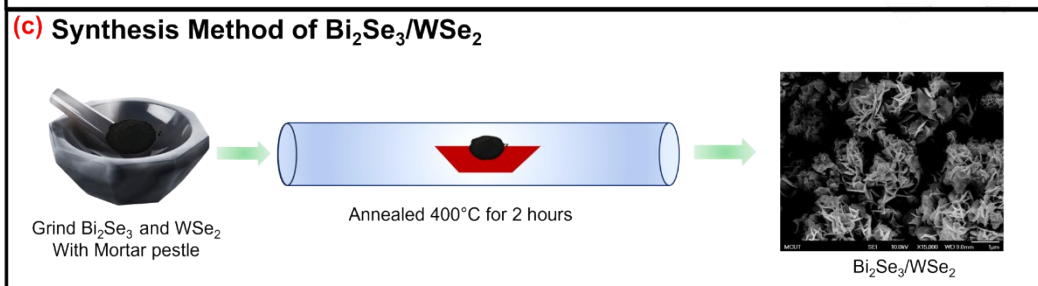
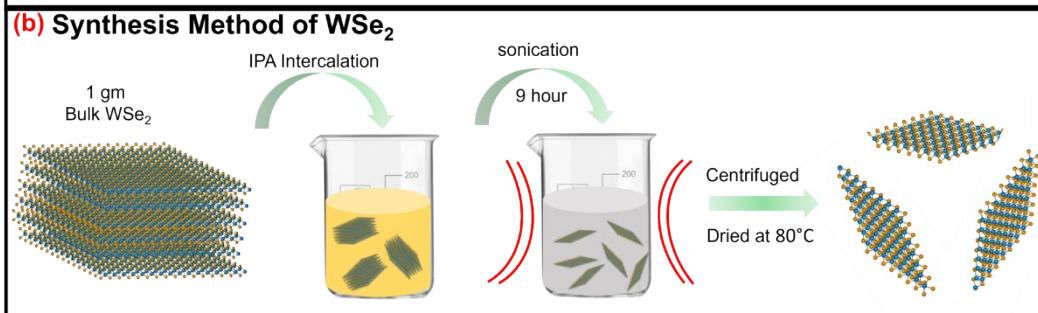
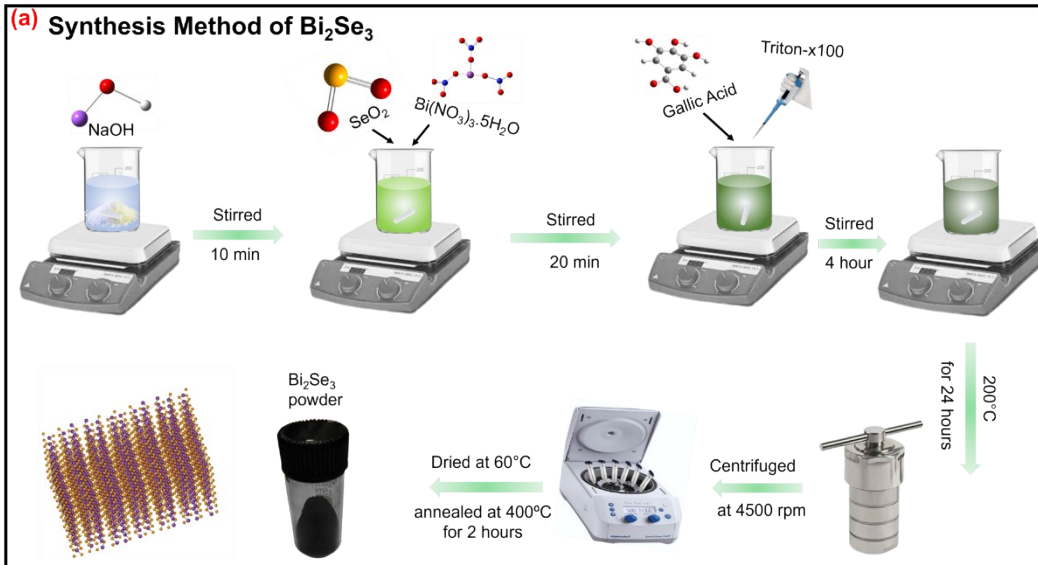
##### S1.2. Synthesis method

As shown in **Figure S1(a)**, Bi<sub>2</sub>Se<sub>3</sub> was synthesized via a hydrothermal method. First, sodium hydroxide (NaOH, 1.28 g) was dissolved in 50 mL of deionized (DI) water under magnetic stirring at 70°C for 10 minutes. Subsequently, selenium dioxide (SeO<sub>2</sub>, 0.47g) and bismuth nitrate pentahydrate (Bi(NO<sub>3</sub>)<sub>3</sub>·5H<sub>2</sub>O, 1.38 g) were added sequentially, followed by continuous stirring for 20 minutes. Then, gallic acid (1.34 g) was added to the solution, along with 50 μL of Triton X-100. The pH of the final mixture was adjusted to

28 ~12, and the reaction was maintained under stirring for 4 hours. The resulting solution  
29 was transferred into a Teflon-lined stainless-steel autoclave and heated at 200 °C for 24  
30 hours. After naturally cooling to room temperature, the obtained precipitate was collected  
31 by centrifugation, washed 5 times with DI water and ethanol, and dried at 60°C. Finally,  
32 the powder was annealed at 400 °C for 2 hours under vacuum conditions to enhance the  
33 crystallinity. As shown in **Figure S1(b)**, few-layer WSe<sub>2</sub> nanosheets were prepared via a  
34 mechanical exfoliation process. Specifically, 1 g of bulk WSe<sub>2</sub> powder was dispersed in  
35 30 ml of isopropanol and subjected to ultrasonication for 9 hours to exfoliate the layered  
36 structure, as shown in Figure S1b. The dispersion was then centrifuged to remove  
37 unexfoliated particles, and the supernatant, containing exfoliated WSe<sub>2</sub> nanosheets, was  
38 collected and dried at 80°C for 8 hours. As presented in **Figure S1(c)**, the Bi<sub>2</sub>Se<sub>3</sub>/WSe<sub>2</sub>  
39 nanohybrids were synthesized by thoroughly grinding the exfoliated WSe<sub>2</sub> nanosheets  
40 with as-prepared Bi<sub>2</sub>Se<sub>3</sub> powder using a mortar and pestle to form a homogeneous  
41 mixture. The blended material was subsequently annealed at 400°C for 2 hours under  
42 vacuum, yielding the final Bi<sub>2</sub>Se<sub>3</sub>/WSe<sub>2</sub> nanohybrid.

43

44



46 **Figure S1.** Synthesis Procedure of (a)  $\text{Bi}_2\text{Se}_3$ , (b) Exfoliated  $\text{WSe}_2$ , (c)  $\text{Bi}_2\text{Se}_3/\text{WSe}_2$   
47 nanohybrid. (d) Device fabrication of the actuator and photodetector.

48 **S1.3. Device Fabrication** The device fabrication procedure is shown in **Figure S1(d)**.  
49 Rapid near-infrared (NIR) responsive actuators were developed by leveraging the  
50 mismatch in thermal expansion. First, a polycarbonate  $\text{Bi}_2\text{Se}_3/\text{WSe}_2$ -PC composite was  
51 prepared as the upper layer of the actuator. Due to the high coefficient of thermal  
52 expansion of the PC membrane ( $\sim 98$  ppm/K), the photothermal conversion properties of  
53 modified  $\text{WSe}_2$  nanosheets were utilized to generate effective thermal actuation. The  
54 resulting PC- $\text{Bi}_2\text{Se}_3/\text{WSe}_2$  composite exhibited rapid photothermal conversion and  
55 efficient heat transfer pathways. The interconnected nanosheet network of  $\text{Bi}_2\text{Se}_3/\text{WSe}_2$   
56 facilitated rapid heat transfer to cooler surrounding regions upon NIR irradiation. Kapton,  
57 characterized by a low coefficient of thermal expansion of nearly 20 ppm/k<sup>1</sup>, was  
58 employed as the bottom layer. By combining the  $\text{Bi}_2\text{Se}_3/\text{WSe}_2$ -PC composite with Kapton,  
59 a flexible actuator was constructed. Upon NIR light irradiation, the actuator bent away  
60 from the light source due to differential expansion and reverted to its original shape when  
61 the light was turned off, demonstrating excellent, fast, and reversible actuation  
62 performance. This responsive behavior is largely attributed to the strong NIR absorption  
63 and photothermal conversion capability of the  $\text{Bi}_2\text{Se}_3/\text{WSe}_2$  nanohybrids. Following the  
64 compressive chemical and physical processes, a bifunctional device was further  
65 fabricated to serve as both a photodetector and actuator, capable of detecting varying  
66 NIR power levels. Nanohybrid powders of  $\text{Bi}_2\text{Se}_3/\text{WSe}_2$  at various mass ratios were  
67 prepared (1:1, 1:2, 1:3, 2:1, 3:1). The various compositions were achieved by fixing the  
68 mass of one component ( $\text{Bi}_2\text{Se}_3$  or  $\text{WSe}_2$ ) at 0.25 g and adjusting the mass of the other  
69 components to obtain the target ratio. Each mixture was dissolved in 30 mL of DI water.  
70 To form a stable suspension, it was first subjected to ultrasonication for 2 hours, followed  
71 by continuous magnetic stirring overnight. For the fabrication of each composite  
72 membrane, a uniform volume of 15 mL of the final nanohybrid solution was isolated and  
73 vacuum-filtered using a PC filter membrane with a pore size of 0.22  $\mu\text{m}$  and a thickness  
74 of 150  $\mu\text{m}$  for 3 hours. The resulting film was dried at 80°C for 4 hours. To complete the  
75 actuator structure, commercial Kapton tape was firmly laminated onto the  $\text{Bi}_2\text{Se}_3/\text{WSe}_2$ -

76 PC film, creating a flexible actuator. The dried composite film was then cut into actuator  
77 strips (length: 4 cm; varying widths) for actuation testing. For performance evaluation,  
78 each actuator strip was fixed at one end and vertically positioned in front of the NIR light  
79 source. Aluminum Tape was applied on the surface of the  $\text{Bi}_2\text{Se}_3/\text{WSe}_2$  layer to serve as  
80 electrodes for photodetection measurements. Bending angle of this soft actuator was  
81 quantified by measuring the angular change from the initial position to the deformed state  
82 using a protractor, with motion captured via a video camera.

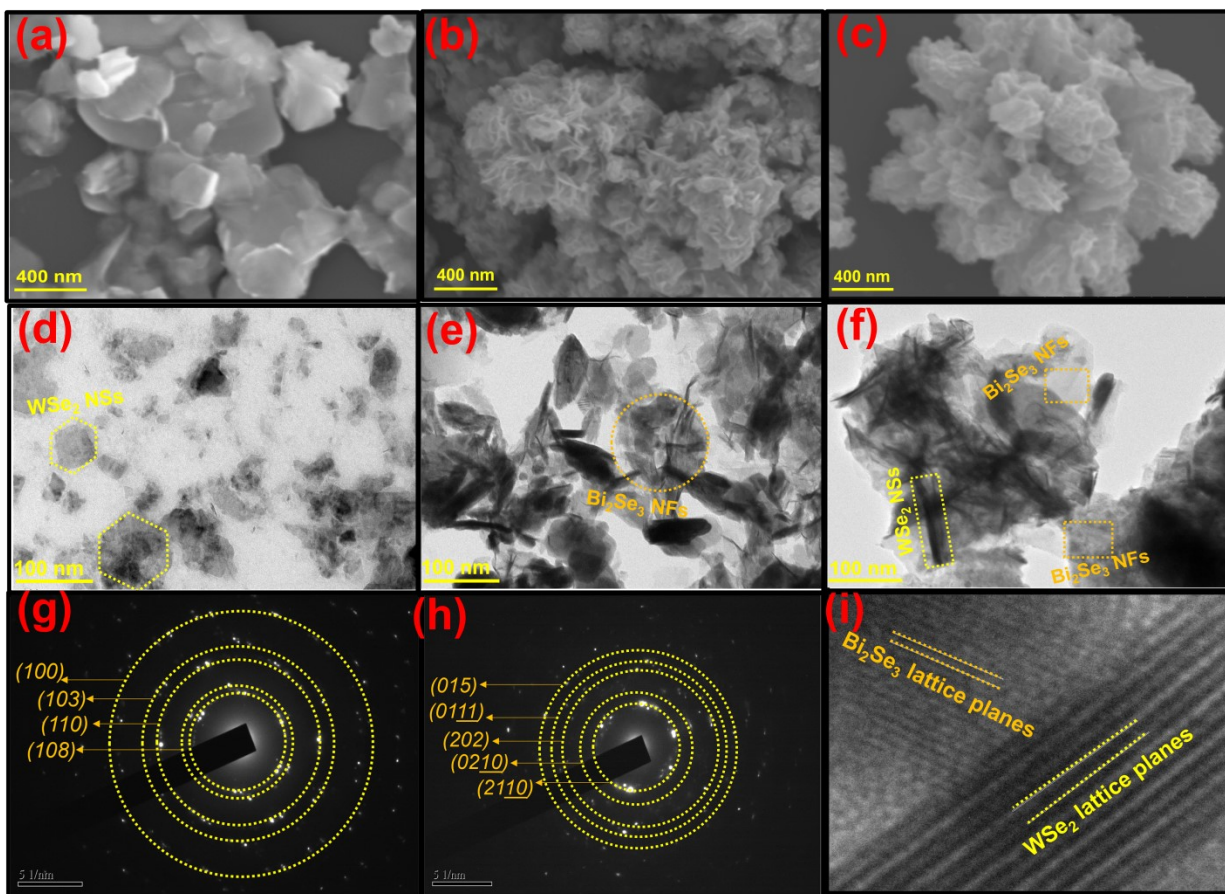
## 83 **S2. Characterization Techniques**

84 Surface morphology was examined using a scanning electron microscope (SEM; Hitachi,  
85 Model S-3400N) operated at an accelerating voltage of 15 kV. Raman spectroscopy  
86 (Horiba, Model IHR 550) with a 532 nm excitation wavelength was employed to examine  
87 the vibrational modes of the materials. Fourier-transform infrared spectroscopy (FT-IR;  
88 PerkinElmer, Model Spectrum) was used to identify the functional groups present on the  
89 surface of the  $\text{Bi}_2\text{Se}_3/\text{WSe}_2$  nanohybrids. For external stimulation, an NIR light source  
90 (Philips, Model PH070004) and a DC power supply (Hila Model DP-30052) were used.  
91 The power density of the NIR light was measured using a Linsang NIR power meter  
92 (Model LS122 IR). Actuator motion was recorded using a video camera, with angle  
93 tracked by a protractor and a laser displacement sensor (Keyence Model LK-H080).  
94 Temperature variations during actuation were monitored using a thermal imaging camera  
95 (Perfect Prime, Model IR0019). Photodetection performance was measured from the  
96 Keithley electrometer (Model 6517B).

## 97 **S3 Results and Discussion**

98 **Figure S2(a)** displays a SEM micrograph of the  $\text{WSe}_2$  nanostructure, revealing its  
99 characteristic 2D, sheet-like morphology. The nanosheets exhibit a flat, thin, and few-  
100 layered structure with smoothly folded edges. Numerous overlapping sheets are evident  
101 on the surface, contributing to a high specific surface area that enhances the transport of  
102 electrons. As shown in **Figure S2(b)**, the SEM image of  $\text{Bi}_2\text{Se}_3$  exhibits a flower-like  
103 aggregation of ultrathin nanoflakes arranged in a petal-like configuration. These  
104 hierarchical structures form well-defined nanoflowers, indicating successful  
105 morphological control during the synthesis process. As depicted in **Figure S2(c)**, the

106  $\text{Bi}_2\text{Se}_3/\text{WSe}_2$  nanohybrid successfully retains its original flower-like morphology of  $\text{Bi}_2\text{Se}_3$   
107 while integrating  $\text{WSe}_2$  nanosheets. The attachment of  $\text{Bi}_2\text{Se}_3$  nanoflowers to the surface  
108 of  $\text{WSe}_2$  forms a cauliflower-like hybrid nanostructure, suggesting strong interfacial  
109 interaction between the two components. **Figure S2(d)-(f)** presents TEM images of  
110  $\text{WSe}_2$ ,  $\text{Bi}_2\text{Se}_3$ , and the  $\text{Bi}_2\text{Se}_3/\text{WSe}_2$  nanohybrid, respectively, all at a scale of 100 nm.  
111 The TEM image of  $\text{WSe}_2$  in **Figure S2(d)** and **(e)** confirms a thin, planar hexagonal sheet-  
112 like morphology, highlighted by the yellow dotted line. The measured lattice fringe spacing  
113 of 0.287 nm corresponds to the (002) planes of  $\text{WSe}_2$ , consistent with the XRD results.  
114 The SAED pattern shown in **Figure S2(g)** exhibits well-defined diffraction spots,  
115 confirming the single-crystalline nature of  $\text{WSe}_2$ . The measured interplanar spacing of  
116 0.306 nm matches the (101) plane of rhombohedral  $\text{Bi}_2\text{Se}_3$ . The corresponding SAED  
117 pattern (Figure S2h) displays multiple concentric rings, indicating the polycrystalline  
118 nature of the sample. These rings can be indexed to the (015), (0111), (202), (0210), and  
119 (2110) planes of the  $\text{Bi}_2\text{Se}_3$ . **Figure S2(f)** shows the TEM image of the  $\text{Bi}_2\text{Se}_3/\text{WSe}_2$   
120 nanohybrid. The observed lattice fringes reveal interplanar spacing of 0.498 nm and 0.311  
121 nm, corresponding to  $\text{WSe}_2$  and  $\text{Bi}_2\text{Se}_3$ , respectively. These results further confirm the  
122 successful formation of the nanohybrid and the preservation of the crystallinity of both  
123 constituents.

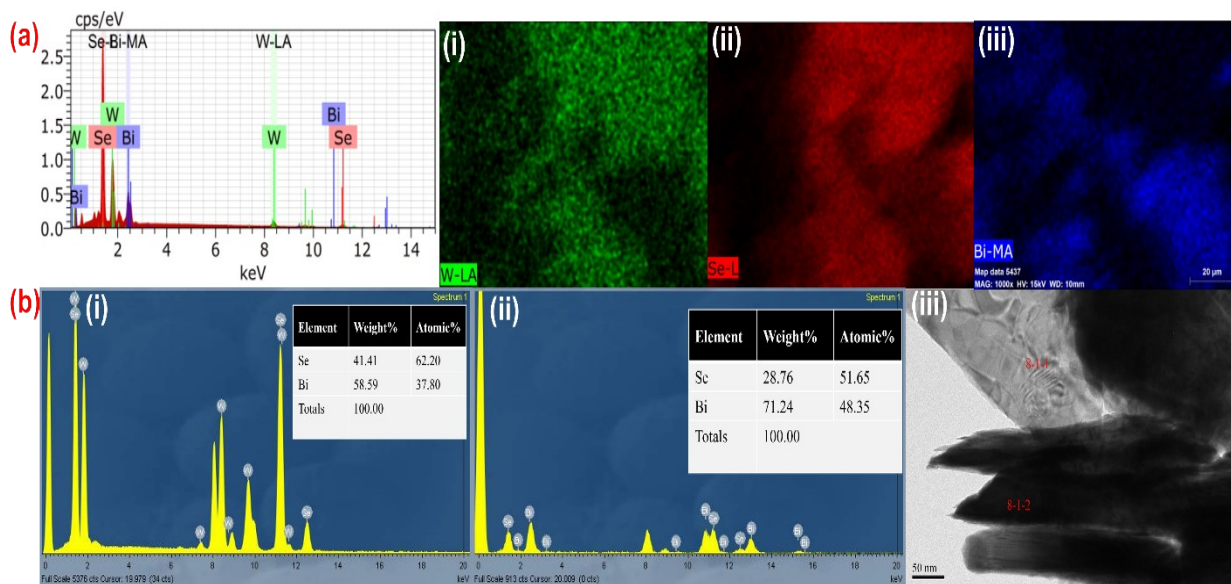


124

125 **Figure S2.** SEM images at a 400 nm scale of materials, (a) WSe<sub>2</sub>, (b) Bi<sub>2</sub>Se<sub>3</sub>, (c)  
 126 Bi<sub>2</sub>Se<sub>3</sub>/WSe<sub>2</sub>. TEM images at a 100 nm scale of materials, (d) WSe<sub>2</sub>, (e) Bi<sub>2</sub>Se<sub>3</sub>, (f)  
 127 Bi<sub>2</sub>Se<sub>3</sub>/WSe<sub>2</sub>. SAED pattern of (g) WSe<sub>2</sub>, (h) Bi<sub>2</sub>Se<sub>3</sub>. (i) Representation of Bi<sub>2</sub>Se<sub>3</sub> and  
 128 WSe<sub>2</sub> lattice planes in Bi<sub>2</sub>Se<sub>3</sub>/WSe<sub>2</sub> nanohybrid.

129 To further elucidate the chemical composition within the domains of WSe<sub>2</sub>, and Bi<sub>2</sub>Se<sub>3</sub>,  
 130 EDS elemental mapping was performed on the Bi<sub>2</sub>Se<sub>3</sub>/WSe<sub>2</sub> shown in **Figure S3a**. The  
 131 mapping results reveal a nearly uniform distribution of W, Se, and Bi across the domains.  
 132 Selenium distribution is red in color which is uniform and intense across the entire region.  
 133 This indicates Se rich composition. The EDS spectrum obtained from the mapped region  
 134 shows characteristic peaks of Bi, W, and Se. This confirms the presence of WSe<sub>2</sub> and  
 135 Bi<sub>2</sub>Se<sub>3</sub> within the mapped domains, which approves the formation of Bi<sub>2</sub>Se<sub>3</sub>/WSe<sub>2</sub>  
 136 nanohybrid. Furthermore, the EDS elemental spectra obtained by TEM presented in the  
 137 supplementary **Figure S3b**, show good compatibility with the composition of Bi<sub>2</sub>Se<sub>3</sub>/WSe<sub>2</sub>

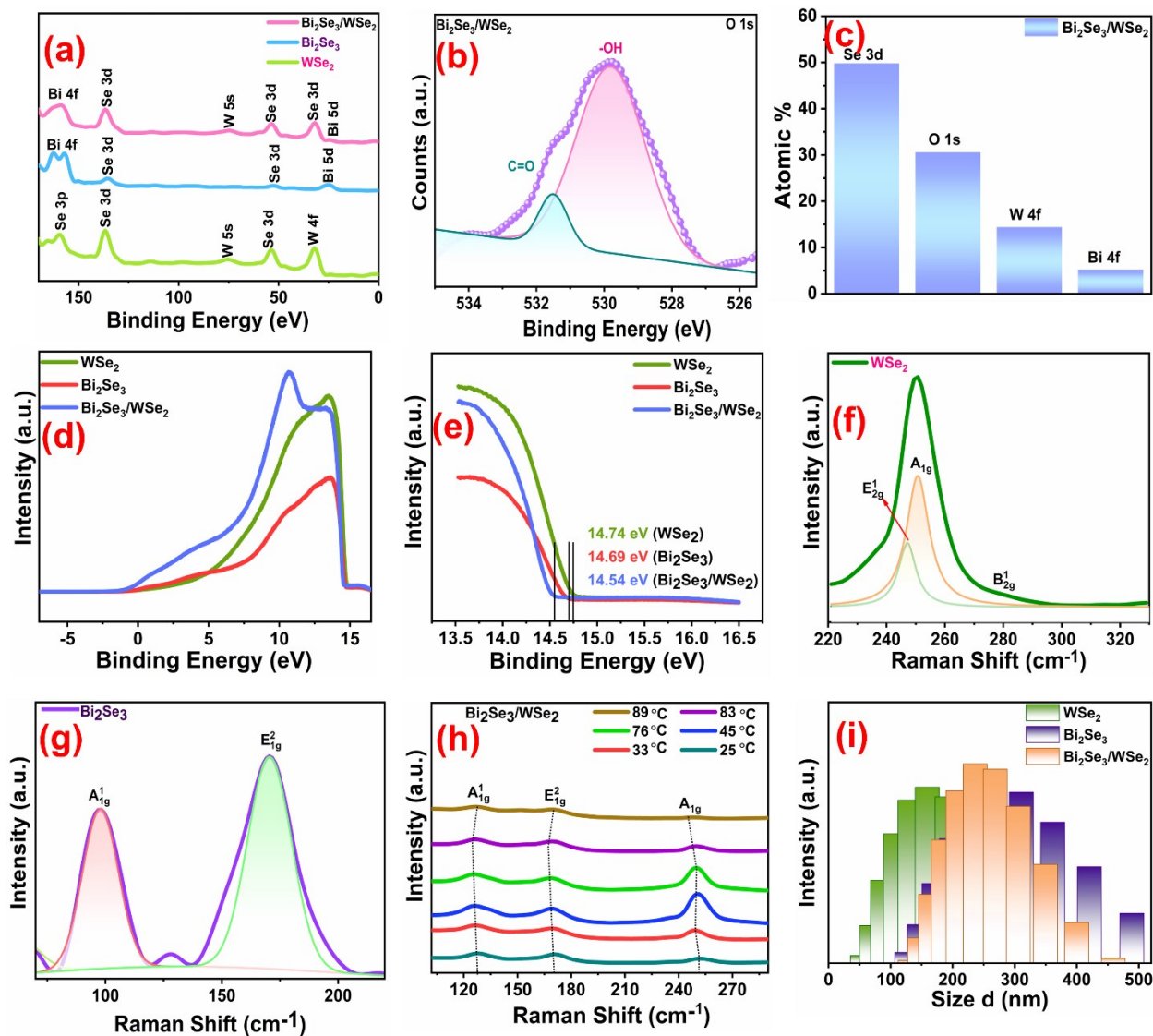
138 nanohybrid. **Figure S3b(i)** shows the EDS spectrum of  $\text{Bi}_2\text{Se}_3$  while **Figure S3b(ii)**  
 139 represents the EDS spectrum of  $\text{WSe}_2$  present in  $\text{Bi}_2\text{Se}_3/\text{WSe}_2$  nanohybrid. These  
 140 regions are identified in **Figure S3b(iii)** where the marked areas correspond to the  
 141 locations from which the spectra were collected. The region marked 8-1-1 corresponds  
 142 to the  $\text{Bi}_2\text{Se}_3$  flakes, while the region indicated as 8-1-2 represents the  $\text{WSe}_2$   
 143 nanostructures, confirming the presence of both in the  $\text{Bi}_2\text{Se}_3/\text{WSe}_2$  nanohybrid.



144

145 **Figure: S3** (a) Elemental mapping showing homogeneous distribution of Bi, W and Se  
 146 located on as prepared  $\text{Bi}_2\text{Se}_3/\text{WSe}_2$  nanohybrid. (b) EDS Spectra of  $\text{Bi}_2\text{Se}_3/\text{WSe}_2$   
 147 nanohybrid.

148



149

150 **Figure S4.** (a) XPS survey spectrum of WSe<sub>2</sub>, Bi<sub>2</sub>Se<sub>3</sub>, and Bi<sub>2</sub>Se<sub>3</sub>/WSe<sub>2</sub>, confirming the  
 151 presence of W, B, and Se. (c) Surface elemental percentage of Bi, W, Se, and O  
 152 calculated by XPS. (d) Full range UPS spectrum of all samples WSe<sub>2</sub>, Bi<sub>2</sub>Se<sub>3</sub>, and  
 153 Bi<sub>2</sub>Se<sub>3</sub>/WSe<sub>2</sub>. (e) UPS binding energy profiles showing the secondary electron cut off  
 154 energy. Raman spectrum of (f) WSe<sub>2</sub>, (g) Bi<sub>2</sub>Se<sub>3</sub>. (h) Raman spectra of Bi<sub>2</sub>Se<sub>3</sub>/WSe<sub>2</sub>  
 155 nanohybrid at different temperatures. (i) DLS histogram of WSe<sub>2</sub>, WSe<sub>2</sub> and Bi<sub>2</sub>Se<sub>3</sub>/WSe<sub>2</sub>  
 156 nanohybrid.

157 In pristine WSe<sub>2</sub>, the W 4f spectrum shows two peaks at 30.75 eV (W4f<sub>7/2</sub>) and 32.91 eV  
 158 (W 4f<sub>5/2</sub>), confirming the W<sup>4+</sup> oxidation state associated with W-Se bonding. A small peak

159 at 36.48 eV suggests minor surface oxidation ( $WO_x$  species). The Se 3d spectrum shows  
 160 two prominent peaks at 52.98 eV and 53.72 eV, corresponding to the Se  $3d_{5/2}$  and Se  
 161  $3d_{3/2}$  states, confirming selenium's presence in  $WSe_2$ . Further, the Se element state 3d  
 162 spectrum of pristine  $Bi_2Se_3$  reveals two distinct peaks: Se-I at 52.28 eV, attributed to Bi-  
 163 Se bonds, and Se-II at 53.94 eV, corresponding to Se-Se bonds, likely present due to  
 164 slight non-stoichiometry or surface states. The Bi 4f spectrum shows peaks at 156.32 eV  
 165 and 161.61 eV assigned to  $Bi^{3+}$  (Bi  $4f_{7/2}$  and Bi  $4f_{5/2}$ ). Additional peaks observed at 157.54  
 166 eV and 162.78 eV are indicative of surface oxidation of  $Bi_2Se_3$ . The spin-orbit splitting of  
 167 5.29 eV between Bi  $4f_{7/2}$  and Bi  $4f_{5/2}$  aligns well with reported values for  $Bi_2Se_3$ , confirming  
 168 its oxidation state and purity. The complete survey, shown in **Figure S4(a)**, displays the  
 169 pure formation of synthesized materials, with the presence of atoms and their  
 170 corresponding binding energies. As shown in **Figure S4(b)**, the O 1s spectra of  
 171  $Bi_2Se_3/WSe_2$  exhibit peaks at 532.30 eV, 532.29 eV, and 531.50 eV, respectively, which  
 172 are typically attributed to adsorbed oxygen or water molecules—a common observation in  
 173 samples exposed to ambient air. The atomic weight percentage of surface elements  
 174 present in the  $Bi_2Se_3/WSe_2$  nanohybrid is presented in **Figure S4(c)** with the highest  
 175 atomic % Se in the 3d state.

176 In UPS (**Figure S4d and e**), the measurement of work function using the width of the  
 177 photoemission spectrum, that is, from the secondary electron cut off to the Fermi edge,  
 178 as presented in the Equations S(1) and (2).<sup>2</sup>

$$179 \quad \phi = h\nu - K.E_{max} \quad (S1)$$

$$180 \quad \phi = h\nu - (E_{SEC} - E_F) \quad (S2)$$

181 Where  $\phi$ ,  $h$ ,  $\nu$ ,  $E_{SEC}$ , and  $E_F$  are the work function, Planck's constant, frequency of  
 182 Ultraviolet light, onset energy or cut-off energy of secondary electrons, and Fermi level,  
 183 respectively.

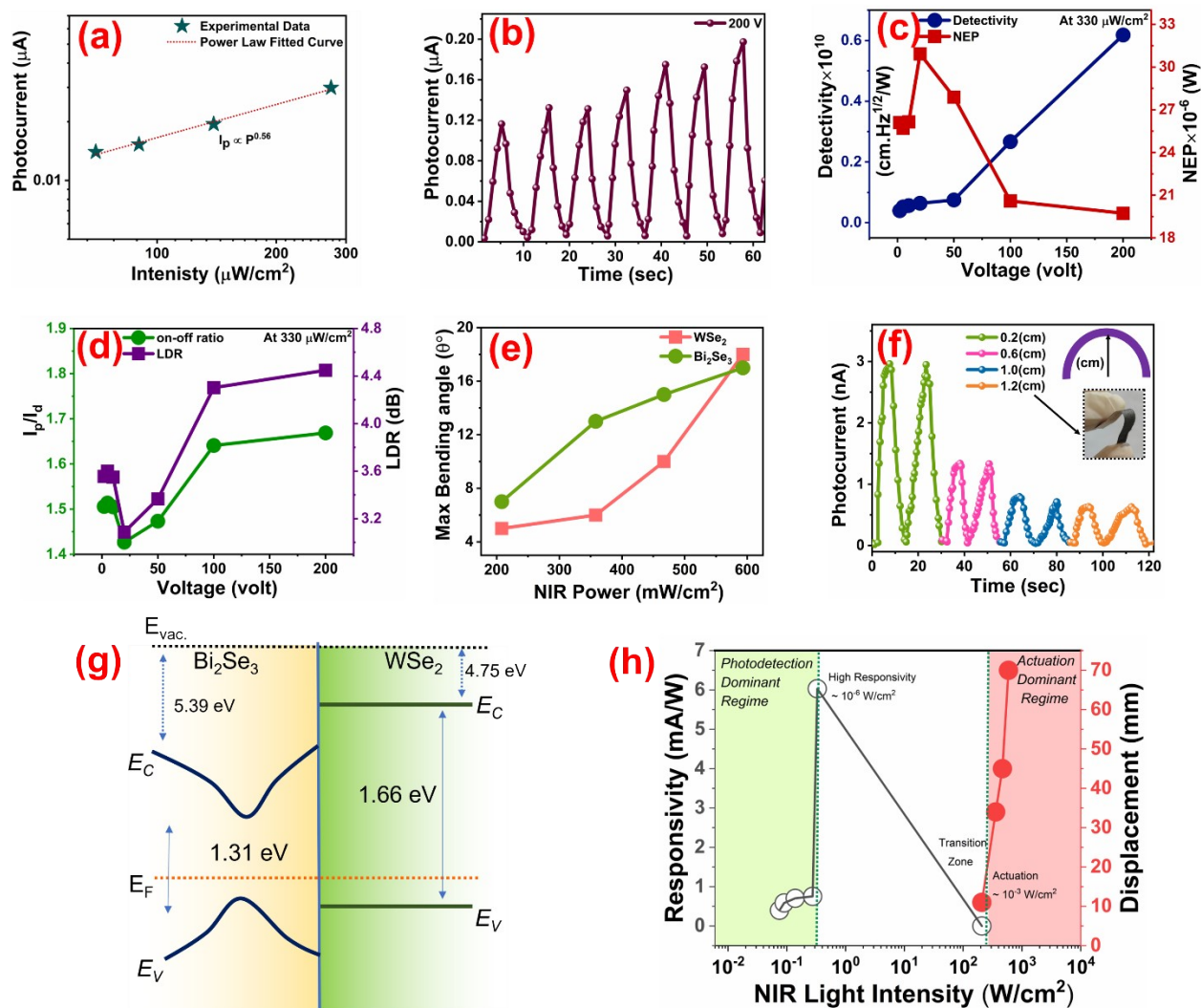
184 By the UV-visible spectroscopy, the absorption coefficient can be determined by  
 185 Equations S (3) and (4)

$$186 \quad \alpha = 2.303 \frac{A}{t} \quad (S3)$$

187 Where  $t$  is the thickness. The theory of optical absorption defines the relationship between  
188 the absorption coefficient and the photon energy for direct allowed transitions, as in  
189 Equation S4

$$190 \quad \alpha h\nu = A(h\nu - E_g)^{\frac{1}{2}} \quad (S4)$$

191 Where  $A$  is the constant,  $E_g$  shows the band gap, and  $h\nu$  represents the photon energy.  
192 The band gap  $E_g$  can be determined by extrapolating the linear region of the  $(\alpha h\nu)^2$  versus  
193  $h\nu$  plot is extrapolated at  $\alpha=0$ . **Figure S4f and g**, represent the Raman spectrum of pure  
194  $WSe_2$  and pure  $Bi_2Se_3$ . Raman spectra of  $Bi_2Se_3/WSe_2$  nanohybrid at different  
195 temperatures is presented in **Figure 4h**. **Figure S4i**, shows the particle size obtained  
196 using the DLS technique, all the values ranging between for  $WSe_2$ ,  $Bi_2Se_3$ , and  
197  $Bi_2Se_3/WSe_2$  nanohybrid is 92-488 nm, 125-568 nm, and 92-420 nm respectively.



198

199 **Figure S5.** (a) Power-dependent photocurrent of the device at 200 V bias under NIR  
 200 illumination. (b) Transient photo response of PD at 200V under 330 μW/cm<sup>2</sup>. (c,d)  
 201 Detectivity, NEP, on/off ratio, and LDR under 330 μW/cm<sup>2</sup> at different bias potentials for  
 202 Bi<sub>2</sub>Se<sub>3</sub>/WSe<sub>2</sub> nano-hybrid. (e) Flexibility test of PD under 330 μW/cm<sup>2</sup>. (f) Actuation  
 203 performance of Bi<sub>2</sub>Se<sub>3</sub> and WSe<sub>2</sub> actuators of 3 mm width in response to different light  
 204 stimulation intensities. (g) Energy band diagram of Bi<sub>2</sub>Se<sub>3</sub>/WSe<sub>2</sub> nano-heterojunctions  
 205 showing the VB and CB. (h) An intensity-dependent phase diagram clearly indicates the  
 206 threshold between sensing-dominant and actuation-dominant regimes.

207 The relationship between the photocurrent and light intensity, as in a power law function,  
 208 is shown in **Figure S5(a)**. **Figure S5(b)** depicts the transient photo response of PD at

209 200V under 330  $\mu\text{W}/\text{cm}^2$ . Responsivity, defined as the photocurrent generated per unit  
210 incident optical power on the active device area, is expressed as equation **S5**:<sup>3</sup>

$$211 \quad R = \frac{I_{\text{light}} - I_{\text{dark}}}{P \cdot A} \quad (\text{S5})$$

212 Where  $I_{\text{light}}$  and  $I_{\text{dark}}$  denote the current under illumination and in the dark, respectively.  $P$   
213 is the incident light intensity, and  $A$  is the photo-absorbing active area of the device.

214 The specific detectivity ( $D$ ), a critical parameter reflecting the ability to detect weak  
215 signals, was extracted using equation **S6**:<sup>4</sup>

$$216 \quad D = \frac{1}{(2qI_d)^{1/2}} R \cdot A^2 \quad (\text{S6})$$

217 where  $q$  is the elementary charge and  $I_d$  is the dark current. This power-dependent trend  
218 is also evident in responsivity, detectivity, and linear dynamic range (LDR). The LDR, a  
219 critical figure of merit for image sensors, is expressed as equation **S7**:<sup>5</sup>

$$220 \quad \text{LDR} = 20 \log \left( \frac{J_{\text{upper}}}{J_{\text{lower}}} \right) \quad (\text{S7})$$

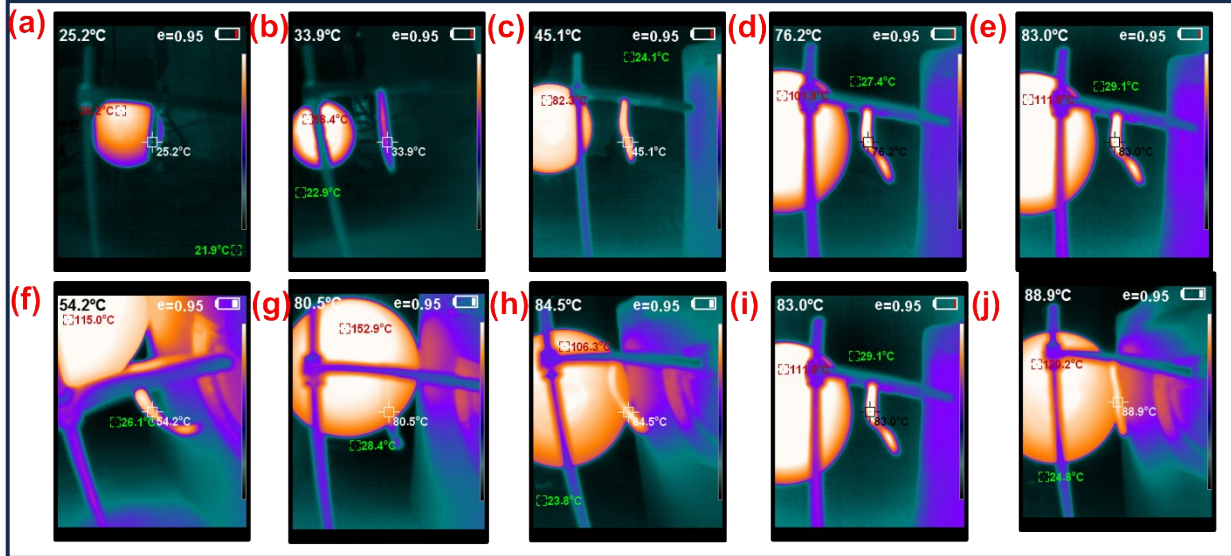
221 where  $J$  represents the current density. The upper limit corresponds to the deviation from  
222 linear photocurrent response, typically governed by slower carrier transit times.<sup>6</sup> **Figure**  
223 **S5c** and **d** represent the detectivity, NEP, on-off ratio, and LDR of the PD device at  
224 different bias potentials under NIR illumination of intensity 330  $\mu\text{W}/\text{cm}^2$   $\text{Bi}_2\text{Se}_3/\text{WSe}_2$   
225 nanohybrid. As represented in **Figure S5e**, The I-t curves were recorded under different  
226 bending conditions at an applied bias of 200 V and an illumination intensity of 330  $\mu\text{W}/\text{cm}^2$   
227 to evaluate the flexibility of the device. **Figure S5e** demonstrates the time-dependent  
228 response of the flexible device's photocurrent under periodic illumination at various  
229 bending radii. The flexible device shows distinct ON/OFF switching characteristics under  
230 various bending states. It is also interesting to see that the photocurrent increases when  
231 the bending radius decreases. The highest response is observed when the radius is 0.2  
232 cm. It is possible that the strain effect enhances the charge of transport and separation  
233 at the interface. A band diagram of the  $\text{Bi}_2\text{Se}_3$  and  $\text{WSe}_2$  nano-heterojunctions formed  
234 within the  $\text{Bi}_2\text{Se}_3/\text{WSe}_2$  nanohybrid is shown in **Figure S5(g)**. As shown in **Figure S5h**,  
235 we have plotted a phase diagram featuring three distinct regions: the photodetection-

236 dominant regime, the transition zone, and the actuation-dominant regime. In the first  
 237 region, the photodetection dominant regime, the responsivity increases as the light  
 238 intensity level rises from  $75 \mu\text{W}/\text{cm}^2$  to  $330 \mu\text{W}/\text{cm}^2$  at low intensity ( $\mu\text{W}$  order). As the  
 239 intensity increases into the high-intensity region (mW order), at  $208 \text{ mW}/\text{cm}^2$ , the  
 240 photoresponsivity begins to decrease. At such high intensities, thermally generated  
 241 electron-hole pairs become highly active, which increases the dark current and  
 242 concurrently increases the device's temperature. This temperature rise is utilized for  
 243 photothermal actuation, effectively transforming the device into an actuator.  
 244 Consequently, in the actuation dominant regime, spanning  $208 \text{ mW}/\text{cm}^2$  to  $593 \text{ mW}/\text{cm}^2$ .  
 245 We analyzed the performance as an actuator, achieving a maximum bending angle of  
 246  $70^\circ$ . Above this intensity, the device suffers from material defects and ultimately burns  
 247 out.

248 **Table S1:** Photodetection performance of parameter of  $\text{Bi}_2\text{Se}_3/\text{WSe}_2$

| Ip<br>(A)             | Id<br>(A)              | On-<br>off<br>ratio | Responsi<br>vity<br>(A/W)               | Detectivity<br>( $\text{cm}\cdot\text{Hz}^{1/2}/\text{W}$<br>) | NEP<br>(W)            | LDR<br>(dB) | Voltage<br>(Volt) |
|-----------------------|------------------------|---------------------|---|--|-----------------------|-------------|-------------------|
| $1.99 \times 10^{-7}$ | $1.19 \times 10^{-7}$  | 1.67                | <b><math>6.03 \times 10^{-3}</math></b> | $6.18 \times 10^9$   | $1.97 \times 10^{-5}$ | 4.45        | 200               |
| $3.97 \times 10^{-8}$ | $2.42 \times 10^{-8}$  | 1.64                | $1.17 \times 10^{-3}$                   | $2.67 \times 10^9$   | $2.06 \times 10^{-5}$ | 4.30        | 100               |
| $5.14 \times 10^{-9}$ | $3.49 \times 10^{-9}$  | 1.47                | $1.25 \times 10^{-4}$                   | $7.49 \times 10^8$   | $2.79 \times 10^{-5}$ | 3.3         | 50                |
| $4.49 \times 10^{-9}$ | $3.15 \times 10^{-9}$  | 1.43                | $1.02 \times 10^{-4}$                   | $6.41 \times 10^8$   | $3.09 \times 10^{-5}$ | 3.09        | 20                |
| $2.63 \times 10^{-9}$ | $1.75 \times 10^{-9}$  | 1.50                | $6.68 \times 10^{-5}$                   | $5.65 \times 10^8$   | $2.61 \times 10^{-5}$ | 3.55        | 10                |
| $2.17 \times 10^{-9}$ | $1.43 \times 10^{-9}$  | 1.51                | $5.58 \times 10^{-5}$                   | $5.21 \times 10^8$   | $2.57 \times 10^{-5}$ | 3.60        | 5                 |
| $1.30 \times 10^{-9}$ | $8.60 \times 10^{-10}$ | 1.51                | $3.30 \times 10^{-5}$                   | $3.97 \times 10^8$   | $2.61 \times 10^{-5}$ | 3.56        | 2                 |

249

250 **S4. Photothermal performance**

251

252 **Figure S6.** Temperature infrared images of the actuator (1:3) of 3 mm when exposed to  
 253 different intensities of infrared light. (a) Dark, (b) 208, (c) 358, (d) 467, and (e) 593  
 254 mW/cm<sup>2</sup>. Temperature infrared images of Bi<sub>2</sub>Se<sub>3</sub>/WSe<sub>2</sub> actuator (3 mm) when exposed to  
 255 593 mWcm<sup>-2</sup> intensity near infrared light. (f) 1:1, (g) 1:2, (h) 1:3, (i) 2:1, and (j) 3:1.

256 The photothermal conversion efficiencies of the Bi<sub>2</sub>Se<sub>3</sub>, WSe<sub>2</sub>, and the Bi<sub>2</sub>Se<sub>3</sub>/WSe<sub>2</sub> films  
 257 were calculated using the following photothermal conversion efficiency equation S8

$$258 \quad \eta = \frac{Cm\Delta T}{Pst} \times 100 \% \quad (\text{S8})$$

259 where  $\eta$  represents the photothermal conversion efficiency. C is the specific heat  
 260 capacity. It quantifies how much heat must be delivered to a substance to heat it over a  
 261 temperature range<sup>10</sup>. we have incorporated literature values of C for the individual  
 262 components; 147.3 J/mol K<sup>11</sup> for WSe<sub>2</sub>, 160 J/mol K<sup>12</sup> for Bi<sub>2</sub>Se<sub>3</sub>, 153 J/mol K  
 263 (Bi<sub>2</sub>Se<sub>3</sub>/WSe<sub>2</sub>), for Bi<sub>2</sub>Se<sub>3</sub>/WSe<sub>2</sub> we estimate an average value. m is the mass of the film,  
 264 and is the change in the film temperature during irradiation, P corresponds to the power  
 265 density of the incident NIR light, S is the effective illuminated area of the film surface, and

266 t is the exposure time under illumination. calculated photothermal conversion efficiencies  
 267 for Bi<sub>2</sub>Se<sub>3</sub>, WSe<sub>2</sub>, and the Bi<sub>2</sub>Se<sub>3</sub>/WSe<sub>2</sub> are 12 %, ~ 15%, 29.6% respectively. The  
 268 incident power density was maintained at 593 mW/cm<sup>2</sup> during irradiation. Under these  
 269 conditions the temperature of the WSe<sub>2</sub>, Bi<sub>2</sub>Se<sub>3</sub>, and Bi<sub>2</sub>Se<sub>3</sub>/WSe<sub>2</sub> nanohybrid films  
 270 increased within 7 sec, 8 sec and within 7 sec. The maximum temperature for WSe<sub>2</sub> and  
 271 Bi<sub>2</sub>Se<sub>3</sub> was 60°C and 64° C.

272 **Table S2:** Performance parameters of TMDCs and Bi<sub>2</sub>Se<sub>3</sub> as a photodetector and light-  
 273 driven actuator

| Material   | Photocurrent (nA)    | Detectivity (cm·Hz <sup>1/2</sup> /W) | Max Displacement/ angle | Max Temperature (°C) | Intensity (mW/cm <sup>2</sup> ) | References |
|--|----------------------|---------------------------------------|-------------------------|----------------------|---------------------------------|------------|
| WS <sub>2</sub> -PNiPAM                            | -                    | -                                     | 7 mm                    | 70                   | 8000 (NIR)                      | 7          |
| Bi <sub>2</sub> Se <sub>3</sub>                    | ~110                 | 3.3×10 <sup>10</sup>                  | -                       | -                    | 142.93 (NIR)                    | 8          |
| WSe <sub>2</sub> single crystal                    | ~938×10 <sup>3</sup> | 10 <sup>7</sup>                       | -                       | -                    | 3 (Visible)                     | 9          |
| GO   | -                    | -                                     | 1.6 cm <sup>-1</sup>    | 102                  | 600 (NIR)                       | 10         |
| MoS <sub>2</sub> -PI                               | -                    | -                                     | ~70°                    | 148                  | 148 (Sunlight)                  | 11         |
| PDMS/WS <sub>2</sub>                               | -                    | -                                     | 23.99°                  | -                    | 300 (NIR)                       | 12         |
| WSe <sub>2</sub> -graphene/PDMS                    | -                    | -                                     | ~82°                    | 142                  | 970 (NIR)                       | 13         |
| WSe <sub>2</sub>                                   | ~ 160                | 3.03×10 <sup>11</sup>                 | -                       | -                    | 105 (Visible)                   | 14         |
| WSe <sub>2</sub> , Au contact                      | ~80                  | 2.26×10 <sup>11</sup>                 | -                       | -                    | 8 (Visible)                     | 15         |
| Bi <sub>2</sub> Se <sub>3</sub> /ReSe <sub>2</sub> | 265×10 <sup>-6</sup> | 1.1×10 <sup>11</sup>                  | -                       | -                    | 1,600 (Visible)                 | 16         |

|   |            |                            |            |             |                    |                 |
|---|------------|----------------------------|------------|-------------|--------------------|-----------------|
| <b>Bi<sub>2</sub>Se<sub>3</sub>/WSe</b> | <b>199</b> | <b>6.18×10<sup>9</sup></b> | <b>70°</b> | <b>88.9</b> | <b>0.330 (NIR)</b> | <b>Our work</b> |
|---|------------|----------------------------|------------|-------------|--------------------|-----------------|

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