

**Supplementary Information for**

**Layered-material WS<sub>2</sub>/topological insulator Bi<sub>2</sub>Te<sub>3</sub> heterostructure  
photodetector with ultrahigh responsivity in the range from 370 to 1550 nm**

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**S1. The (002) diffraction peaks of WS<sub>2</sub> grown on Bi<sub>2</sub>Te<sub>3</sub> with different thickness and their full width at half maximum (FWHM).**

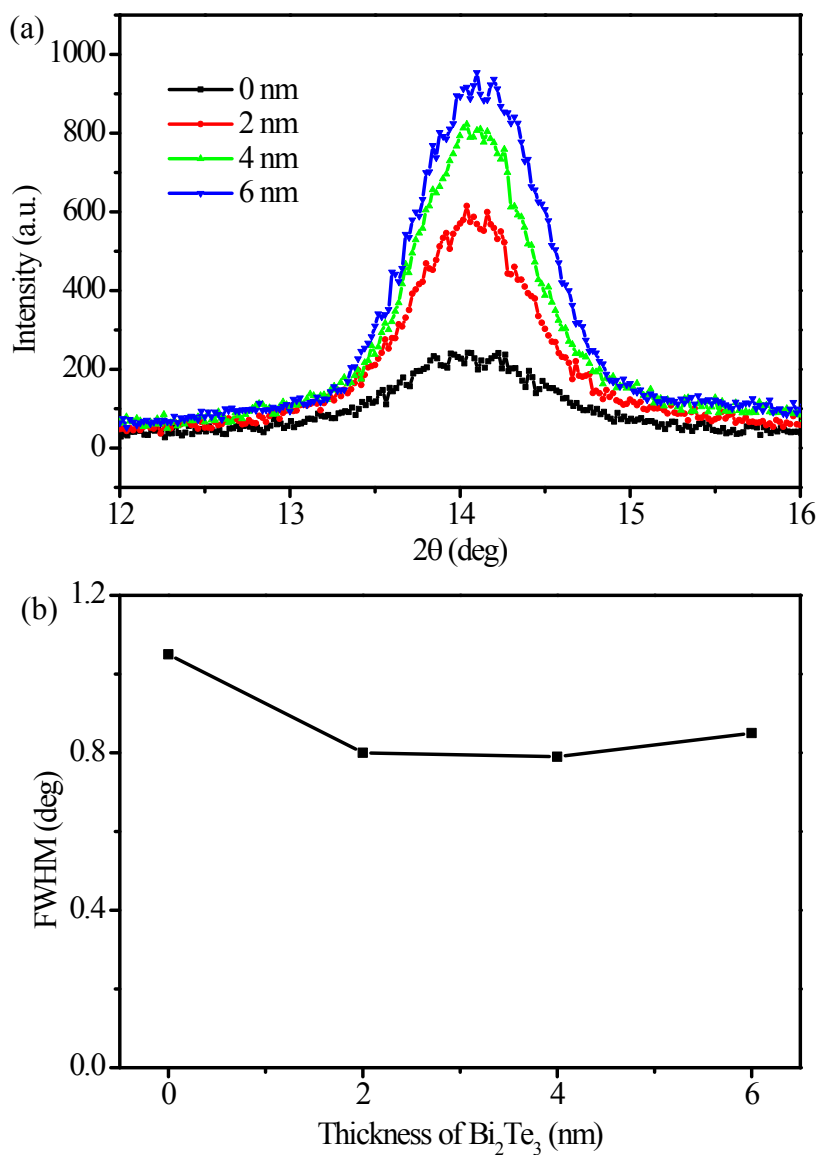


Figure S1. (a) The (002) diffraction peaks of WS<sub>2</sub> grown on Bi<sub>2</sub>Te<sub>3</sub> with different thickness and their (b) FWHM. Thickness of WS<sub>2</sub>: 15 nm.

## S2. AFM thickness profiles of PLD grown $\text{WS}_2$ and $\text{Bi}_2\text{Te}_3$ .

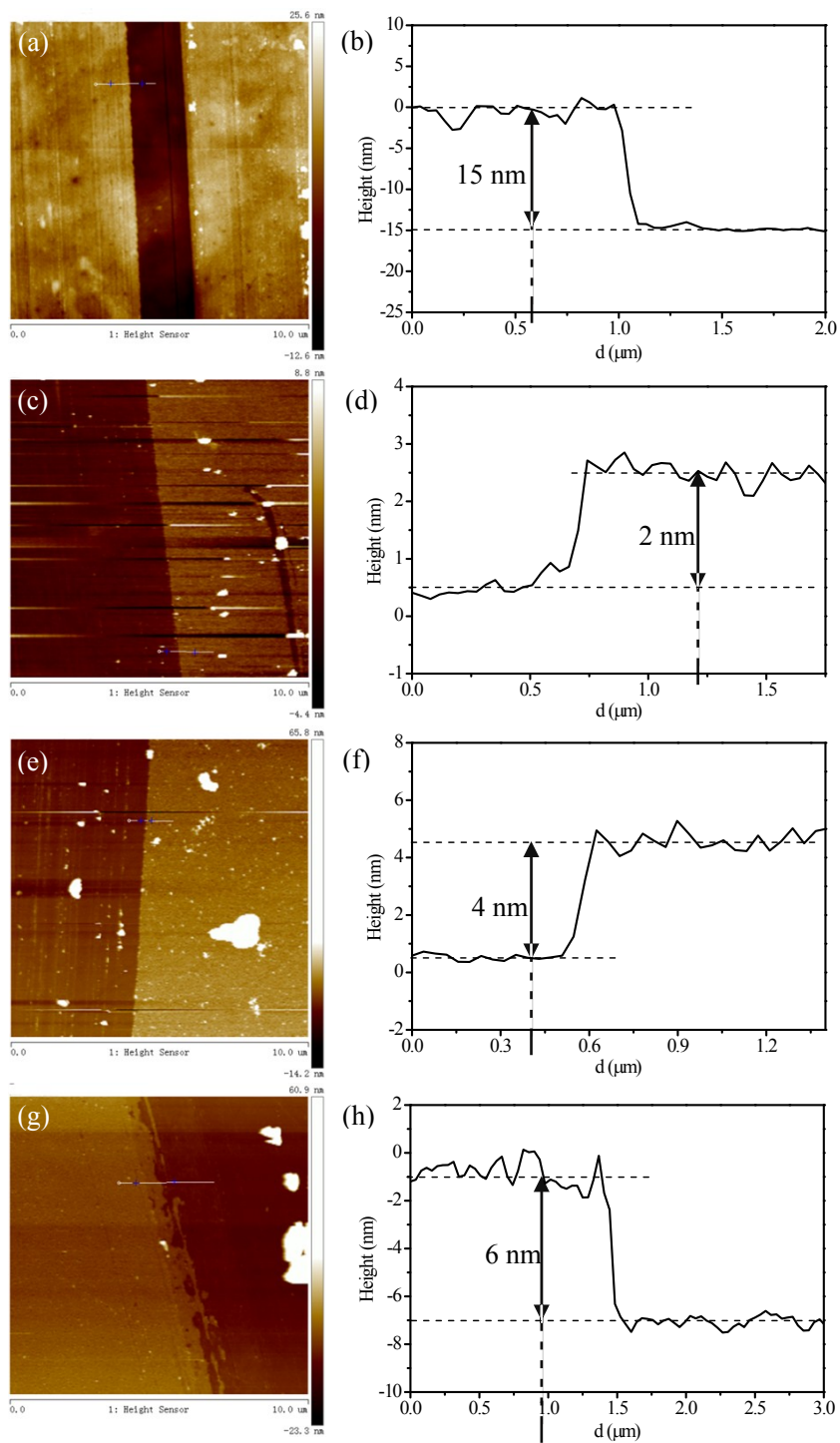


Figure S2. AFM images of PLD-grown (a)  $\text{WS}_2$  specimen of 10000 pulse and  $\text{Bi}_2\text{Te}_3$  specimen of (a) 26, (b) 52 and (c) 78 pulse.

### S3. Long-term stability of the WS<sub>2</sub>/Bi<sub>2</sub>Te<sub>3</sub> photodetectors

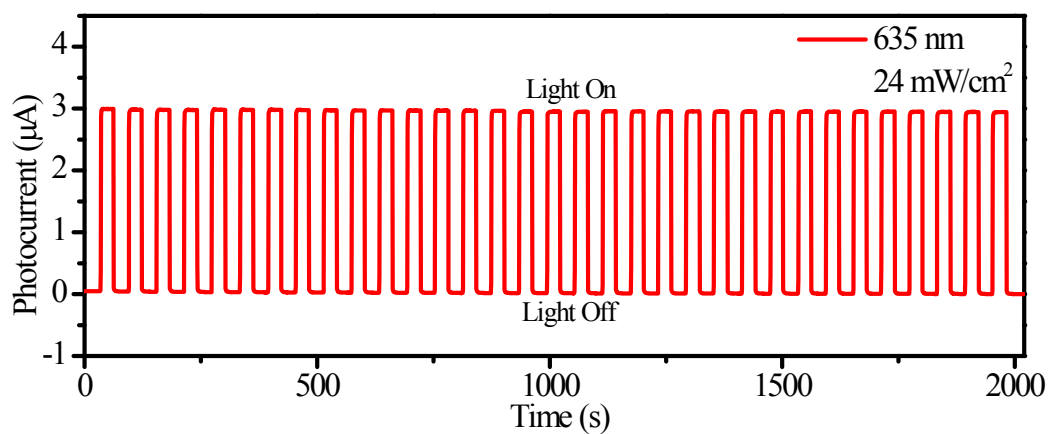


Figure S3. Photoswitching curve of the WS<sub>2</sub>/Bi<sub>2</sub>Te<sub>3</sub> photodetector in response to long-term periodic 635-nm illumination (~ 2000 s, 33 cycles).

#### S4. Absorption spectrum of the pure WS<sub>2</sub> film and WS<sub>2</sub>/Bi<sub>2</sub>Te<sub>3</sub> heterojunction film

Fig. S4 presents the absorption spectrum of the pure WS<sub>2</sub> film (black) and WS<sub>2</sub>/Bi<sub>2</sub>Te<sub>3</sub> heterojunction film (red). To exclude the absorption from the substrates, transparent mica was exploited as substrates. Note that the periodic oscillation of absorption spectrum comes from the interference effect of the layered mica substrates, which, however, doesn't hinder us from drawing the conclusion. Obviously, absorption edge appears at around 1000 nm for the pure multilayer WS<sub>2</sub> film. After the addition of a Bi<sub>2</sub>Te<sub>3</sub> layer, no absorption edge can be observed in the whole measured range extending from 400 to 2000 nm, which is benefit from the small bandgap of Bi<sub>2</sub>Te<sub>3</sub> (0.15 eV). Therefore, the above results provide convincing evidence that the photoresponse to the 1550-nm illumination of the WS<sub>2</sub>/Bi<sub>2</sub>Te<sub>3</sub> heterojunction photodetector originates from the Bi<sub>2</sub>Te<sub>3</sub> layer.

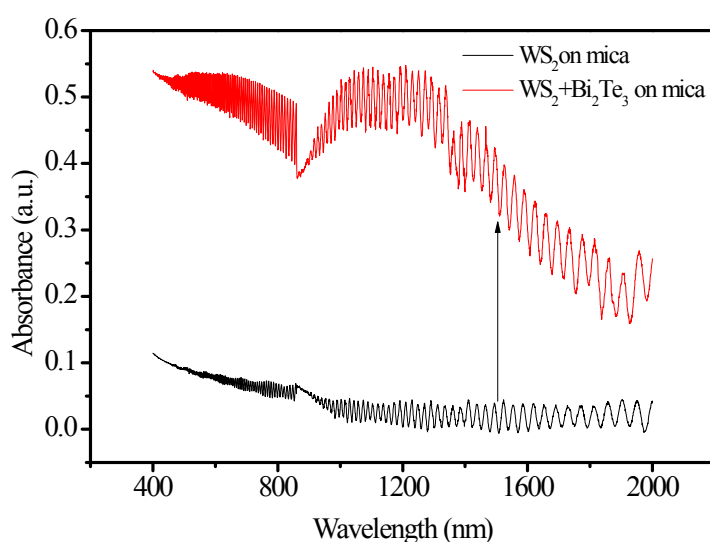


Figure S4. Absorption spectrum of the pure WS<sub>2</sub> film and WS<sub>2</sub>/Bi<sub>2</sub>Te<sub>3</sub> heterojunction film.

### S5. Dark current of the device with different thickness of $\text{Bi}_2\text{Te}_3$ .

The dark current of the devices with different thickness of  $\text{Bi}_2\text{Te}_3$  is summarized in Fig. S5. In general, the dark current increases as the thickness of  $\text{Bi}_2\text{Te}_3$  increases. Note that the dark current of  $\text{WS}_2/\text{Bi}_2\text{Te}_3$  (2 nm) is slightly larger than that of  $\text{WS}_2/\text{Bi}_2\text{Te}_3$  (4 nm), which seems to be counterintuitive. However, it is actually reasonable. When the thickness of  $\text{Bi}_2\text{Te}_3$  is thinner than ca. 4 nm, the dark current of  $\text{Bi}_2\text{Te}_3$  is relative smaller on account of its discontinuous nature. Thus, the  $\text{WS}_2$  channel dominates the dark current of the  $\text{WS}_2/\text{Bi}_2\text{Te}_3$  photodetector. Since the quality of  $\text{WS}_2$  increases as the thickness of  $\text{Bi}_2\text{Te}_3$  increases, its defect doping thus decreases, resulting in the decrease of the dark current of the  $\text{WS}_2$  channel. Thus, the dark current of  $\text{WS}_2/\text{Bi}_2\text{Te}_3$  (2 nm) is slightly larger than that of  $\text{WS}_2/\text{Bi}_2\text{Te}_3$  (4 nm). As the thickness of  $\text{Bi}_2\text{Te}_3$  increases to 6 nm, it becomes totally continuous and its dark current is much larger than that of  $\text{WS}_2$ . As a result, the dark current  $\text{WS}_2/\text{Bi}_2\text{Te}_3$  (6 nm) is much larger. Other parameters such as the variation of the contact barrier between the electrode and the channel may also affect, which demands further investigation.

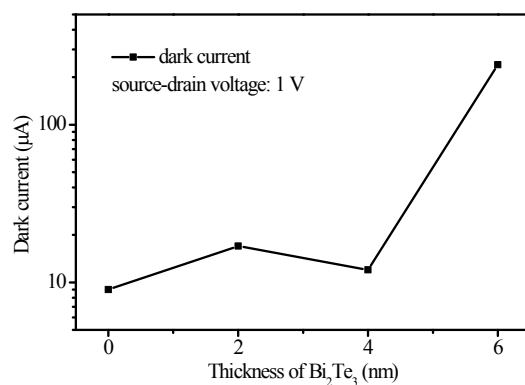


Figure S5. Dark current of the  $\text{WS}_2/\text{Bi}_2\text{Te}_3$  photodetectors.

## S6. Application of the interface engineering methodology to PLD-grown MoS<sub>2</sub>

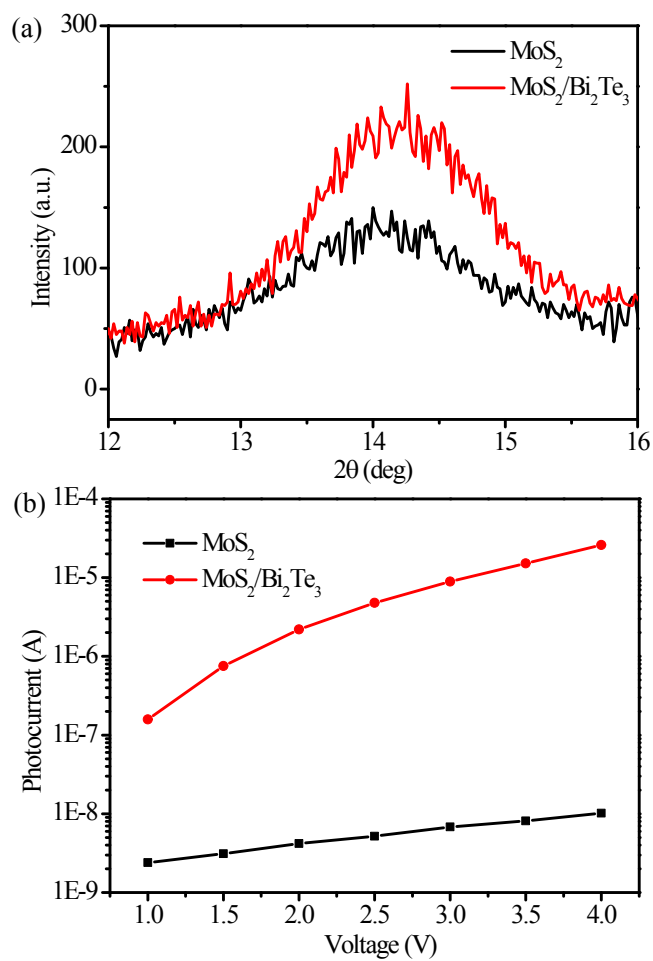


Figure S6. (a) (002) diffraction peaks of the MoS<sub>2</sub> grown on SiO<sub>2</sub> and Bi<sub>2</sub>Te<sub>3</sub>. (b)

Voltage dependent photocurrent of the MoS<sub>2</sub> and MoS<sub>2</sub>/Bi<sub>2</sub>Te<sub>3</sub> photodetectors.