Supplementary Information

High-Temperature Flexible WSe2 Photodetectors with Ultrahigh Photoresponsivity

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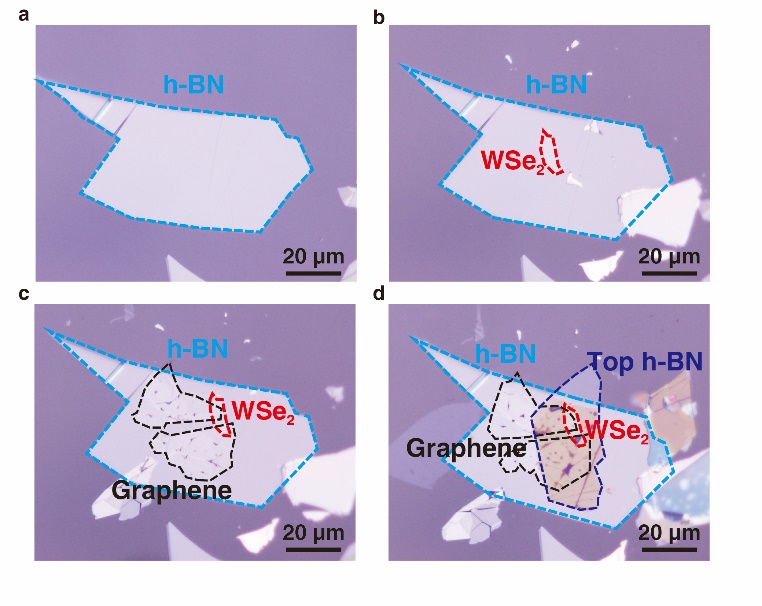
Figure S13. Photo responsivity of WSe2 device with mica encapsulation under 385 nm (purple), 440 nm (blue), and 520 nm (green) illumination at different temperatures (*Vds*=0.4 V, *Vg*=0 V).

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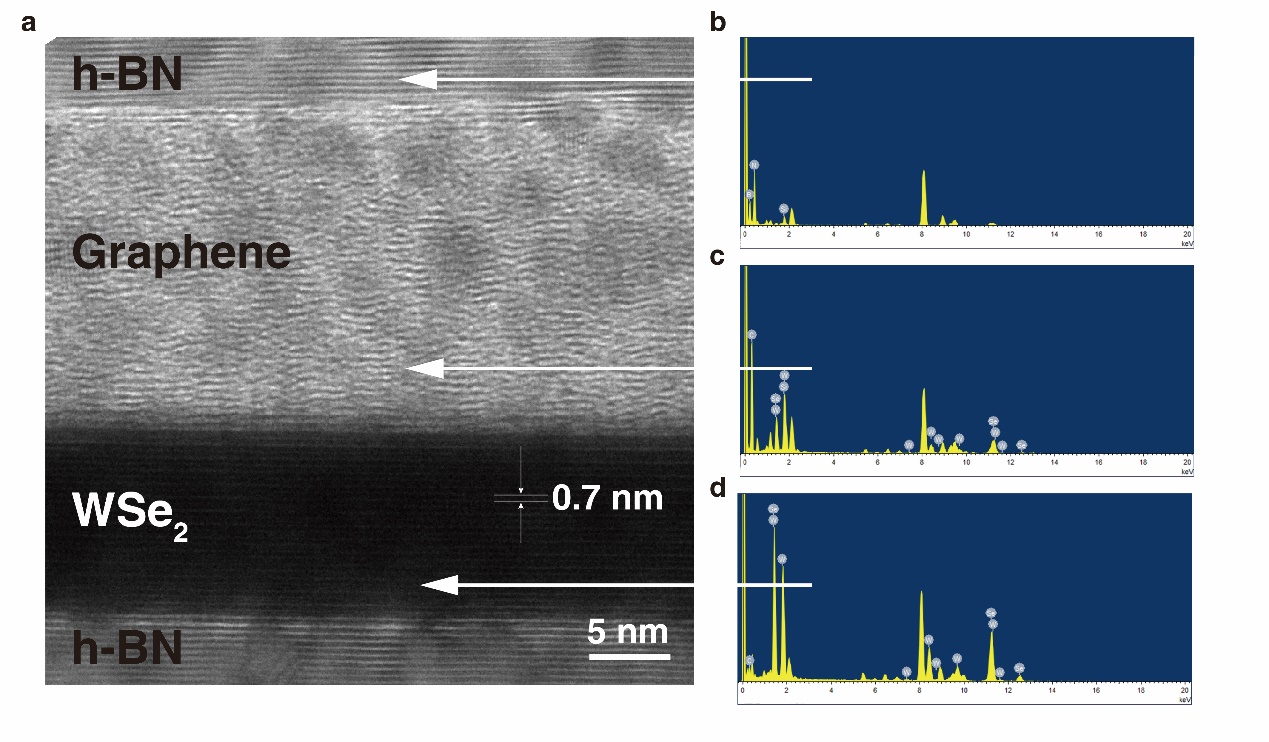
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Figure S17. Negative photoconductivity phenomenon at 400 ℃ and 500 ℃ under 0.2 W/m2 365 nm light illumination.

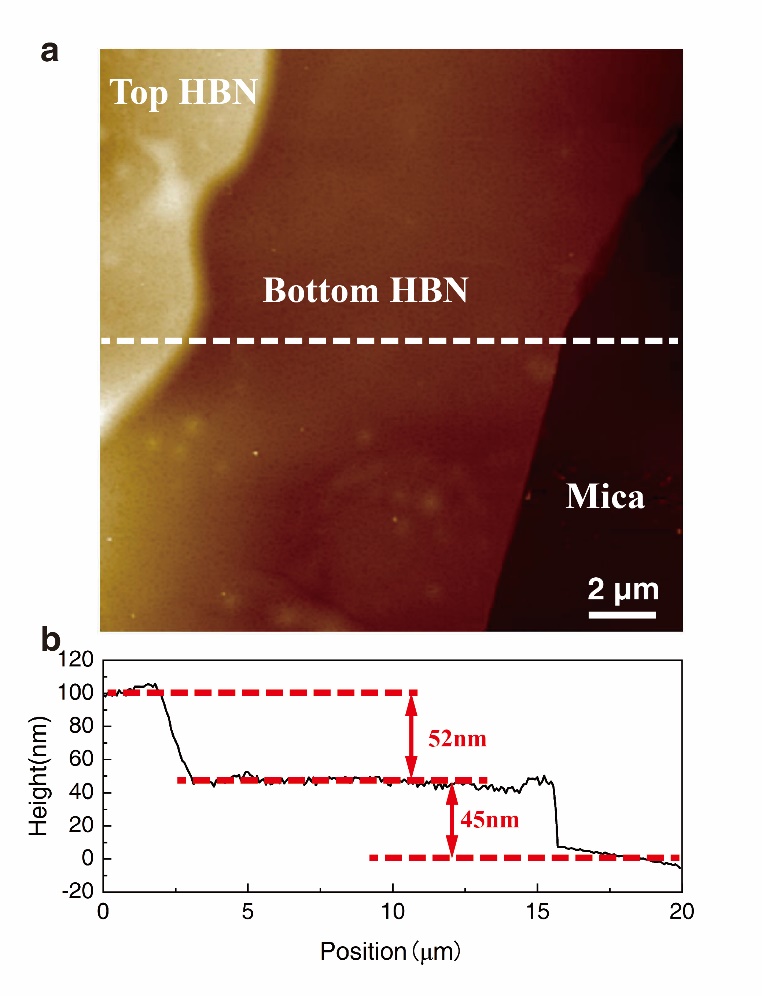


**Figure S1. Optical images of step-by-step transfer process of the stacked** van der **Waals heterostructures. a,** Transfer of the bottom h-BN. **b,** Transfer of the WSe2. **c,** Transfer of the two graphene electrodes. **d,** Transfer of the top h-BN.

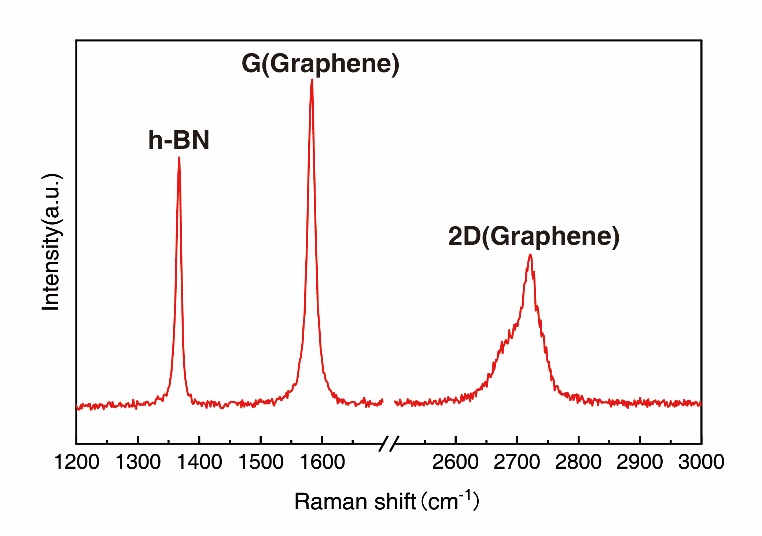
We exfoliated the mica sheet to about 100 μm thick to obtain clean, flat and flexible substrate. We cleaned the substrate using an ultrasonic cleaner to remove possible residues from the substrate surface. Then, mechanically exfoliated h-BN, WSe2 and two pieces of graphene as electrodes were transferred to the substrate step by step, as shown in (a), (b), (c). Another h-BN was transferred to form fully encapsulation, as shown in (d).



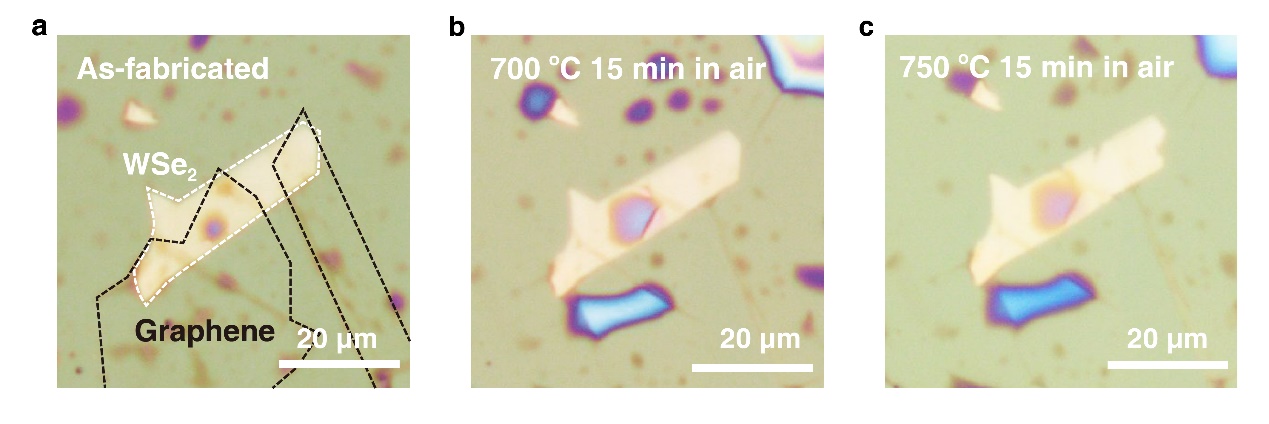
**Figure S2. High Resolution Transmission electron microscope (HRTEM) images of the stacked structure and elemental analysis results of different areas. a,** HRTEM image of the stacked structure. Clear lattice structure can be seen in the image, indicating excellent material quality. **b, c, d** are elemental analysis results of h-BN, graphene, WSe2, respectively. According to the results of element analysis, we can well determine the type of material in each area.



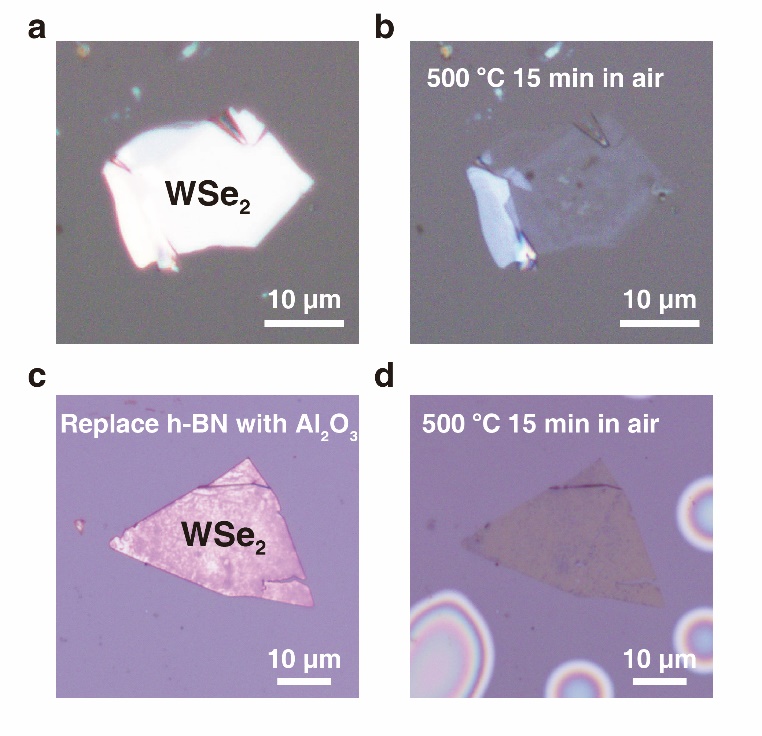
**Figure S3. Atomic force microscope (AFM) topographic image and height-position curve of h-BN. a,** AFM topographic image of bottom and top h-BN. **b,** Height-position curve along the dotted line in (a). The thickness of top h-BN is 52 nm and the bottom h-BN is 45 nm. Generally, 40-60 nm thick h-BN were preferred to be chosen considering both the thermal protection and gate dielectric isolation.



**Figure S4. Raman spectra of h-BN and graphene.** Three peaks at 1375 cm-1, 1580 cm-1 and 2720 cm-1 corresponded to the h-BN, G peak and 2D band of graphene, respectively. The sharp peaks indicate that the materials are of high quality and have no major defects.



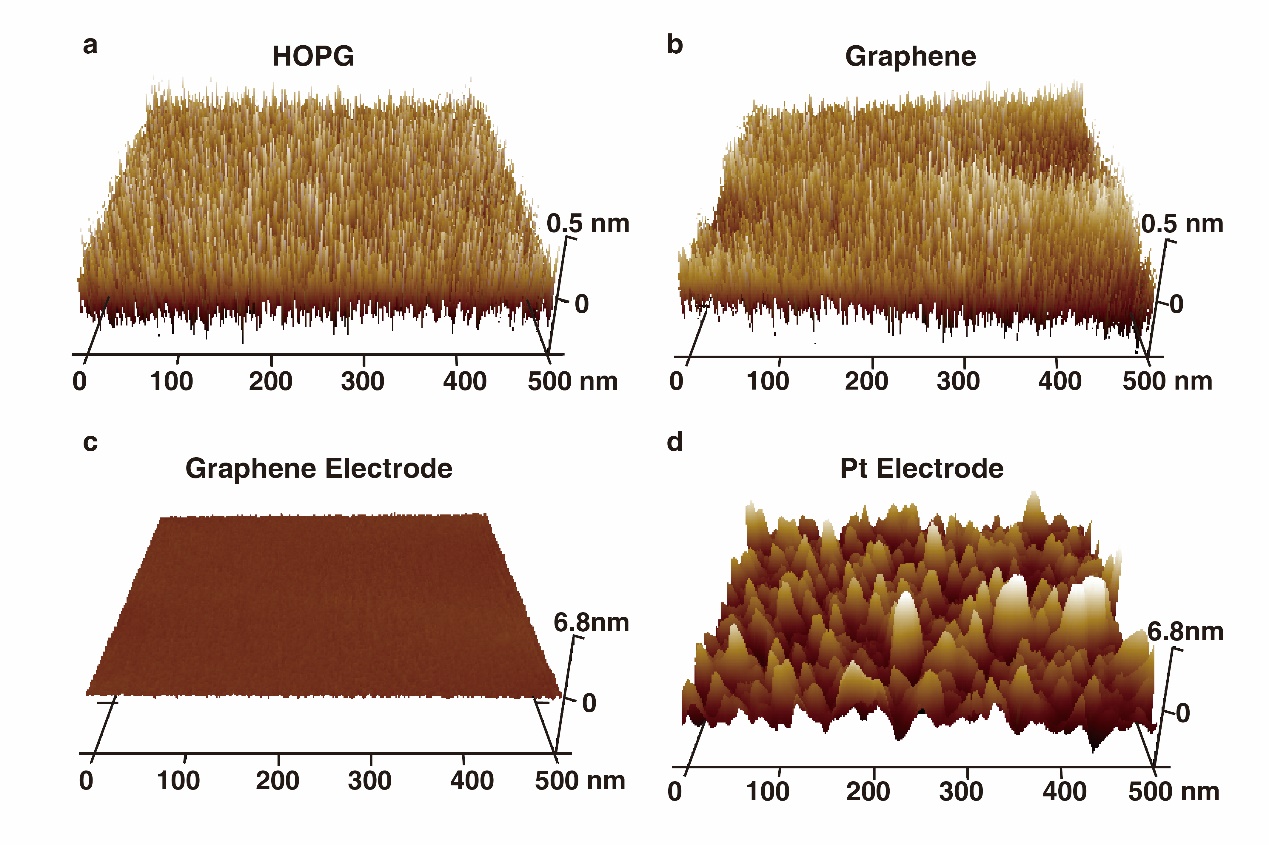
**Figure S5. Optical images of WSe2 FET before and after 700 ℃/750 ℃** **burning. a,** As-fabricated WSe2 FET with h-BN encapsulation and graphene electrodes. **b,** WSe2 FET after 700 ℃ heating for 15 min in air. No significant change of WSe2 was observed. **c,** WSe2 FET after 750 ℃ heating for 15 min in air. WSe2 showed only very small defects at the edge.



**Figure S6. Thermal stability of bare WSe2 and WSe2 covered with 100 nm Al2O3. a,** Optical image of exfoliated WSe2 on mica substrate. **b,** Optical image of the same WSe2 after 500 ℃ burning for 15 min in air. The WSe2 flake was strongly oxidized and became nearly transparent inasmuch as WO3 is transparent under visible light. **c,** Optical image of exfoliated WSe2 on mica substrate covered with 100 nm Al2O3. **d,** Optical image of the WSe2 covered with 100 nm Al2O3 after 500 ℃ burning for 15 min in air. The WSe2 flakes were strongly oxidized and became nearly transparent.

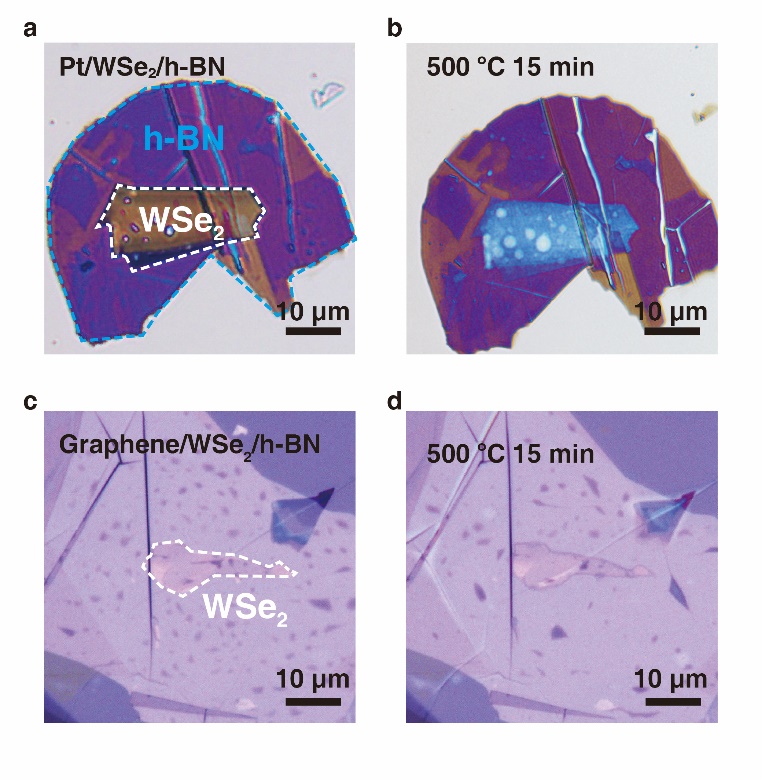


**Figure S7. WSe2 FET before and after 1000 ℃ burning in vacuum.** **a,** Bare WSe2 before 1000 ℃ burning. **b,** The WSe2 is vanished after 1000℃ burning. **c,** Raman spectrum of WSe2 after 1000℃ burning in vacuum. **d,** WSe2 FET before 1000℃ burning. **e,** WSe2 FET after 1000℃ burning in vacuum.



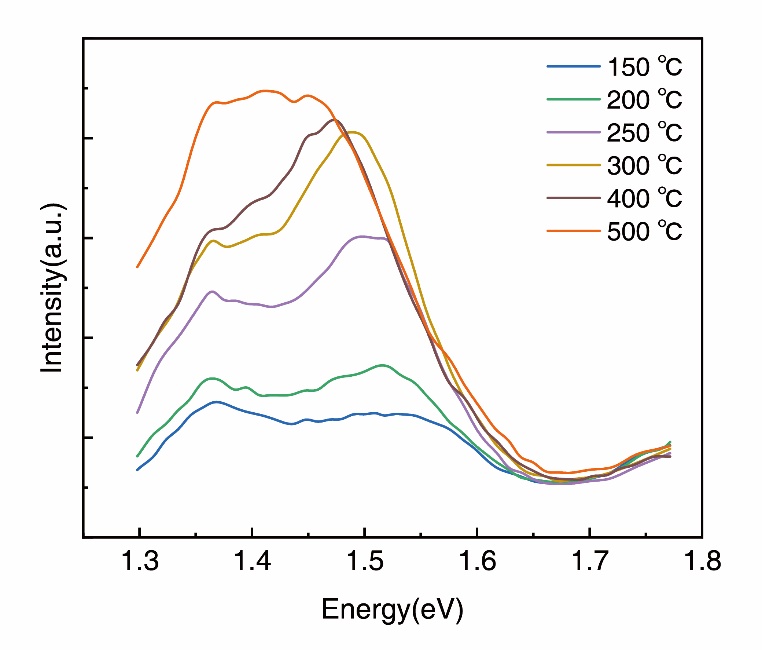
**Figure S8. Atomic force microscope (AFM) images of HOPG ((highly oriented pyrolytic graphite), graphene, and Pt electrode. (a)** AFM image of HOPG. **(b), (c)** AFM image of exfoliated graphene on h-BN in different Z direction scales. **(d)** AFM image of Pt electrode.

HOPG is reported to have atomic flat surface. The height variation of HOPG surface is ~0.5 nm and exfoliated graphene is ~0.6 nm, indicating that exfoliated graphene has a surface flatness close to that of HOPG. The height variation of Pt electrode is ~6.8 nm, much larger than that of graphene electrode (~0.6 nm). Instrument noise affected the measurement by approximately 0.4 nm, indicating that the height variation of graphene was mainly limited by instrument noise. The result demonstrated that exfoliated graphene had a much flatter surface than Pt electrodes. Consequently, good encapsulation can be formed between h-BN and graphene.

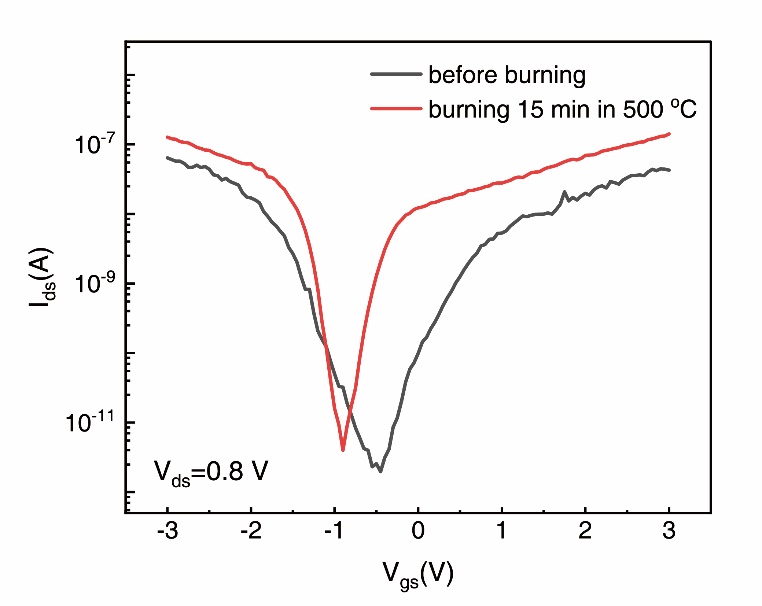


**Figure S9. Comparison of graphene and Pt as bottom encapsulation layer. a,** Optical images ofPt/WSe2/h-BN structure using 30 nm thick Pt as bottom layer. **b,** Optical images ofPt/WSe2/h-BN structure after burning 15 min at 500 °C. **c,** Optical images ofgraphene/WSe2/h-BN structure using exfoliated graphene as bottom layer. **d,** Optical images ofgraphene/WSe2/h-BN structure after burning 15 min at 500 °C.

The Pt/WSe2/h-BN structure device using 30 nm thick Pt as bottom layer shown in (a). Mechanically exfoliated WSe2 and h-BN were transferred to the Pt electrode sequentially. WSe2 was heavily oxidized after burning 15 min at 500 °C, indicating the poor oxidation protection of Pt/WSe2/h-BN structure, as shown in (b).Then we tested graphene/WSe2/h-BN structure using exfoliated graphene as bottom layer in same experimental condition. Contrary to former structure, WSe2 was well protected after burning 15 min at 500 °C, as shown in (d). The results demonstrated that graphene/WSe2/h-BN structure is the better way to prevent WSe2 from oxidation in high temperature.

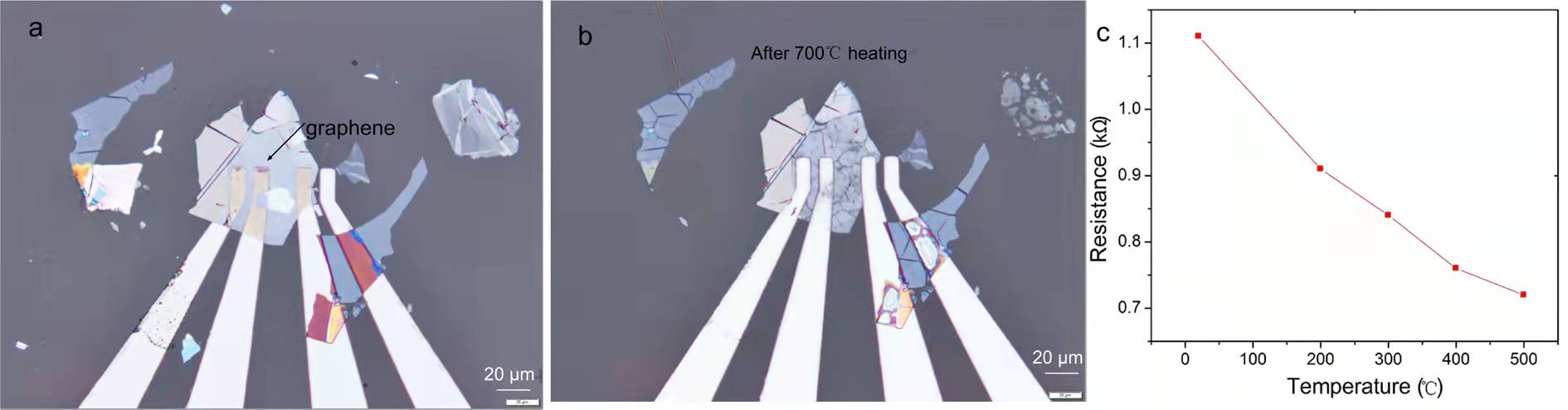


**Figure S10. Photoluminescence spectroscopy image of WSe2 in heat resistant structure from 150 ℃ to 500 ℃.** The leftward shift of the PL peak with increasing temperature indicated a decreasing energy bandgap of WSe2.

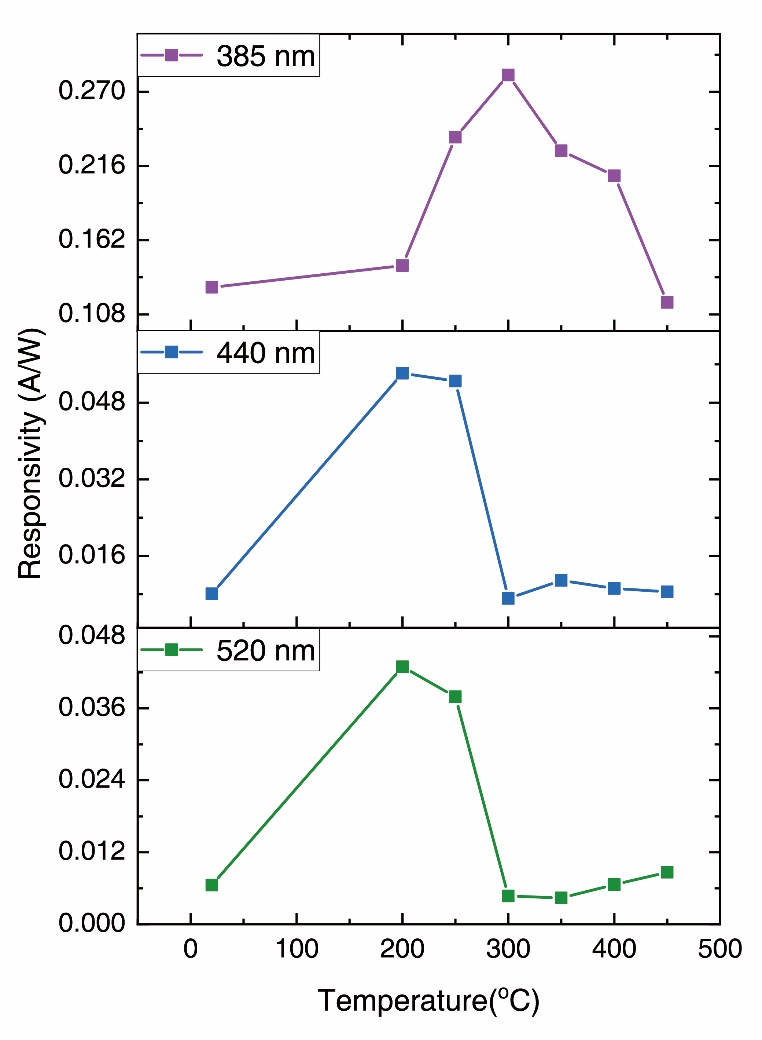


**Figure S11. Ids-Vgs curves (Vds=0.8V) measured at room temperature before and after burning 15 min at 500 °C in air.**

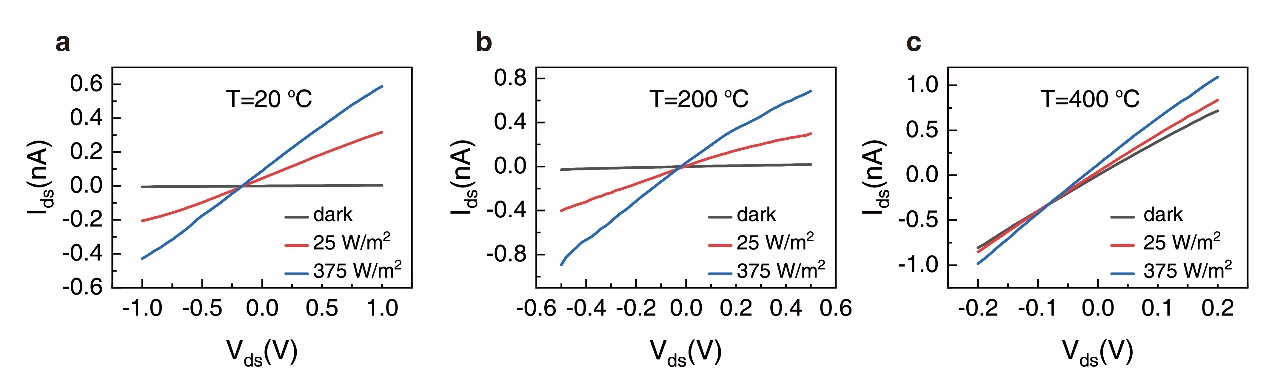
Both p-type and n-type currents were enhanced after burning at 500 °C in air, which may be due to improved contact by high temperature heating. The results further proved that heating at 500 °C will not cause significant damage to our devices, even improving the performance of the device as a result.



**Figure S12. Conducting properties of graphene electrode at high-temperature.** **a,** Graphene flake before heating. The resistance of the fresh graphene flake is 1.2 kΩ.  **b,** Graphene flake after 700℃ heating for 15 min in air. The bare graphene without h-BN protection is still in good shape with smaller resistance of 0.6 kΩ at room temperature. **c,** The resistance of a typical graphene flake reduces from 1.1 kΩ to 0.7 kΩ as temperature varied from 20℃ to 500℃.

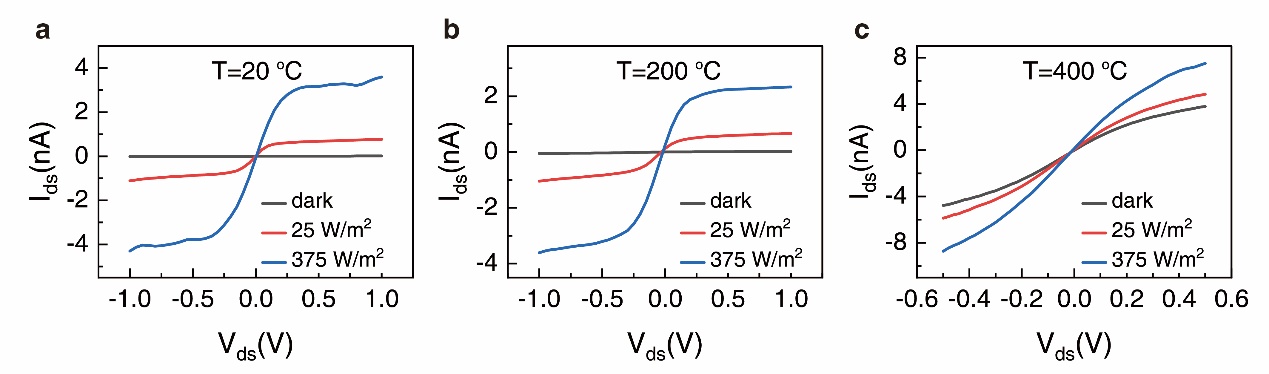


**Figure S13.** **Photo responsivity of WSe2 device with mica encapsulation under 385 nm (purple), 440 nm (blue), and 520 nm (green) illumination at different temperatures (*Vds*=0.4 V, *Vg*=0 V).** Under the illumination of three different lights, all the responsivity increased and then decreased with increasing temperature from 20 ℃ to 450 ℃ in air, but remains positive. The device with top mica exhibited positive photoconductivity in 400 ℃, indicating h-BN encapsulation contributed to the novel negative photoconductivity phenomenon in 400 °C.



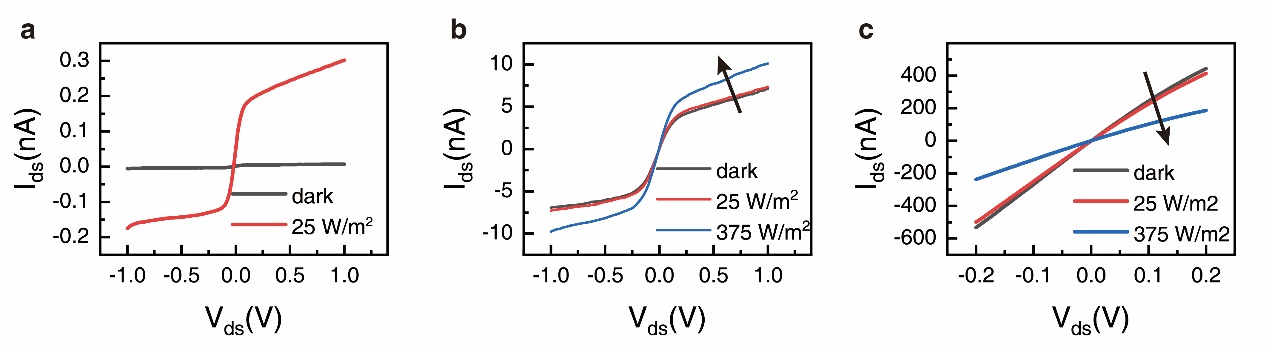
**Figure S14. The Ids-Vds curves of bare WSe2 with graphene electrodes (without top h-BN) under different light intensity and temperature. a,** The Ids-Vds curves of bare WSe2 with graphene electrodes under white light illumination at 20 °C. **b,** The Ids-Vds curves of bare WSe2 with graphene electrodes under white light illumination at 200 °C. **c,** The Ids-Vds curves of bare WSe2 with graphene electrodes under white light illumination at 400 °C.

At three different temperature, positive photoconductive effect was observed as illumination power increased, indicating that bare WSe2 with graphene electrodes (without top h-BN) showed positive photoconductive effect under illumination. The contribution of WSe2 and WSe2/graphene electrode contact to the novel photoconductivity phenomenon in 400 °C have been excluded.



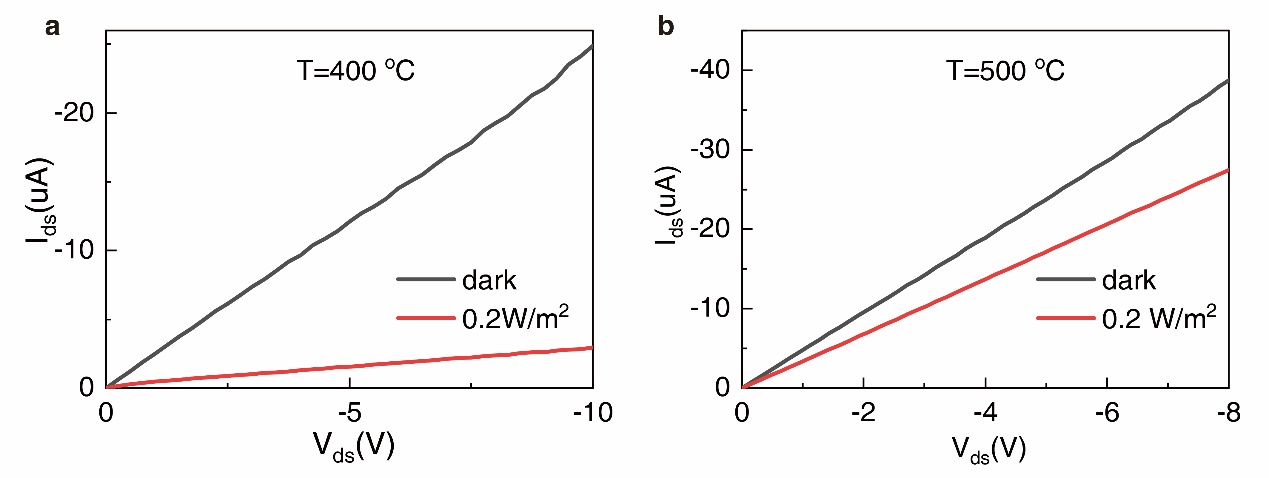
**Figure S15. The Ids-Vds curves of bare WSe2 with** **Pt electrodes (without top h-BN) under different light intensity and temperature. a,** The Ids-Vds curves of bare WSe2 with Pt electrodes under white light illumination at 20 °C. **b,** The Ids-Vds curves of bare WSe2 with Pt electrodes under white light illumination at 200 °C. **c,** The Ids-Vds curves of bare WSe2 with Pt electrodes under white light illumination at 400 °C.

In three different temperature, positive photoconductive effect was observed as illumination power increased, indicating that contact between WSe2 and Pt is not the cause of the negative photoconductivity phenomenon in 400 °C.



**Figure S16. The Ids-Vds curves of WSe2 with Pt S/D electrodes and h-BN encapsulation (without top gate) under different light intensity and temperature. a,** The Ids-Vds curves of bare WSe2 with Pt electrodes under white light illumination at 20 °C. **b,** The Ids-Vds curves of bare WSe2 with Pt electrodes under white light illumination at 200 °C. **c,** The Ids-Vds curves of bare WSe2 with Pt electrodes under white light illumination at 400 °C.

The current of WSe2 device increased as light intensity increased from 25 W/m2 to 375 W/m2 at 20 °C and 400 °C. The current decreased with increasing light intensity, indicating negative photoconductivity.



**Figure S17. Negative photoconductivity phenomenon at 400 ℃ and 500 ℃ under 0.2 W/m2 365 nm light illumination.** The photoresponsivity derived can reach 2.2×106 A/W and 1.1×106 A/W respectively.