Electronic Supplementary Information

Synthesis of semiconducting 2H phase WTe_2 nanosheets with large positive magnetoresistance

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S1. Experimental section

Materials

Tungsten powder and tellurium powder were commercially available from Aladdin Chemical Co. Ltd. N-butyl-lithium (n-BuLi) (2.5 M in hexane) was purchased from Sinopharm Chemical Reagent Co. Ltd and used without further purification.

Preparation of bulk WTe_2 powder

Bulk WTe_2 powder was synthesized by a high-temperature solid reaction. In detail, 1 mmol W powder and 2 mmol Te powder were ground to mix homogeneously and then sealed in an evacuated quartz tube. The tube was heated in a horizontal tube furnace at 900 °C for 12 h with a heating rate of about 3 °C/min. The system was then allowed to cool to room temperature with a cooling rate of about 5 °C/min. The product was directly collected from the quartz tube without washing.

Preparation of precursor Li_xWTe_2

Li_xWTe_2 precursor was synthesized through a lithium intercalation process. Typically, the 40 mg of WTe_2 powder was dispersed in 4 mL of hexane with 1.0 mL of n-BuLi in a customized glass vacuum system. The reaction system was stirred and kept at 60°C for 12 h. The obtained dispersion was filtered, washed with cyclohexane and anhydrous ethanol for several times, and dried at 60°C for 30 min under vacuum.

Exfoliation of Li_xWTe_2 into WTe_2 ultrathin nanosheets

In a typically procedure, 20 mg of the as-obtained Li_xWTe_2 precursor and 20 mL of formamide (FA)/ethanol mixture (V_{FA}/V_{EtOH} = 9:1) were loaded in a weighing bottle with a total capacity of 20 mL. Then the solution was bubbled with nitrogen to eliminate the dissolved oxygen molecules for avoiding oxidation. The above dispersion was ultra-sonicated in ice water for 4 h. Here, the ice water was used to keep the system in a relatively low temperature. Afterward, the resultant suspension was centrifuged at 2000 rpm for 3 min to remove the residual un-exfoliated particles, and the supernatant was finally collected by high-speed centrifugation and washed by ethanol for further characterizations.

Characterizations

X-ray diffraction (XRD) was performed on a Philips X'Pert ProSuper diffractometer with Cu Kα radiation (λ = 1.54178 Å). The field emission scanning electron microscopy (FE-SEM) images were taken on a JEOL JSM-6700F SEM. The transmission electron microscopy (TEM) image was carried out on a JEM-2100F field emission electron microscope at an acceleration voltage of 200 kV. High-angle annular dark-field scanning transmission spectroscopy (HAADF-STEM) image and corresponding energy-dispersive spectroscopy mapping analyses were performed on a JEOL JEM-ARF 200F TEM/STEM with a spherical aberration corrector. Raman spectra were recorded on a Renishaw RM3000 Micro-Raman system with a 514.5 nm Ar laser. X-ray photoelectron spectra (XPS) was acquired on an ESCALAB MK Ⅱ with Mg K_α as the excitation source. The electrical transport property measurements were carried out using a Keithley 4200-SCS Semiconductor Charactetization System and a four-point probe method. Magnetoresistance was measured by a four-point

technique using a Quantum Design Physical Property Measurement System (PPMS)-9 using the van der Pauw method. Electrochemical station (CHI 660B) was used for the photodetector measurement. The photodetector based on WTe_2 nanosheets was fabricated by depositing the WTe_2 nanosheets on the gap between the ITO electrode.[1]

DFT calculation details

The electronic structure calculations have been performed by the VASP package (PAW).[2-4] To achieve the accurate density of the electronic states, the plane wave cutoff energy was 500 eV, a 7×7×5 for bulk Td WTe_2 and 11×11×1 for 2H WTe_2 nanosheet k-point mesh were used. Ionic relaxations were carried out under the conventional energy (10^{-4} eV) and force (0.02 eV/Å) convergence criteria. Of note, from the HRTEM data (d_{100}=0.307 nm) and XRD, we calculated the unit cell parameters of the 2H nanosheet are a=b=0.355 nm, c=1.4 nm, while the unit cell parameters of the Td bulk are a=0.347 nm, b=0.625 nm, c=1.4 nm.

S2. Additional characterization information

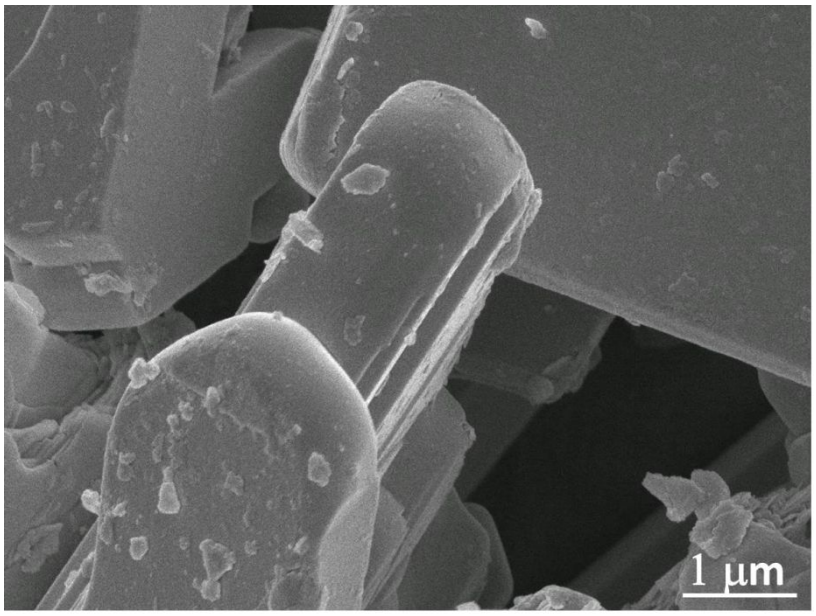

Figure S1. SEM image of the bulk Td WTe_2.

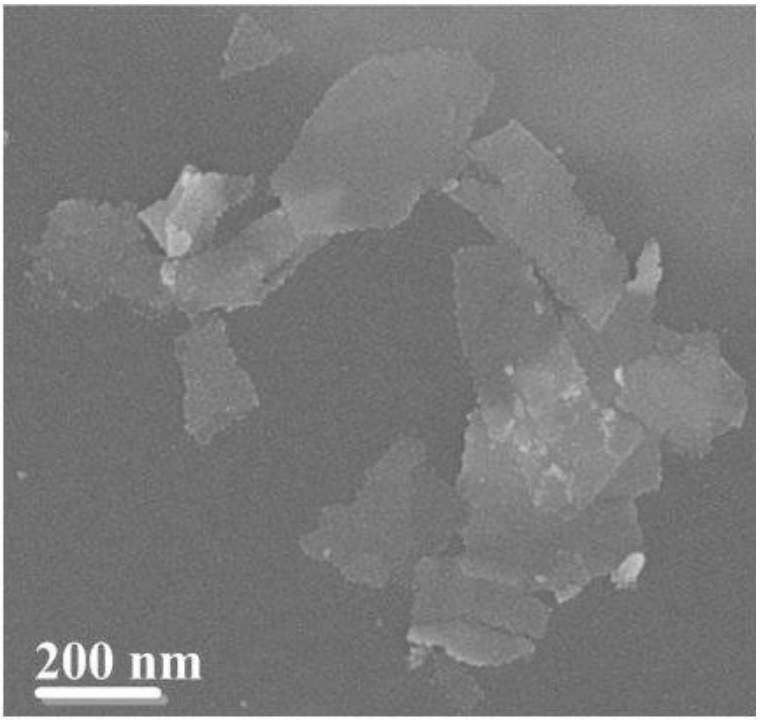

Figure S2. SEM image of the 2H WTe_2 ultrathin nanosheets.

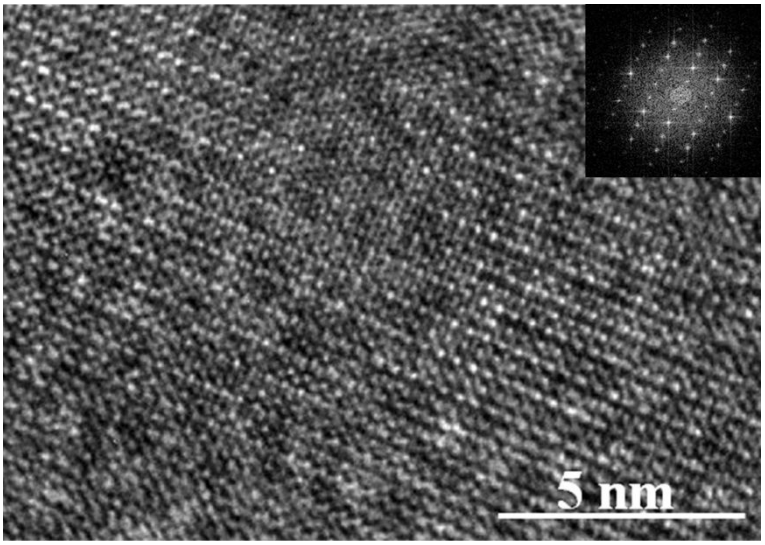

Figure S3. The HRTEM and corresponding FFT images of bulk WTe_2 with the typically orthorhombic phase.

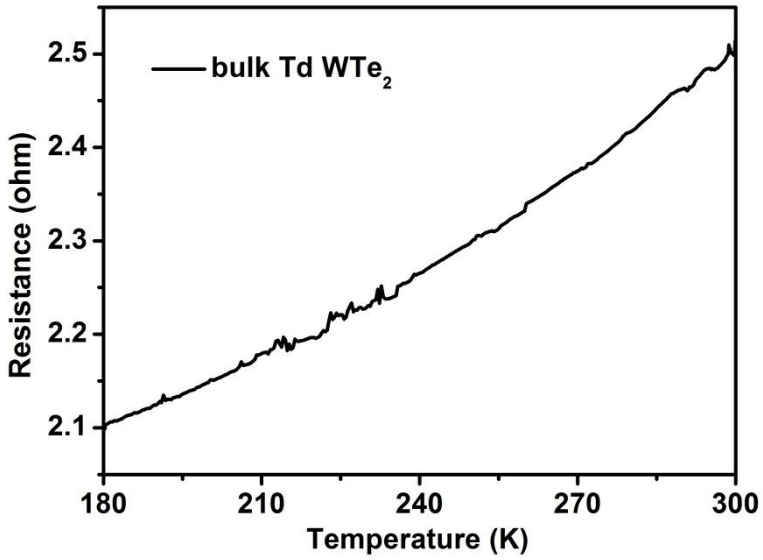

Figure S4. Temperature-dependent resistance of bulk Td WTe_2.

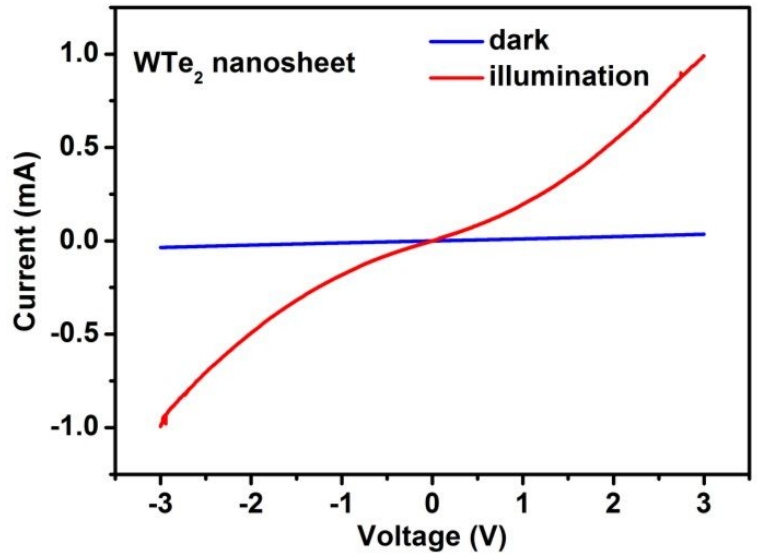

Figure S5. Photocurrent versus voltage plots of the as-exfoliated WTe_2 ultrathin nanosheet in the dark and under light illumination.

References

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