Supplementary Information

Atomic Thin Telluride Superlattices: Toward Spatial Modulation of Bandgaps

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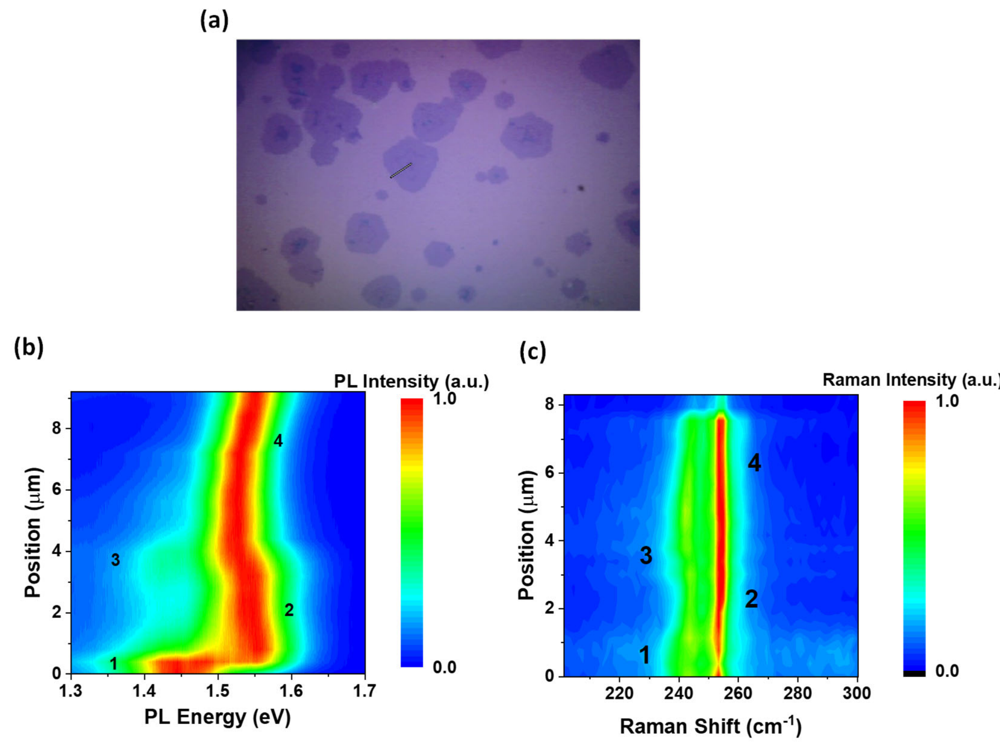

Figure S1. The optical image, PL and Raman contour plots of the fabricated samples in the same growth condition and different batches

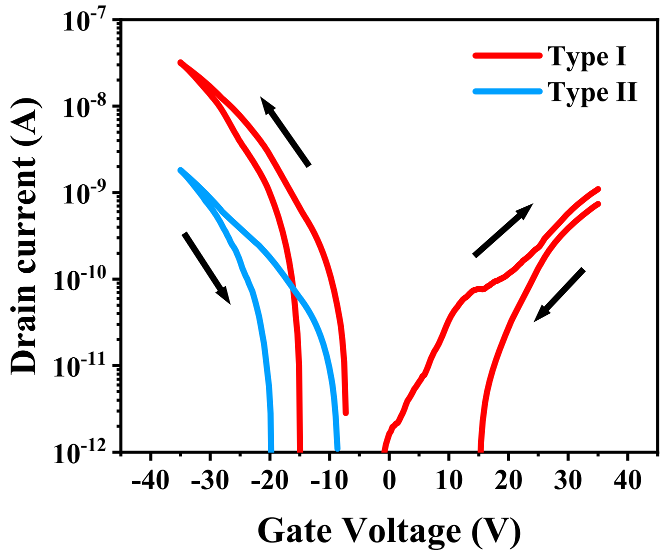

Figure S2. The transfer curves with double sweeping. A hysteresis loops exist in the transfer curves and corresponding sweep directions are labelled.

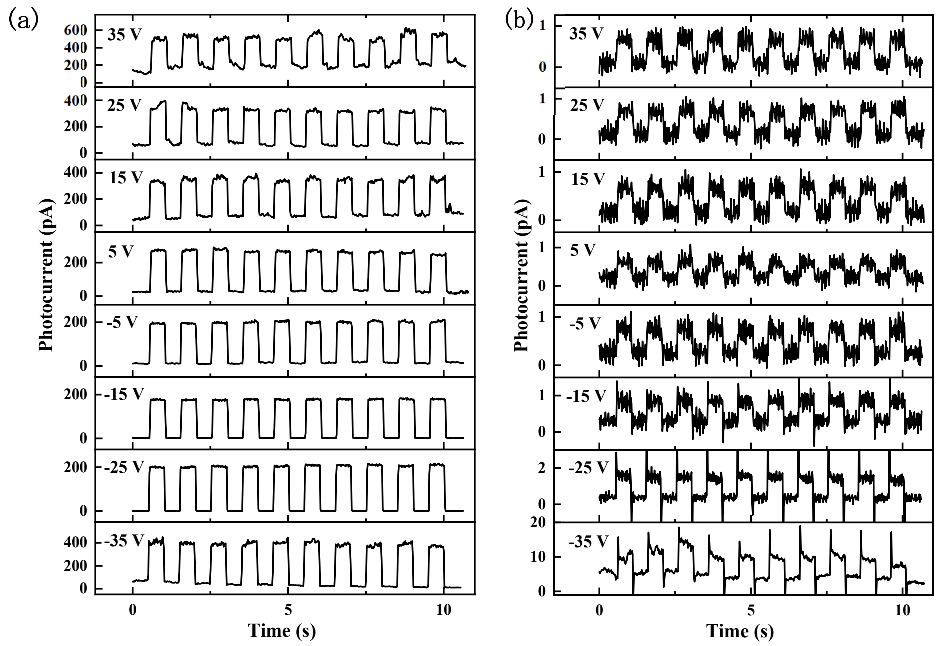

Figure S3. The time-dependent drain current with and without laser illumination measured at various gate voltages for (a) type I and (b) type II $WSe_{2-2x}Te_{2x}$/$WSe_{2-2y}Te_{2y}$ transistors. The photocurrents were extracted by subtracting the laser-on currents by the laser-off currents.

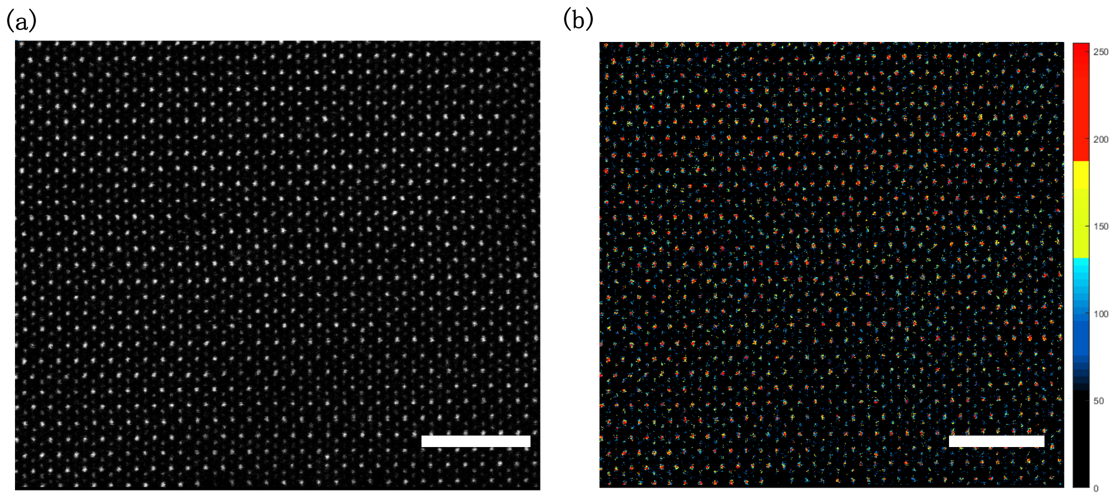

Figure S4. High-angle annular dark-field (HAADF) scanning transmission electron microscopy (STEM) images taken in zone 4 of $WSe_{2-2x}Te_{2x}/WSe_{2-2y}Te_{2y}$ multiheterostructures. Scale bar: 2 nm. (a) Original atomic-resolution HAADF-STEM image. (b) Processed image for Te mole fraction estimation. Red, yellow, light blue and dark blue color spots roughly correspond to W, Te_2, SeTe and Se_2 sites, respectively.

The HAADF-STEM image contrast has a strong dependence on the effective atomic numbers of the elements. The list of the effective atomic numbers of the elements in this alloy is shown in Table S1. Based on the STEM contrast, a Te mole concentration of 20.5% is extracted in Zone 4. Considering the bandgap of the alloy in zone 4 is 1.56 eV, these results are consistent with reference [1], where 20% Te corresponds to ~1.55 eV optical bandgap.

Elements	Effective Atomic number (Z)	$Z^{1.7}$
Se	34	401.34158
Te	52	826.42926
W	74	1505.54563
Se_2	48.1	723.84675
Se-Te	62.1	1117.52107
Te_2	73.5	1488.29313

Table S1. Effective atomic numbers (Z) of various elements in $WSe_{2-2x}Te_{2x}/WSe_{2-2y}Te_{2y}$ multiheterostructure. The HAADF-STEM intensity is roughly proportional to $Z^{1.7}$ [2], thus enabling the manual counting of Te mole fraction based on STEM images. Here, the effective atomic numbers of Se_2, Se-Te and Te_2 were consistent with reference [3].

The Te concentrations were also estimated based on the optical bandgap measured by PL, shown in Table S2, using the correlation between the optical bandgap and the Te composition in reference [1].

Zone	1	2	3	4
Optical bandgap (eV)	1.44	1.53	1.44	1.56
Te concentration (%)	40.0	23.6	40.0	18.1

Table S2. Optical bandgaps extracted from PL and the estimated Te concentrations in zone 1-4 of $WSe_{2-2x}Te_{2x}/WSe_{2-2y}Te_{2y}$ multiheterostructure.

TEM sample preparation:

Polymethyl methacrylate (PMMA) was first spin-coated onto the SiO_2/Si substrate with $WSe_{2-2x}Te_{2x}/WSe_{2-2y}Te_{2y}$ flakes. Then, the sample was soaked in 2 molar KOH solution for 3 hours to peel off $WSe_{2-2x}Te_{2x}/WSe_{2-2y}Te_{2y}$ flakes with the aid of supporting PMMA layer from the SiO_2/Si substrate. The PMMA-$WSe_{2-2x}Te_{2x}/WSe_{2-2y}Te_{2y}$ layer was then transferred onto TEM Cu grid with continuous carbon film (CF100H-CU, Electron Microscopy Sciences). The PMMA was then removed by soaking the TEM Cu grid with $WSe_{2-2x}Te_{2x}/WSe_{2-2y}Te_{2y}$ flakes into acetone and isopropanol (IPA). Finally, the sample was dried on a hotplate at 60 °C for 10 minutes.

STEM Z-contrast imaging:

The $WSe_{2-2x}Te_{2x}/WSe_{2-2y}Te_{2y}$ flakes were characterized by high-angle annular darkfield–scanning transmission electron microscopy (HAADF–STEM) on a ThermoFisher Scientific Themis Z TEM/STEM instrument operated at 300 keV. The instrument is equipped with a high brightness Schottky 'extreme' field emission gun (X-FEG) electron source, an electron energy monochromator, and a 5th-order probe spherical aberration corrector (DCOR). These features enabled STEM with sub-Angstrom imaging spatial resolution without introduction of aberrations. These advanced features, along with a modern drift corrected frame integration (DCFI) algorithm, enabled the high spatial resolution of current 2D materials with atomic resolution. To minimize the electron beam damage to the sample, a small probe current of 10 pA is used for the HAADF-STEM imaging. The imaging was set at a specific condition that a high collection angle of scattered electrons ranging from 50 to 200 mrad was used to form the image. In this condition, the image contrast exhibits a high dependence on Z (with contrast approximately proportional to $Z^{1.7}$), therefore it is feasible to identify atoms of high Z and lower Z elements directly from the image.

References:

[1] P. Yu *et al.*, "Metal–Semiconductor Phase-Transition in $WSe_{2(1-x)}Te_{2x}$ Monolayer," *Advanced Materials,* vol. 29, no. 4, p. 1603991, 2017.

[2] B. Xu, C. R. Fell, M. Chi, and Y. S. Meng, "Identifying surface structural changes in layered Li-excess nickel manganese oxides in high voltage lithium ion batteries: A joint experimental and theoretical study," *Energy & Environmental Science,* vol. 4, no. 6, pp. 2223-2233, 2011.

[3] A. Apte *et al.*, "Telluride-Based Atomically Thin Layers of Ternary Two-Dimensional Transition Metal Dichalcogenide Alloys," *Chemistry of Materials,* vol. 30, no. 20, pp. 7262-7268, 2018.