Supporting Information

High Performance WS_2 Monolayer Light Emitting Tunneling Devices using 2D Materials Grown by Chemical Vapor Deposition

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S1. Device fabrication process

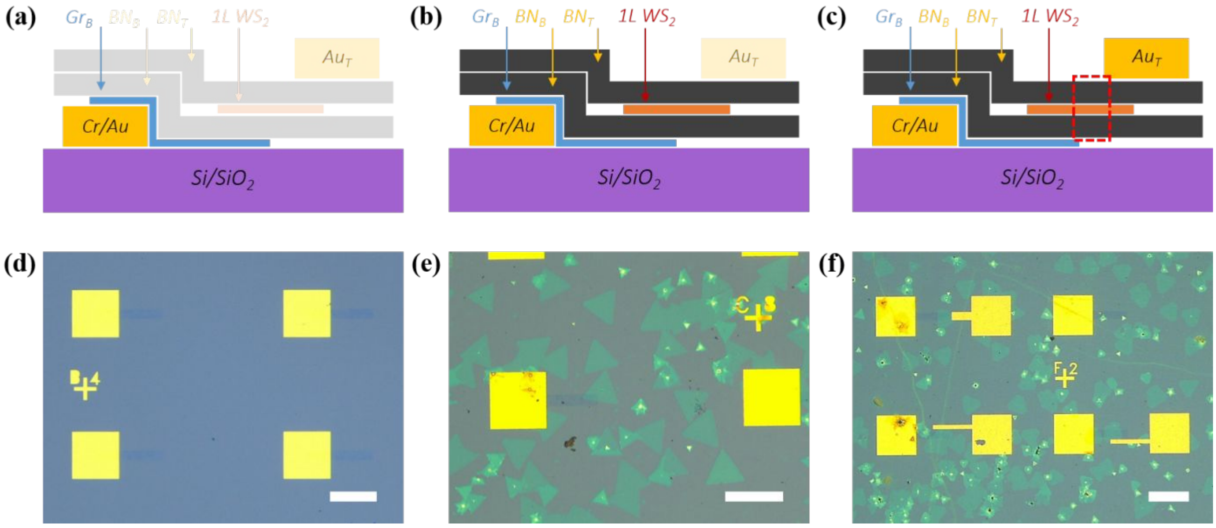

Figure S1. Device fabrication process. (a) Schematics of the asymmetric structure featuring the first two steps including depositing bottom bond pads (step 1) and fabricating graphene electrodes (step 2). (b) Schematic diagram of the structure after step 3, 4 and 5 (*i.e.* transferring bottom BN, monolayer WS_2 and top BN). (c) Schematic illustration of the final structure with customized top gold bond pad deposited (step 6). (d) The corresponding optical image showing bottom gold bond pads and graphene ribbons. Scale bar: 100 µm. (e) Optical image of a typical area after transferring three layers of 2D materials. Scale bar: 100 µm. (f) An optical image of the final device, showing three kinds of top bond pad designs. Scale bar: 100 µm.

S2. Customized top bond pad designs

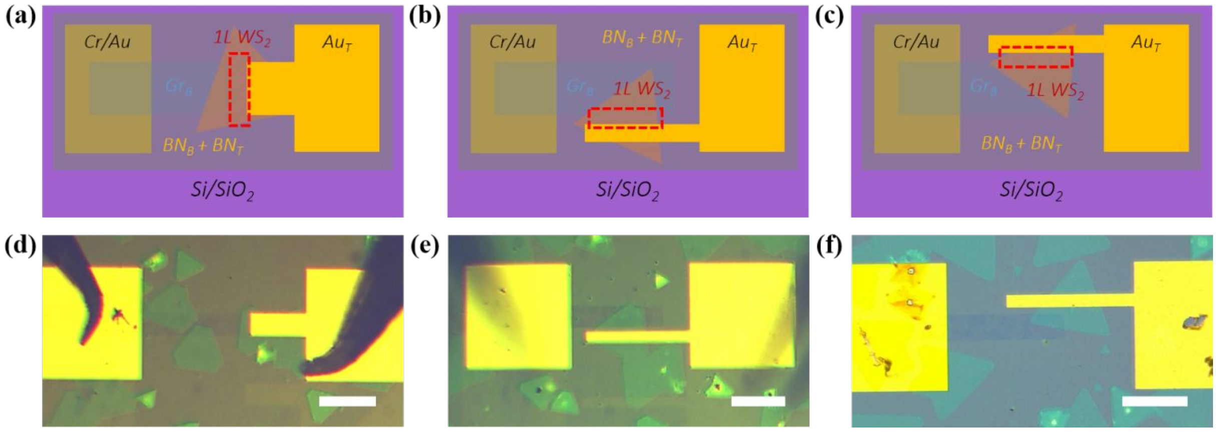

Figure S2. Customized top bond pad designs. (a-c) Schematic illustration showing three different types of top bond pad designs. (d-f) The corresponding optical images of the devices. Each device has a channel length of 5 µm. Scale bar: 50 µm.

S3. Overview of the final product

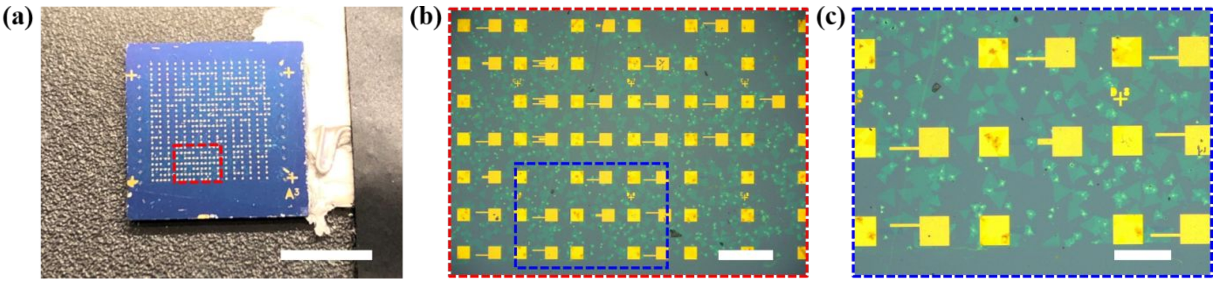

Figure S3. Overview of the final product. (a) Photo of the devices after all the fabrication process, on a 1 cm × 1 cm chip. Scale bar: 5 mm. (b) Optical image of the red dashed rectangle in (a), showing different designs of the top bond pad. Scale bar: 500 µm. (c) Zoomed-in optical image of the blue dashed rectangle in (b). Scale bar: 200 µm.

S4. Characterization of multilayer BN

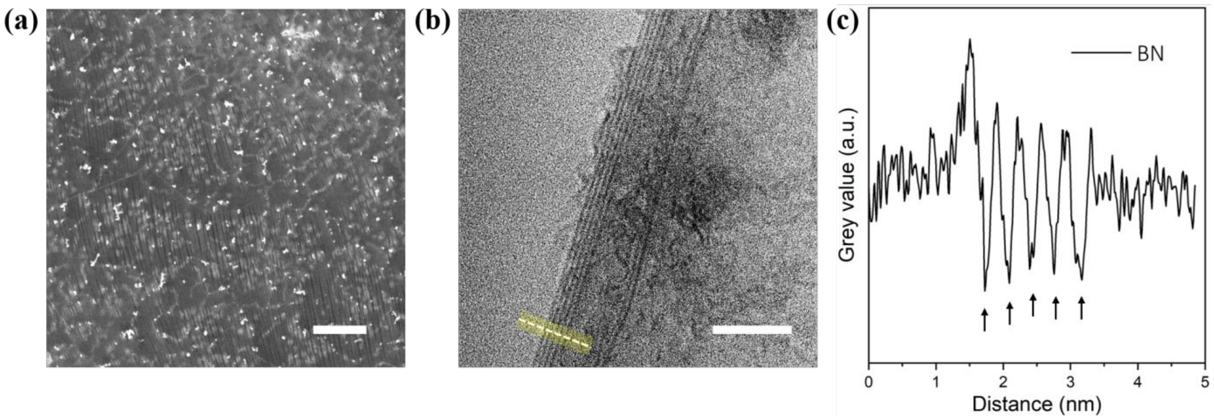

Figure S4. Characterization of multilayer BN. (a) SEM image presenting a typical area of the multilayer CVD-grown BN film. Scale bar: 2.5 μm. (b) Low-magnification TEM image showing the edges of the material. Scale bar: 5 nm. (c) The corresponding line profile taken from the yellow marked region in (b). The black arrows indicates that our BN film is 5 layers thick.

S5. Robustness testing when V_{ds} = −10V

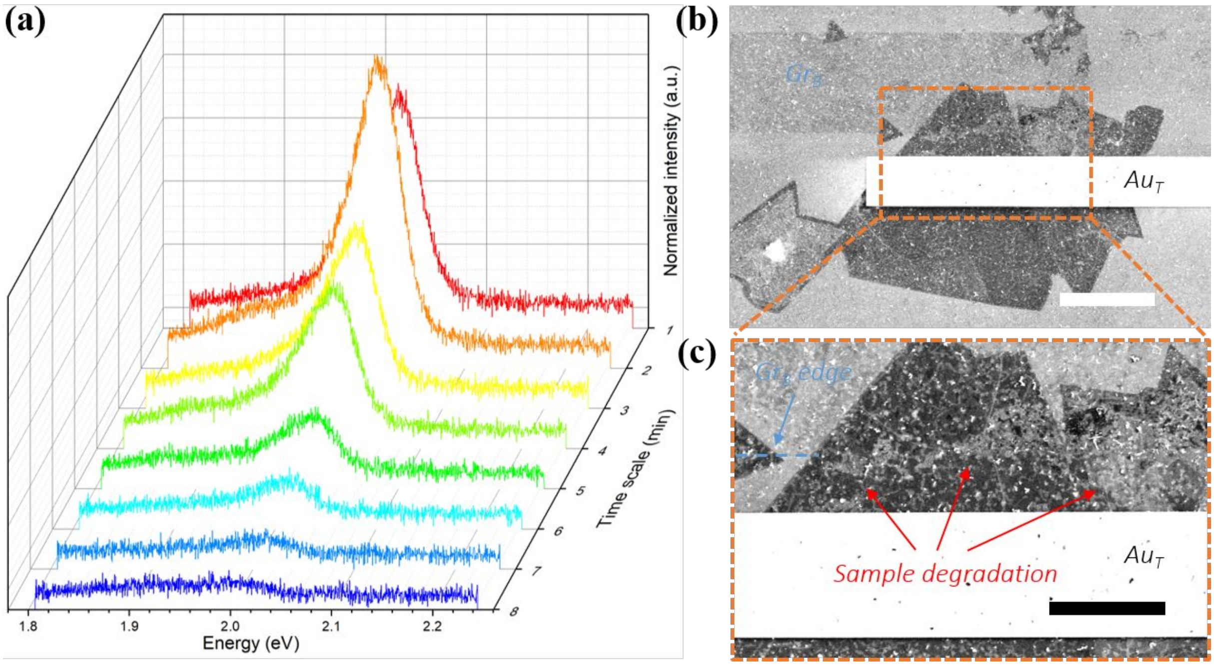

Figure S5. Robustness testing of the device under a source-drain bias of 10V. (a) A time-series of normalized EL spectra when V_{ds} = −10V. With a higher and continuing negative voltage (−10V), the EL intensity started to decrease dramatically after 4 min biasing. (b) SEM image of a light-emitting device after applying a higher and continuing bias for 8 min. Scale bar: 20 μm. (c) Zoomed-in SEM image of the orange dashed rectangle in (b), indicating that the device breakdown results from sample degradation. Scale bar: 10 μm.

S6. Quantum efficiency estimation

The external LED quantum efficiency (EQE) can be calculated as follows:

$$EQE = \frac{N_{LED-photons}}{N_{electrons}}$$

, where $N_{photons}$ is the number of photons emitted per second by LED and $N_{electrons}$ is the number of electrons injected per second.

During our measurement, the EL spectrum is collected by a spectrometer microscope system used for PL measurement. Herein, we are able to estimate the number of photons emitted with help of PL spectrum.

The number of photons emitted per second from photoluminescence ($N_{PL\text{-}photons}$) and electroluminescence ($N_{EL\text{-}photons}$) received by our spectrometer can be estimated as follows:

$$N_{PL-photons} = \eta_{PL}\eta_{collection}N_{laser} = \eta_{PL}\eta_{collection}\frac{P_{laser} \times \lambda}{hc}$$

$$N_{EL-photons} = \eta_{collection}N_{LED-photons} = \eta_{collection} \times EQE \times N_{electrons}$$

, where η_{PL} is the photoluminescence quantum yield (PLQY) of monolayer WS_2, $\eta_{collection}$ is the collection efficiency of our spectrometer, including all the optical components in the optical circuit, N_{laser} is the number of laser photons injected into the sample, P_{laser} is the laser injection power, h is the Planck's constant (6.63×10^{-34} J·s), c is the speed of light (3×10^{8} m/s), λ is the laser wavelength, which is 532 nm in our case.

By adjusting the excitation laser intensity, we can normalize the PL and EL spectrum by obtaining a PL spectrum of monolayer WS_2 that is very similar to our EL spectrum (acquisition time = 1s) in terms of shape and intensity, as well as its corresponding laser injection power P_{laser} (W/s). Then the EQE can be expressed as follows:

$$\frac{N_{EL-photons}}{N_{PL-photons}} = 1 = \frac{EQE \times N_{electrons}}{\eta_{PL}\frac{P_{laser} \times \lambda}{hc}}$$

$$EQE = \frac{\eta_{PL}P_{laser} \times \lambda}{N_{electrons} \times hc}$$

The number of electrons per second can be calculated from the injection current as:

$$N_{electrons} = \frac{I}{e}$$

, where I is the injection current and e is the charge of one electron (1.6×10^{-19} C).

Therefore, the LED quantum efficiency can be calculated as:

$$EQE = \frac{\eta_{PL} P_{laser} e\lambda}{I \times hc}$$

According to previous report,[1,2] the PLQY of monolayer WS_2 (η_{PL}) is about 1.5~6%. Then we can estimate that the EQE of our LEDs is about 0.69~2.78% when $V_{sd}<0$ and 1.13~4.52% when $V_{sd}>0$.

Reference

(1) Yuan, L.; Huang, L. Exciton Dynamics and Annihilation in WS_2 2D Semiconductors. *Nanoscale* **2015**, *7*, 7402-7408.

(2) Liu, J.; Lo, T. W.; Sun, J.; Yip, C. T.; Lam, C. H.; Lei, D. Y. A Comprehensive Comparison Study on the Vibrational and Optical Properties of CVD-Grown and Mechanically Exfoliated Few-Layered WS_2. *J. Mater. Chem. C* **2017**, *5*, 11239-11245.