Supporting Information

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High-Speed Electroluminescence Modulation in Monolayer WS$_2$

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Experimental Section

PL and EL measurements.

A source meter (Keithley 2612A) For I-V curves and EL measurements was used. Optical and electrical investigations were carried out in a cryostat (Oxford Instruments) under high vacuum (10⁻⁶ mbar), integrated in a scanning micro-PL setup. The setup includes a dichroic mirror at λ = 550 nm to separate the PL emission from the excitation. PL and EL were collected using a ×50 long working distance objective lens (NA = 0.5). In PL experiments, the sample was excited by a continuous-wave 532 nm laser. the laser beam size on the sample was ~1 μm. The PL and EL emission were spectrally filtered with a monochromator (Horiba iHR320) and detected with a liquid nitrogen cooled charge-coupled device (CCD). EL image was observed by using a CCD camera (Thorlabs).

Device fabrication

Bulk WS₂ (purchased from HQ Graphene), graphite and hBN were exfoliated by an adhesive tape and transferred onto a SiO₂/Si. The substrates were cleaned by an acetone/isopropanol bath. They were then annealed at 150 °C for 30 min in a dry-air atmosphere to remove adsorbates before the transfer of the exfoliated materials. Then, the tape was peeled-off and the desire flakes were selected with optical microscopy. A dry transfer technique was used to pick up the flakes by a polypropylene carbonate/polydimethylsiloxane (PPC/PDMS) stamp, starting with the top graphene. The heterostructure (Graphene/hBN/Graphene/WS₂/hBN) was then transferred on a clean SiO₂/Si substrate. The transfer process was performed in a nitrogen-purged glovebox. The remaining PPC residues were removed by introducing the
sample in an acetone/chloroform bath. Finally, an additional electrode was fabricated by means of electron-beam lithography and metal evaporation to make an electrical contact to the graphene flakes (Ti/Au, 3/60 nm).

**Frequency Modulation**

In order to investigate the time-resolved light emission from WS₂ tunneling light emitting diodes, we use time-correlated single photon counter (TCSPC) method. The devices were operated by a source meter (Keithley 2612A) and RF signal generator (Hewlett Packard 8648C) to form a sinusoidal signal. Electrically generated photons from the device operation by the AC application were collected to two Si avalanche photodetectors (APDs) via a beam splitter. The electrical signals from APDs are sent to the TCSPC and they are used as start and stop triggers to measure the delay of photon arrival times. For the TCSPC measurement setup, the detection limit depends on the timing jitter (~350 ps) of the APDs (SPCM-AQR-14). The limited timing jitter of the detector can decrease the visibility of the frequency modulation because it is close to our device speed of 1.5 GHz.

![I-V curves of tunnel WS₂ diodes with different top h-BN thicknesses. The increase of h-BN thickness causes the high threshold voltage.](image)

**Figure S1.** I-V curves of tunnel WS₂ diodes with different top h-BN thicknesses. The increase of h-BN thickness causes the high threshold voltage.
Figure S2. PL intensity map of our tunnel WS$_2$ diode with a continuous 532 nm laser and optical power of 5 µW.
Figure S3. PL spectra from our device with different gate voltages of 0, 40 and 80 V₉.

Figure S4. PL spectra of our tunnel WS₂ diode with different gate voltages of 0 and -40 V₉ at low temperature of 5 K.
Figure S5. (a-g) TCSPC histograms of EL electrically modulated at frequencies of 100, 200, 500, 800, 1000, 1200, and 1500 MHz ($V_d = 6.5$ V, $V_p = 1.6$ V). The raw results were fitted by red sine curves. (h-n) Histogram data from a-g for two oscillation periods.
Figure S6. Oscillation amplitude as a function of the modulated frequency at $V_d = 6.5 \text{ V}$. The red dash line indicates a gain of 3 dB relative to the amplitude value of device at 100 MHz. We estimate a cutoff frequency of $f_c \sim 600 \text{ MHz}$.

Figure S7. (a) Optical output from TCSPC method with a modulation frequency of 500 MHz and different DC offset voltages of 5.5, 6, and 6.5 $V_d$ at fixed count time. The raw results of optical output were fitted by sine waves (b) Optical output from TCSPC method with a modulation frequency of 500 MHz and different RF modulation power of 6, 10, and 14 V dBm at fixed count time. The raw results of optical output were fitted by sine waves. (c) Normalized sine waves with different DC offset voltages of 5.5, 6, and 6.5 $V_d$ at fixed maximum count. (d) Normalized sine waves with different RF modulation power of 6, 10, and 14 V dBm at fixed maximum count.