Supplemental Materials for

Interlayer Exciton Optoelectronics in a 2D Heterostructure p-n Junction

Jason S. Ross[1], Pasqual Rivera[2], John Schaibley[2], Eric Lee-Wong[2], Hongyi Yu[3], Takashi Taniguchi[4], Kenji Watanabe[4], Jiaqiang Yan[5,6], David Mandrus[5,6,7], David Cobden[2], Wang Yao[3], Xiaodong Xu[1,2,*]

Affiliations:
[1]Department of Materials Science and Engineering, University of Washington, Seattle, Washington 98195, USA
[2]Department of Physics, University of Washington, Seattle, Washington 98195, USA
[3]Department of Physics and Center of Theoretical and Computational Physics, University of Hong Kong, Hong Kong, China
[4]Advanced Materials Laboratory, National Institute for Materials Science, Tsukuba, Ibaraki 305-0044, Japan
[5]Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee, 37831, USA
[6]Department of Materials Science and Engineering, University of Tennessee, Knoxville, Tennessee, 37996, USA
[7]Department of Physics and Astronomy, University of Tennessee, Knoxville, Tennessee, 37996, USA

*Correspondence to: xuxd@uw.edu

Supplemental Discussion S1. Description of voltages and device function

The reason for the smaller voltage on V_{BG1} is because of the interplay with V_{SD}, which is applied to the source contact electrode on WSe_2, whereas the drain electrode on $MoSe_2$ is grounded. As V_{SD} is ramped up and drives source-drain current, the voltage drop across the boron nitride (BN) dielectric increases and thus the WSe_2 hole doping increases such that at $V_{SD} = 3.5$ V, the gating in WSe_2 is comparable to the gating in $MoSe_2$ when $V_{BG2} = 5$ V. If the gate voltages were equal and opposite, say -5 and +5 V, as V_{SD} is increased the BN dielectric would break down before forward bias conduction occurs. This is a limitation of these electrostatically defined lateral p-n junctions. In the negative V_{SD} regime, we have swept V_{BG1} to -8 V in order to ensure comparable p and n doping on respective sides of the device and no reverse bias conduction is observed, confirming we have a diode.

Supplemental Figure S1. Heterostructure twist angle and interlayer exciton light cones.

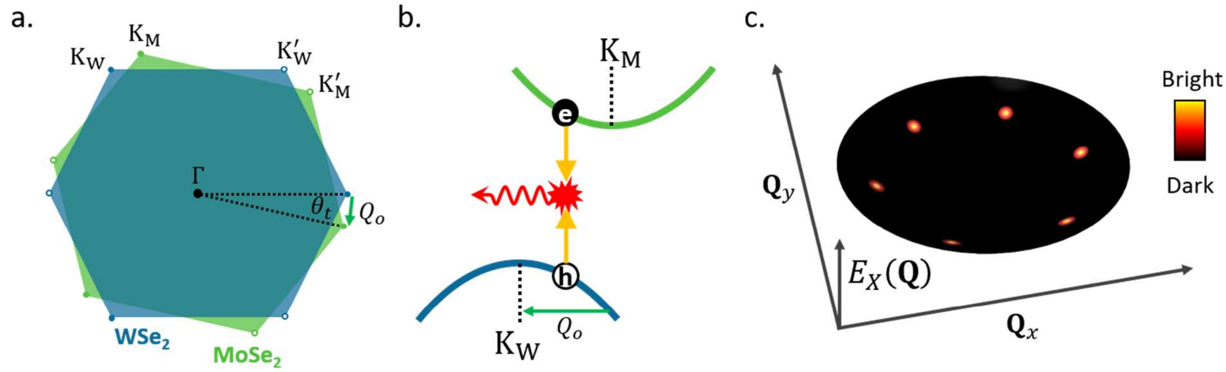

Figure S1 | Heterostructure twist angle and interlayer exciton light cones. a. A small twist angle (θ_t) results in the K valleys of constituent materials becoming momentum mismatched in the first Brillouin zone. $\mathbf{Q_o}$ is the relative displacement between $\mathbf{K}_\mathrm{W}$ and $\mathbf{K}_\mathrm{M}$. **b.** The displacement vector $\mathbf{Q_o}$ (green arrow) is the combined momentum of an electron and hole necessary to recombine for an interlayer transition[1]. This means the optically bright interlayer exciton has a center-of-mass velocity $\propto Q_o$. **c.** Due to the lattice three-fold rotation symmetry and the time-reversal symmetry between the two valleys, there are six light cones in the 2D exciton energy dispersion ($\mathrm{E_X}(\mathbf{Q})$) located at $\mathbf{Q_o}$ and its $\pi/3$ rotations. The lowest energy interlayer exciton at $\mathbf{Q} = 0$ is optically dark. *Figures recreated from reference [1].*

Supplemental Figure S2. Electroluminescence of additional devices

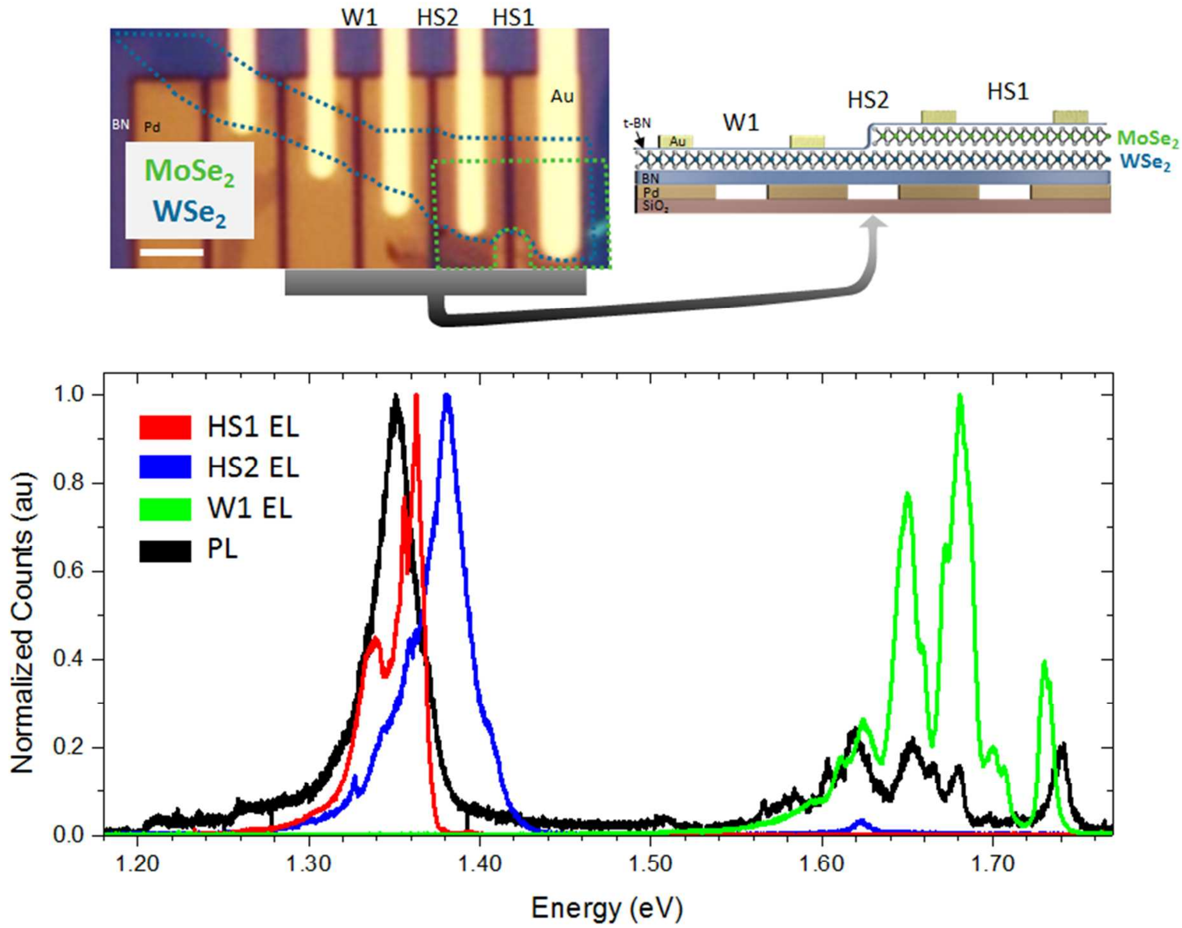

Figure S2 | Electroluminescence of additional devices. Electroluminescence (EL) from devices under similar voltages as discussed in the main text are compared above to the photoluminescence (PL) of undoped HS2 (black). In red is the full-HS device HS1, which has less blue shift to the PL than HS2 in blue. These relative interlayer emission energies can all be explained by a combination of the Stark effect and state filling, as discussed in the main text. In practice, HS1 was found to have less Stark shifts than HS2. In the main text Fig. 2c we see that at lower V_{SD} of about 2.5 V, HS2 EL can indeed match the PL at 1.36 eV. Finally, in green we have the emission from the single layer WSe_2 device W1 which shows EL only from intralayer exciton states between 1.6 and 1.74 eV[2].

Supplemental Figure S3. Global backgate dependent photoluminescence from HS2

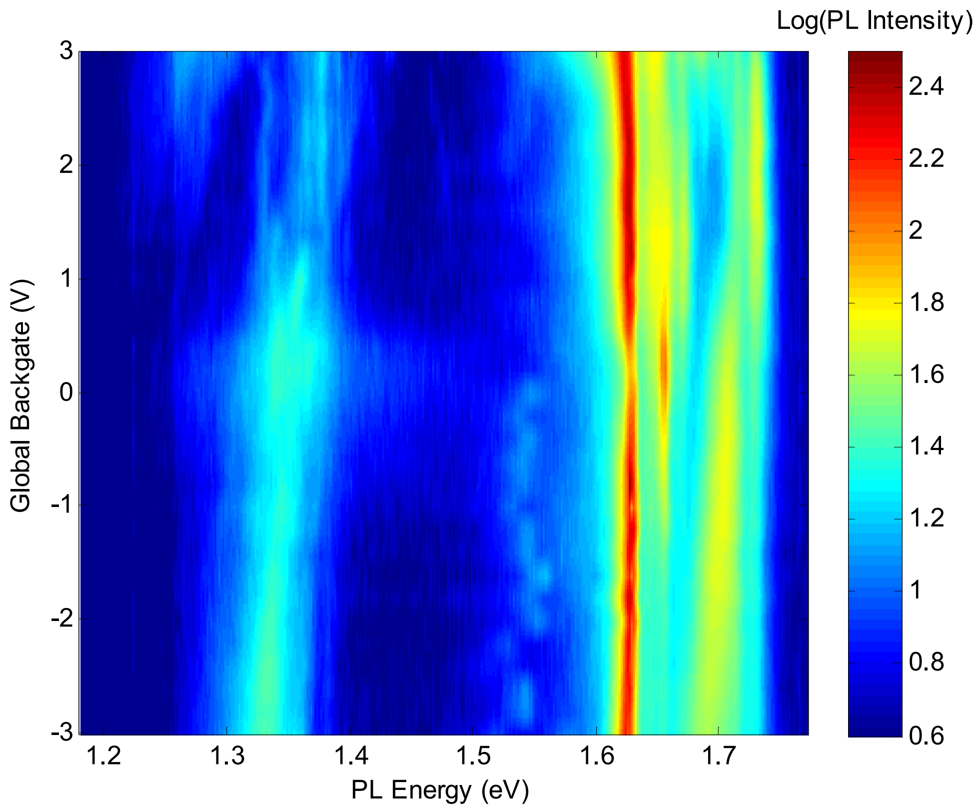

Figure S3 | Global backgate dependent photoluminescence from HS2. Photoluminescence (PL) from HS2 at a temperature of 30K under excitation with 2 μW laser power from a He-Ne light source (632 nm). All backgates are changed together while the top contacts remain grounded. The Stark tuning of the interlayer exciton shifts the low energy PL from ~1.33 eV to 1.38 eV. Importantly, the interlayer exciton PL is always below that of the peak in the wavelength dependent photocurrent, shown in Figs. 3b and 4b.

Supplemental Figure S4. Twist angle characterization

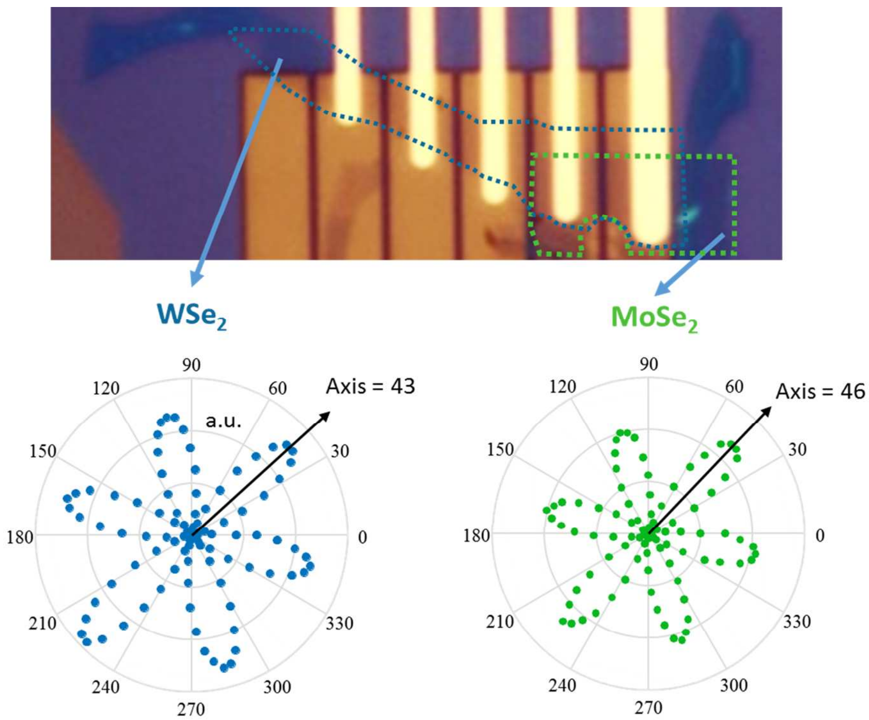

Figure S4 | Second harmonic generation (SHG) vs. linear polarization angle of incident light. Data taken from monolayer areas of constituent materials as indicated by the blue arrows. For WSe_2 ($MoSe_2$), incident light set to 1480 (1560) nm and SHG signal collected at 740 (780) nm. The maximum SHG signal co-polarized with the linearly-polarized excitation laser corresponds to the armchair axes of each crystal[3-5]. Looking at the difference in the armchair axes of each crystal, we estimate a 3-degree twist angle for the heterostructure region.

Supplementary References:

1. Yu, H., Wang, Y., Tong, Q., Xu, X. & Yao, W. Anomalous Light Cones and Valley Optical Selection Rules of Interlayer Excitons in Twisted Heterobilayers. *Physical Review Letters* **115,** 187002 (2015).
2. Ross, J. S. *et al.* Electrically tunable excitonic light-emitting diodes based on monolayer WSe_2 p-n junctions. *Nature Nanotechnology* **9,** 268–72 (2014).
3. Kumar, N. *et al.* Second harmonic microscopy of monolayer MoS_2. *Physical Review B* **87** (2013).
4. Li, Y. *et al.* Probing symmetry properties of few-layer MoS_2 and h-BN by optical second-harmonic generation. *Nano Letters* **13**, 3329-3333 (2013).
5. Malard, L. M., Alencar, T. V., Barboza, A. P. M., Mak, K. F. & de Paula, A. M. Observation of intense second harmonic generation from MoS_2 atomic crystals. *Physical Review B* **87** (2013).