

## Gate-tunable Diode-like Current Rectification and Ambipolar Transport in Multilayer van der Waals ReSe<sub>2</sub>/WS<sub>2</sub> p-n Heterojunctions

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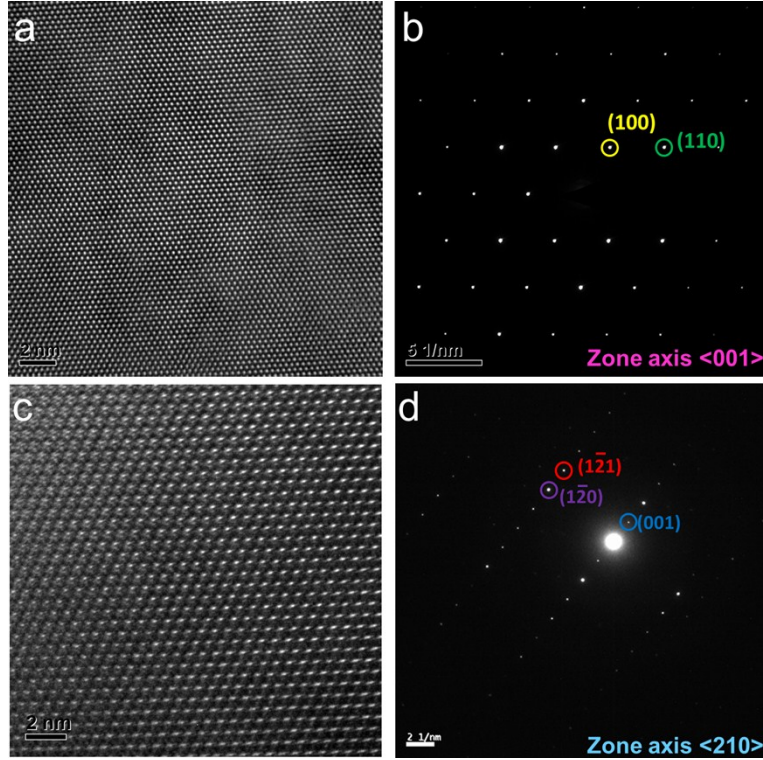


Figure S1 (a) TEM image of individual WS<sub>2</sub>; (b) SAED pattern of the WS<sub>2</sub>; (c) TEM image of individual ReSe<sub>2</sub>; (d) SAED pattern of the ReSe<sub>2</sub>.

The multi-layer WS<sub>2</sub> and the ReSe<sub>2</sub> 2D materials are obtained by micromechanical exfoliation and were transferred onto TEM grids with lacey support by poly (methyl methacrylate) (PMMA) transfer method in order to investigate their structure and crystal quality. The high resolution TEM (HRTEM) images (Figure S1a, c) show the hexagonal ring lattice in the WS<sub>2</sub> and the ReSe<sub>2</sub>, indicating good single crystalline phase. The corresponding selected area electron diffraction (SAED) patterns of the single crystalline WS<sub>2</sub> and ReSe<sub>2</sub> nanosheet (Figure S1b,d) reveal orientation of WS<sub>2</sub> is along (001) zone axis belongs to a hexagonal crystal system and orientation of ReSe<sub>2</sub> is along (210) zone axis.

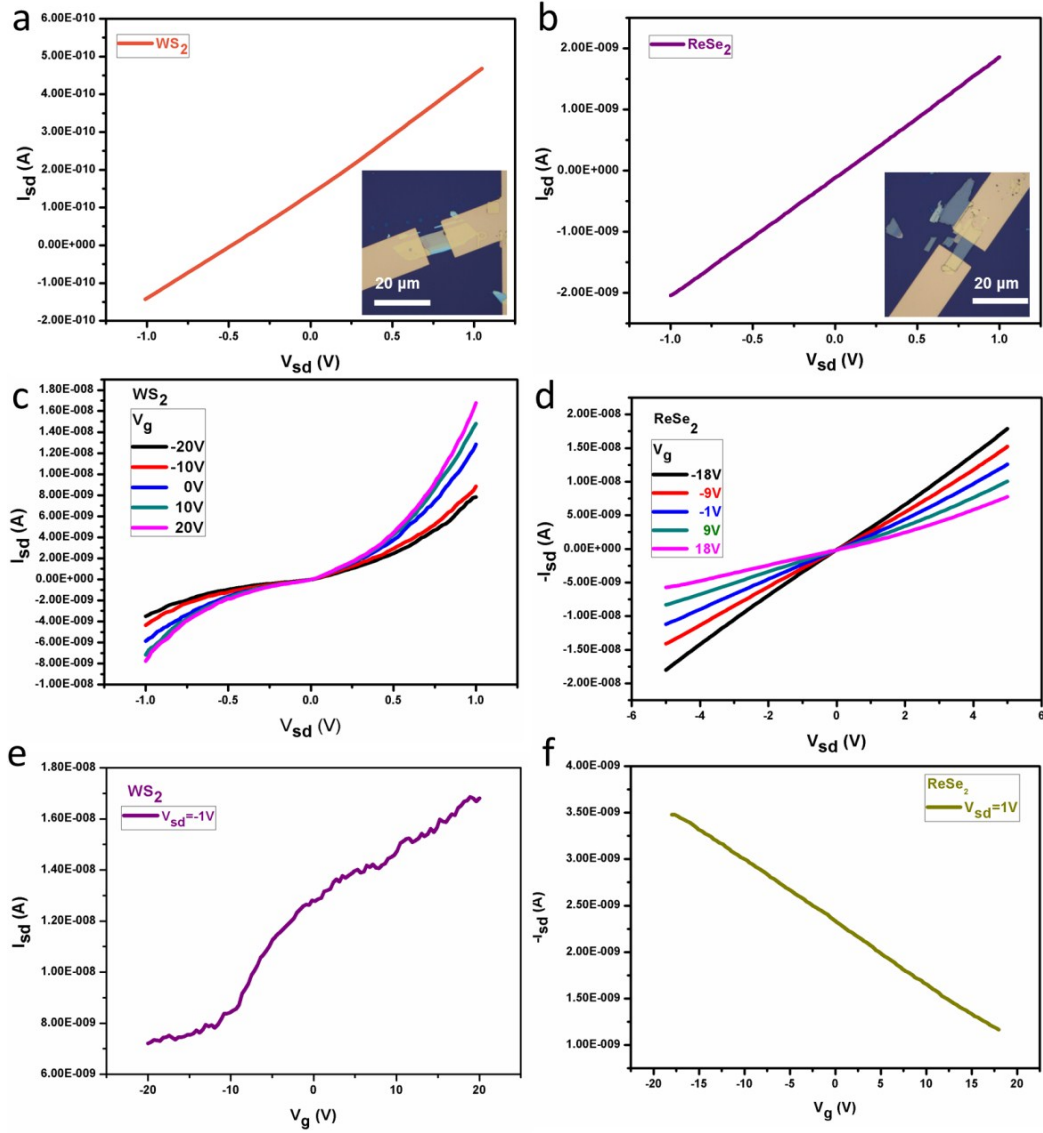


Figure S2  $I_{sd}$ - $V_{sd}$  characteristics of the individual (a) WS<sub>2</sub> multilayer and (b) ReSe<sub>2</sub> multilayer. Output characteristics of the individual (c)WS<sub>2</sub> multilayer and (d) ReSe<sub>2</sub> multilayer. Transfer characteristic of individual (e) WS<sub>2</sub> multilayer and (f) ReSe<sub>2</sub> multilayer.

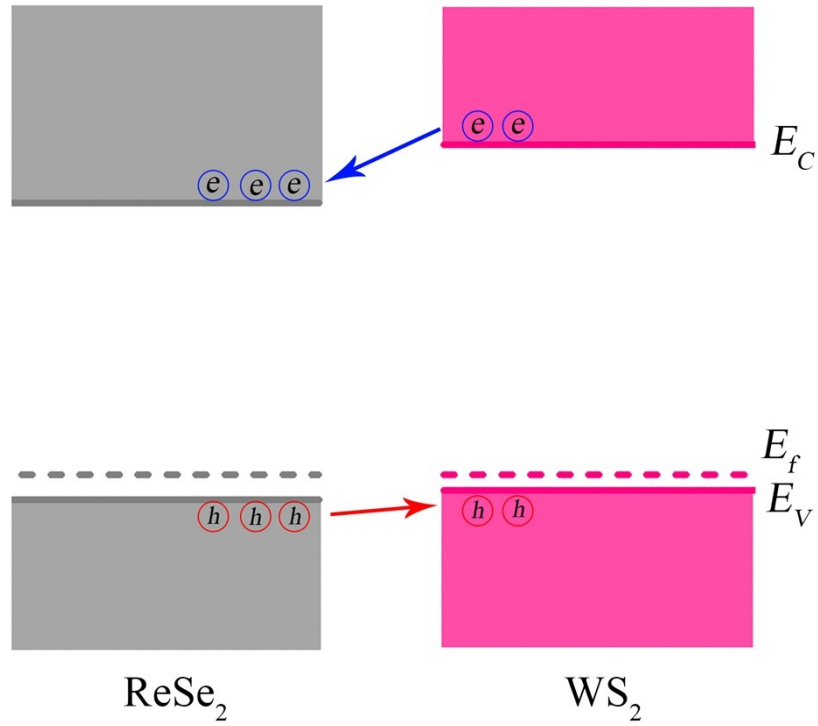


Figure S3. The band offset of type-II ReSe<sub>2</sub>-WS<sub>2</sub> heterostructure in the Fermi level alignment.

## Section A. Details of the ReSe<sub>2</sub>/WS<sub>2</sub> heterojunctions Device Fabrication.

The ReSe<sub>2</sub>/WS<sub>2</sub> heterojunctions were fabricated with transfer method. Both multilayer 2D ReSe<sub>2</sub> and WS<sub>2</sub> materials were first exfoliated mechanically and transferred onto two Si substrates coating 300nm-thick SiO<sub>2</sub> layer, respectively, from the WS<sub>2</sub> and ReSe<sub>2</sub> crystals. And then, a few drops of the self-regulating polymethyl methacrylate (PMMA) solution was spin coated onto ReSe<sub>2</sub> multilayer on Si substrates coating 300nm-thick SiO<sub>2</sub> layer by using rotating speed of 3000 r s<sup>-1</sup> and baked at 180 °C for 2 min as a transfer medium, and the ReSe<sub>2</sub> multilayer were transferred onto another substrate with WS<sub>2</sub> multilayer to partly overlap with the WS<sub>2</sub> multilayer to form ReSe<sub>2</sub>/WS<sub>2</sub> heterojunction sample, and finally the PMMA on ReSe<sub>2</sub> was removed by acetone.

Electrode patterns of the sample were fabricated by electron-beam

lithography (SEM) using an SEM/focused ion beam system (Nava 200 NanoLab) at a voltage of 30 kV and a current of 1.5 nA. And then 8-nm-thick Cr layer followed 80-nm-thick Au layer was deposited by using electron beam evaporation in turn. Finally, well-defined source and drain electrodes were achieved by using the lift-off process with acetone/IPA.

The Raman measurements were conducted by a confocal laser Raman spectrometer (Renishaw Model inVia-Reflex) with He-Ne laser (532 nm) excitation. Samples were further characterized by AFM (Benyuan NanoInstruments Ltd. Model CSPM 5500A) to confirm the thickness of layers.