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Electronic Supplementary Information

In-plane Ferroelectricity in Few-Layered GeS and its van der Waals Ferroelectric Diodes

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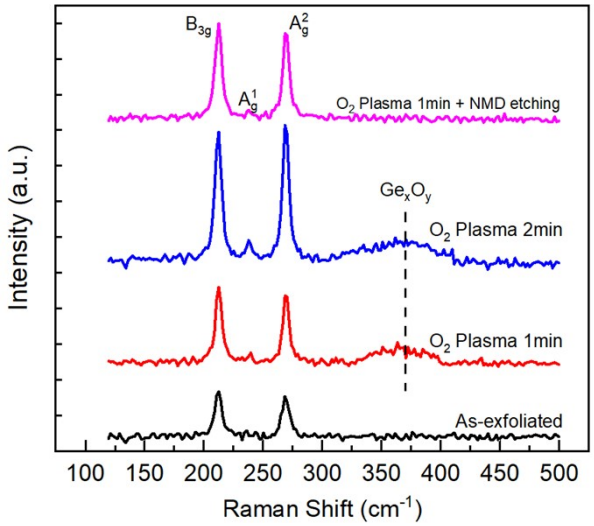

Figure S1 Raman studies of GeS flakes fabricated by exfoliation, O_2 plasma oxidation and NMD etching.

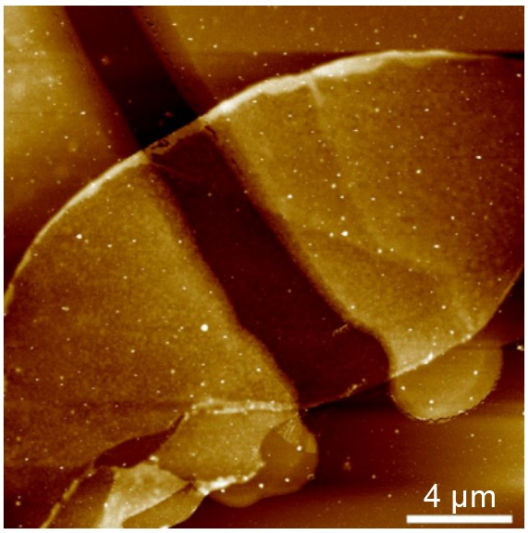

Figure S2 AFM image of the few-layer GeS nanoflake.

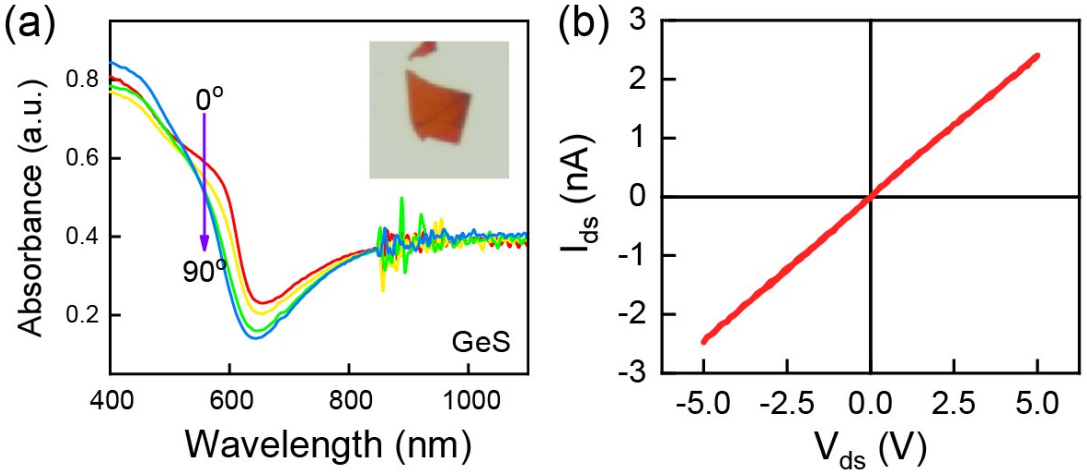

Figure S3 (a) Polarization-resolved absorption spectra with spectral range 400–1100 nm. (b) I-V plot of the thicker GeS sample.

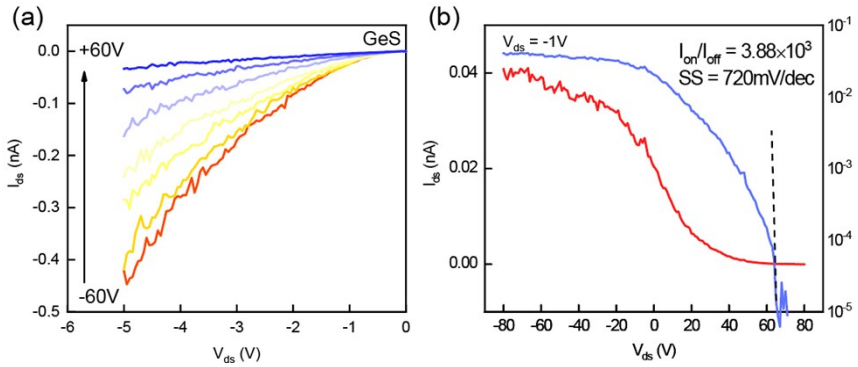

Figure S4 Typical output (a) and transfer (b) curves along the armchair direction.

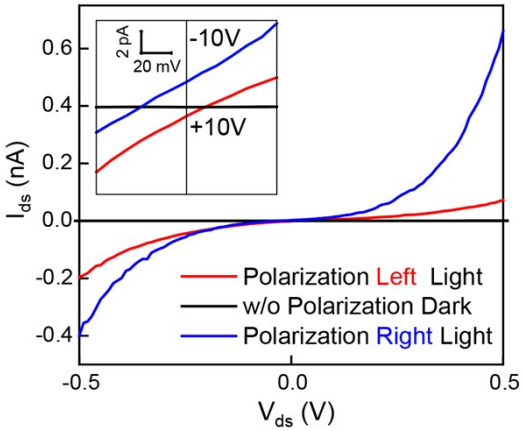

Figure S5 Light and dark I–V measurements before and after polarization, respectively.

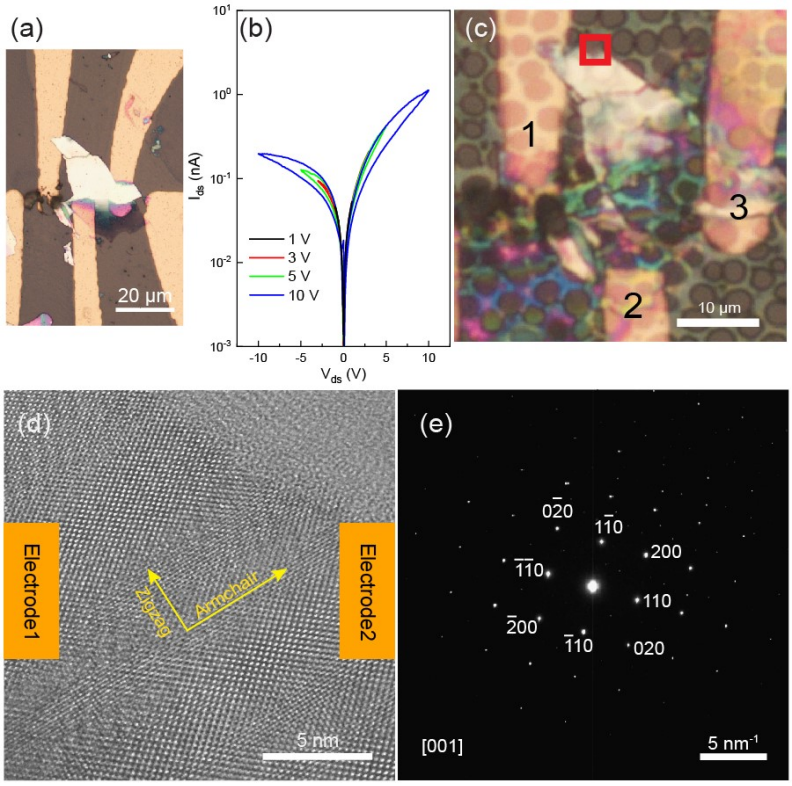

Figure S6 (a) Optical microscope image of the GeS device with three probes on SiO_2/Si substrate. (b) I-V curves of the GeS device in (a). (c) Optical microscope image of the GeS device transferred onto TEM copper grids. The electrode 2 was detached during the transfer process (d) The

magnified ADF images taken from the areas marked by red square in (c). Scale bar 5 nm. (e) SAED pattern taken from (d).

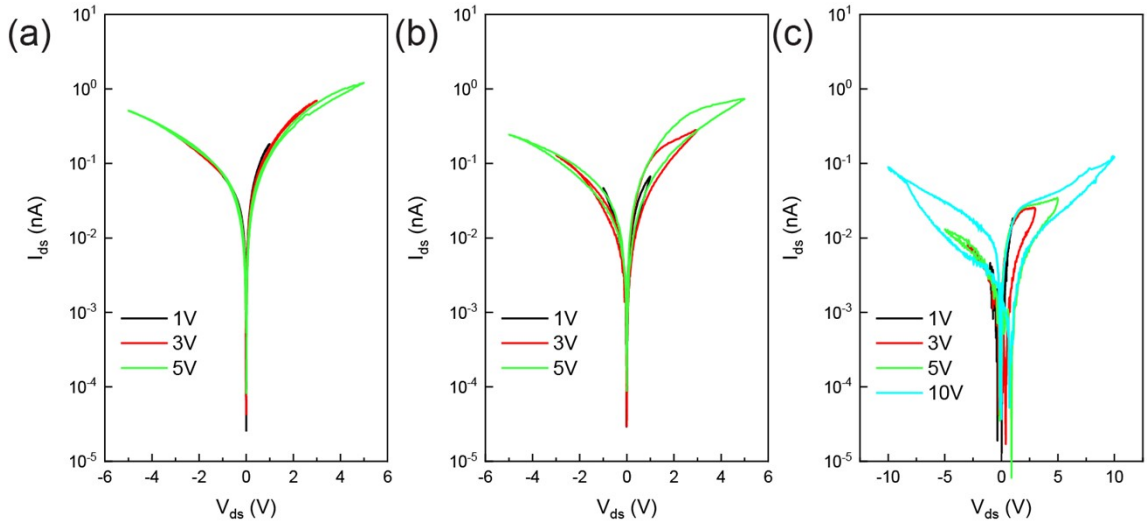

Figure S7 The I-V hysteresis loops for GeS devices with different thickness: (a) 88.94 nm, (b) 43.75 nm and (c) 5.25 nm.

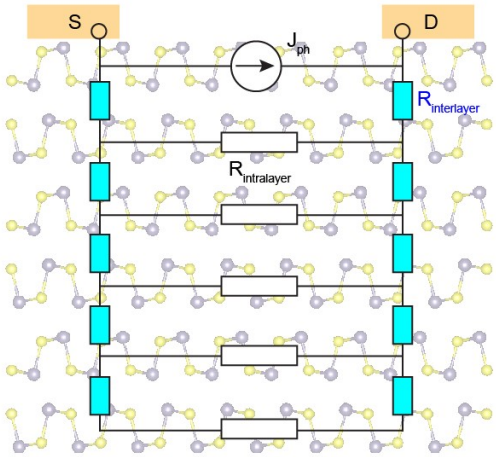

Figure S8 Resistance network model for the GeS BPV device.

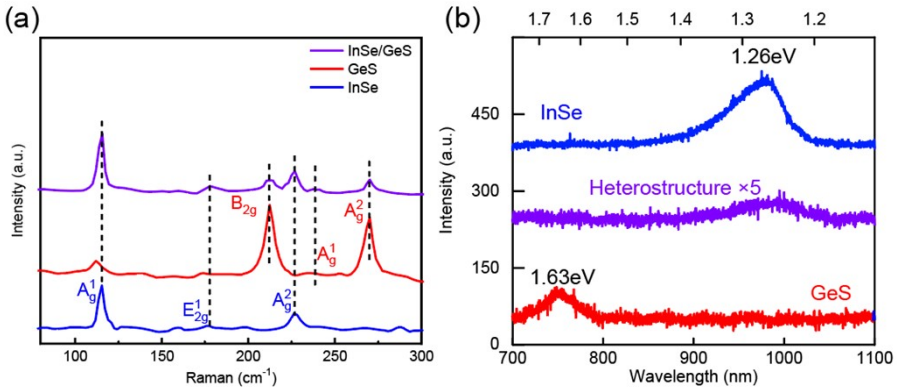

Figure S9 (a) Raman spectra of InSe, GeS and the heterojunction of InSe and GeS. (b) PL spectra of GeS, InSe and the heterojunction of InSe and GeS.

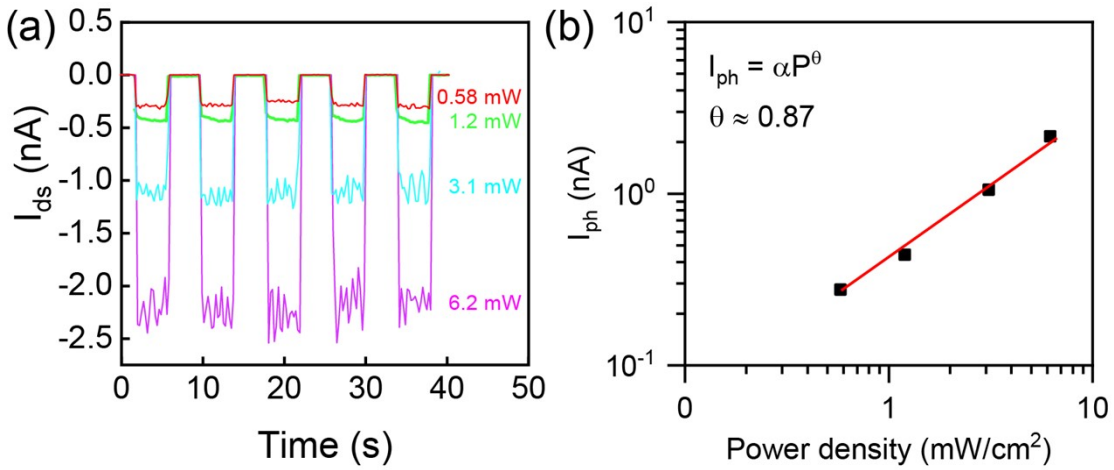

Figure S10 (a) Photocurrent trace of the device illuminated by a chopped light with a frequency of 0.25 Hz at V_{ds} = 0 V and V_{gs} = 0 V. (b) Gate-voltage (V_{gs}) dependent open-circuit voltage (V_{oc}) in the Fe-PV heterojunction under 670 nm light.

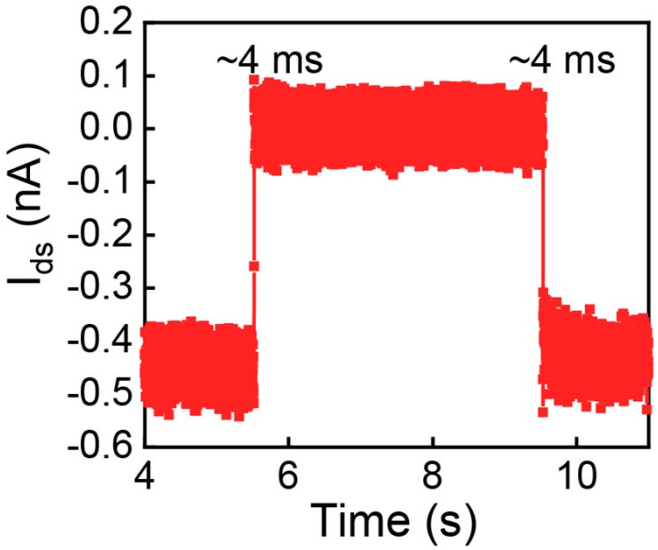

Figure S11 Response time (rise and decay time) of the Fe-PV heterojunction.