## **Near-Infrared Optical Transitions in PdSe<sub>2</sub> Phototransistors**

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## **S1. Device Fabrication**

Degenerately p-doped silicon substrate with 270 nm of thermally grown SiO<sub>2</sub> was first treated by oxygen plasma for 10 min to enhance hydrophilicity. Few-layer PdSe<sub>2</sub> flakes were produced by mechanical exfoliation from bulk grown crystals onto the substrates. Metal electrodes were then defined by electron beam lithography and the subsequent electron beam deposition of 5 nm of Ti covered by 45 nm of Au. Optical microscopy and Park-Systems XE-70 noncontact mode AFM were used to identify and characterize thin PdSe<sub>2</sub> flakes.

S2. Schottky Barrier Height of PdSe2 with Ti/Au Contacts



**Figure S1. Schottky barrier extraction.** (a) The Arrhenius plots for several back-gate bias voltages measured with drain voltage bias at 100 mV. The slope of each curve yields the effective Schottky barrier height at the corresponding gate bias. (b) The extracted effective Schottky barrier height at the various back-gate bias voltage, where the flatband electron Schottky barrier height is measured to be 180 meV.

## **S3. Scanning Photocurrent Measurement**

Spatially-resolved scanning photocurrent measurements were executed using an Olympus microscope setup (BX51W). A linearly-polarized continuous wave laser beam (NKT Photonics SuperK Supercontinuum Laser) was expanded and focused by a 40× objective (N.A. = 0.6) into a diffraction - limited spot ( $\approx 1 \mu m$ ) on the devices. The position of the laser beam was changed by a nanometer-resolution scanning mirror. All experiments were performed in a Janis ST - 500 microscopy cryostat in a near zero-bias and high vacuum ( $\approx 10^{-6}$  Torr) environment.

## **S4.** Photocurrent Generation Mechanism



**Figure S2**. (a) Conductance of a  $PdSe_2$  device as a function of temperature; (b) photocurrent response of the device as a function of drain-source bias (red dots represent experimental data and black line is a linear fitting); and (c) a line profile of the photocurrent response across the metal-PdSe<sub>2</sub> junction is shown by the black dots. The red solid curve is the related Gaussian fitting. The green arrow denotes the photocurrent "tail" on the electrode. The orange background indicates the position of the electrode.