Supporting Information

High Performance van der Waals Junction Field Effect Transistors Utilizing Organic Molecule/Transition Metal Dichalcogenide Interface

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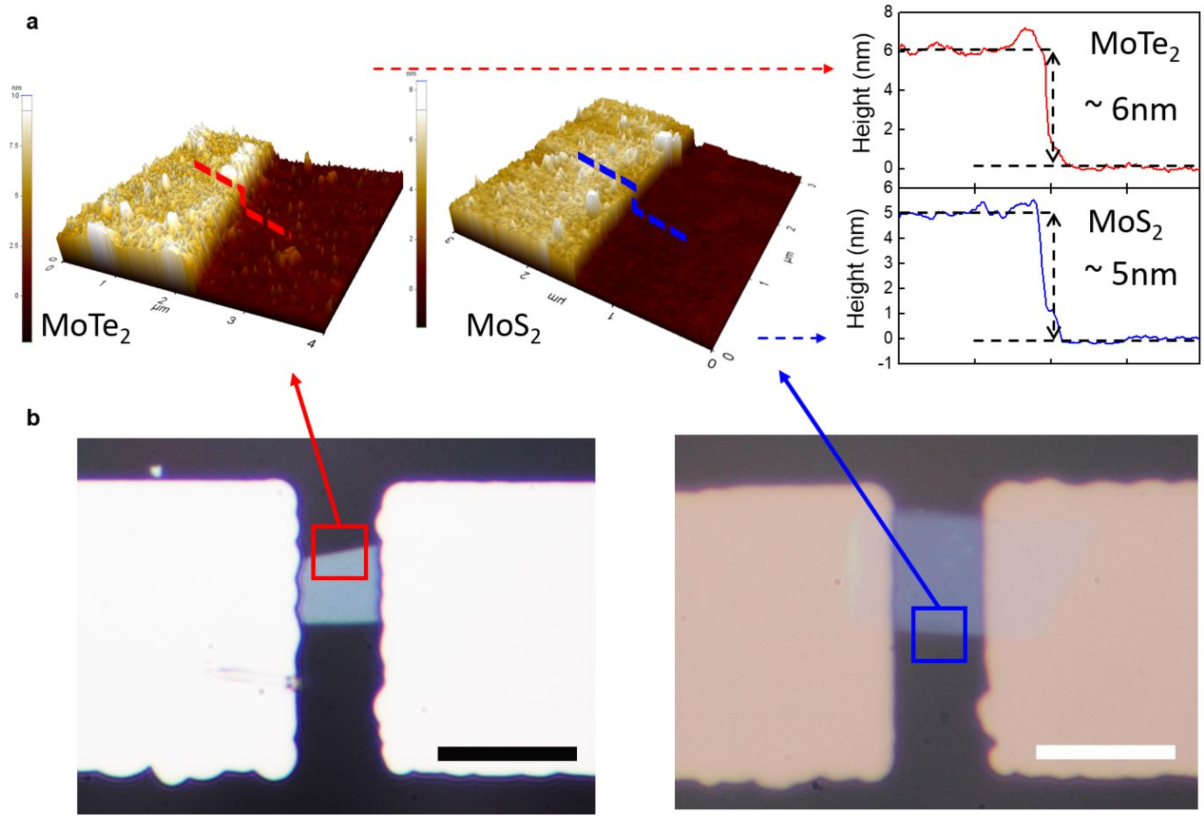

Figure S1. AFM measurement. (a) 3D AFM images and Thickness profiles for MoTe$_2$ and MoS$_2$ flakes. Thickness appears to be ~6 and ~5 nm for MoTe$_2$ and MoS$_2$ flakes, respectively. (b) Corresponding optical microscope images of *p*- and *n*- JFET before patterning the gate. (scale bar, 10 μm)

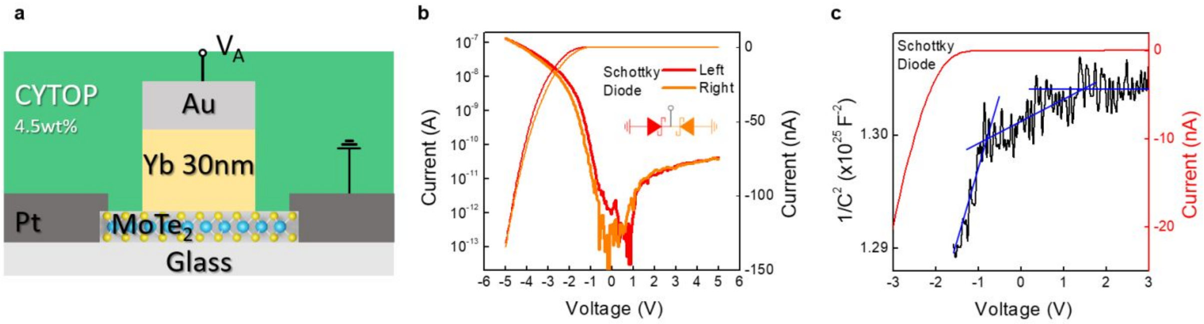

Figure S2. I-V & C-V measurement of Yb/*p*-$MoTe_2$ Schottky diode. (a) Schematic cross section view of our Yb/$MoTe_2$ Schottky junction MESFET. (b) I-V curves of the Yb/$MoTe_2$ Schottky diode that was presented in Figure 1f of main manuscript. In the logarithmic scale I-V plots, turn-on voltage seems to be -1 ~ -2 V. (c) C-V measurement of Yb/*p*-$MoTe_2$ Schottky diode was conducted and plotted along with I-V curve. According to the I-V and C-V curves, *p*-type only region starts from -1 V. On the one hand, carrier concentration of holes in $MoTe_2$ is again extracted out, to be ~ 7×10^{16} cm^{-3}, using the slope at 500 kHz for C-V measurements. So the hole density is only slightly different from the case at 400 kHz, which is but regarded ignorable.

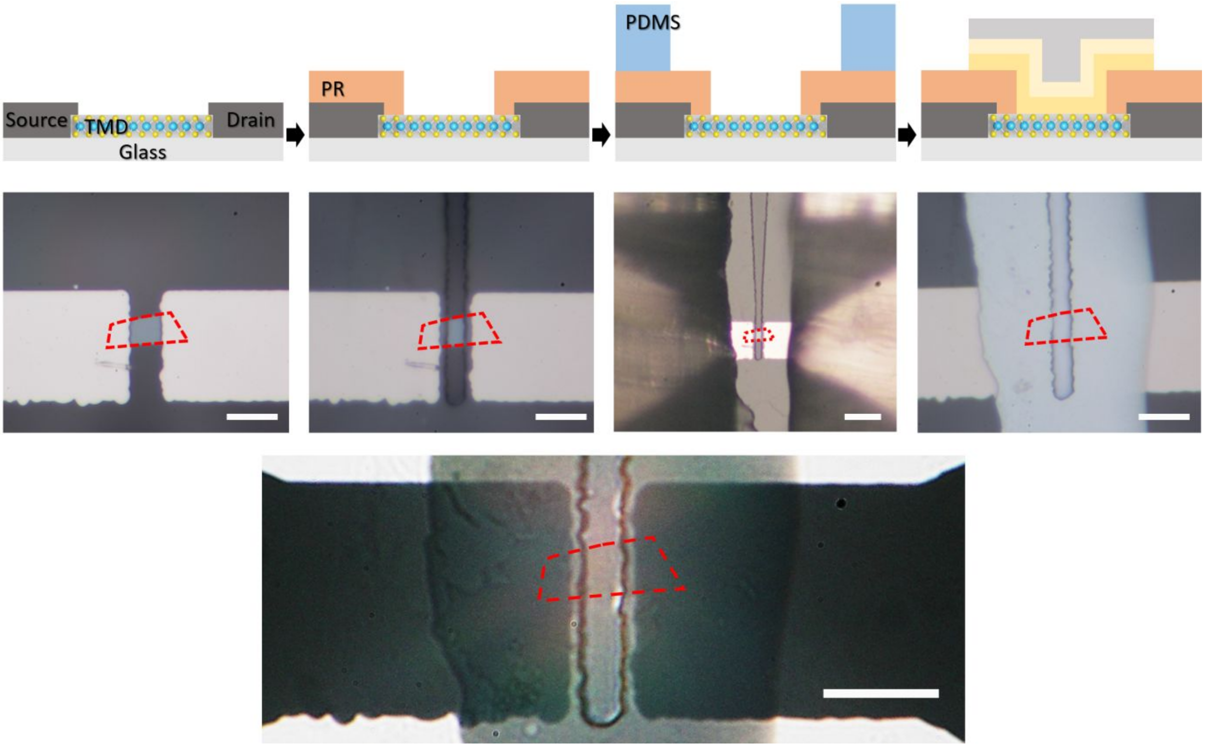

Figure S3. Fabrication process of organic/TMD JFET. 1. The initially transferred TMD is patterned with S/D electrode on glass by conventional photolithography and lift-off. 2. Photoresist (PR) is patterned on the device as shown above, to protect ungated $MoTe_2$ region from any gate metal, and in fact the PR has dipoles inside causing $MoTe_2$ channel to be more strong *p*-type in polarity. 3. Then, we used a thick PDMS to pattern the gate area looking through an optical microscope. 4. Thermal evaporation deposition of organic and gate metal was conducted sequentially. 5. Finally, PDMS has been removed to show a wanted device architecture. Final OM image was obtained by bottom illumination, clearly displaying a whole JFET area. Flake is too thin (5~6 nm) to clearly distinguish its contour. The size of inset scale bar is 10, 10, 20, 10 and 10 μm respectively in order from the left OM image.

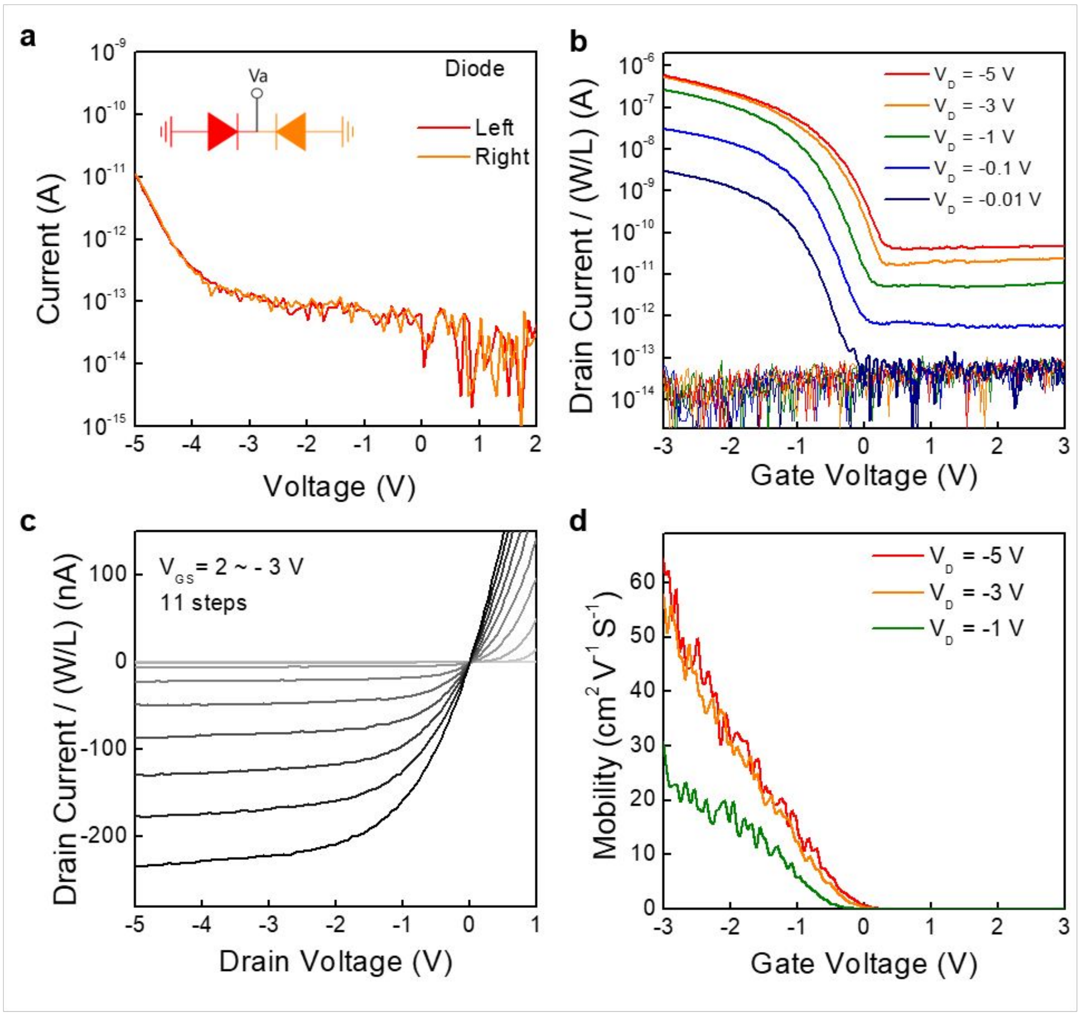

Figure S4. van der Waals *n*-organic/*p*-channel $MoTe_2$ junction FET with LiF/Alq3 gate as fabricated for reproducibility test. (a) I-V curves of Alq_3/*p*-$MoTe_2$ PN diode. (b) I_D-V_{GS} transfer, I_G-V_{GS} gate leakage, (c) I_D-V_{DS} output curves and (d) mobility plot of another *p*-JFET with same structure as the JFET in Figure 2 of main manuscript. Highest mobility and I_{ON} value appear to be ~60 cm^2/V s and 240 nA, respectively which are somewhat lower than those of the other JFET presented in Figure 2. It is probably because the $MoTe_2$ thickness might be here thinner than the other case, however, 60 cm^2/V s is still regarded quite high.

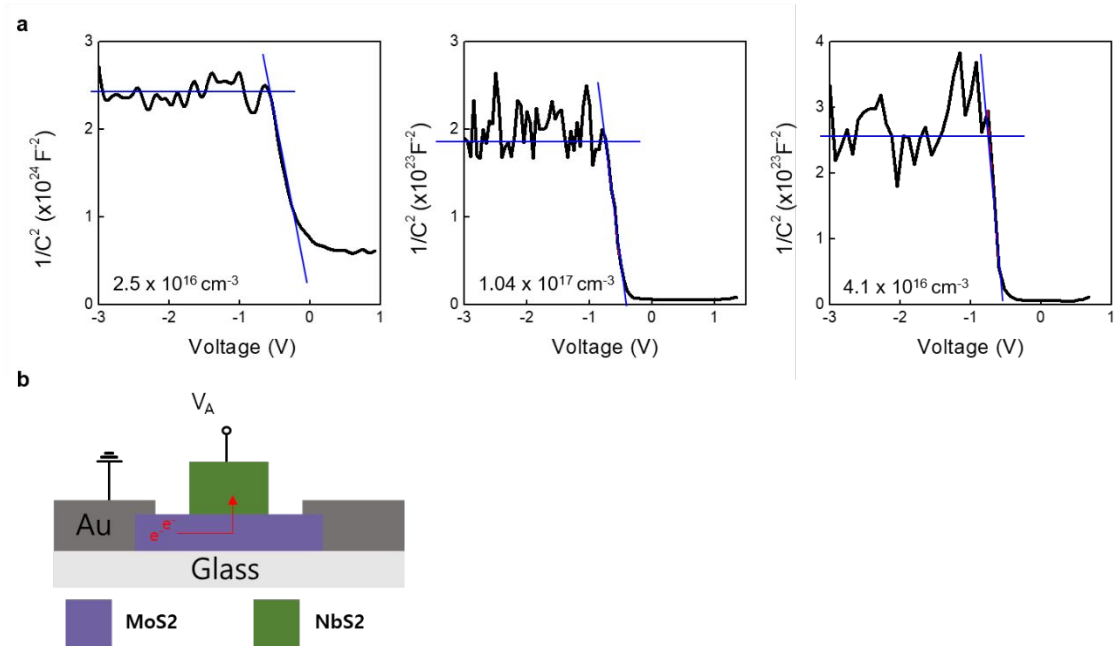

Figure S5. C-V measurement of NbS_2/*n*-MoS_2 Schottky diodes for estimating electron concentration. (a) C-V measurement results from NbS_2/*n*-MoS_2 Schottky Diode. (b) Schematic cross section of *n*-channel MESFET with two Schottky diodes.

In order to estimate the carrier density of *n*-MoS_2, we fabricated a NbS_2/*n*-MoS_2 MESFET, which contains two Schottky diodes in one MESFET device, although such experimentations are already found in our previous report.[1] Schematically shown above is the device. After C-V measurement, the C-V data was converted to $1/C^2$-V plot. The slope of $1/C^2$-V plot is $-2/(q\varepsilon_s N_d)$ according to the charge equation involving the bias-dependent depletion thickness (X_d). Here, q is electronic charge, ε_s is dielectric constant of MoS_2 $=12\varepsilon_o$, N_d is electron concentration # /cm^3. As we work out electron carrier concentration, N_d of 5 nm-thin MoS_2 channel, from $1/C^2$-V measurement on the diode. Schottky diode would show an abrupt capacitance change right at the moment when the MoS_2 layer thickness under NbS_2 gets to the total depletion state by a certain gate voltage, as shown in the figure. As a result, the slope of $1/C^2$-V plot is extracted out to estimate N_d value, which turns out to be 2.5×10^{16}, 4.1×10^{16}, and 1.04×10^{17} cm^{-3} as obtained from three NbS_2/*n*-MoS_2 Schottky diodes. We thus averaged the three values, and determine N_d as 5.67×10^{16} cm^{-3}.

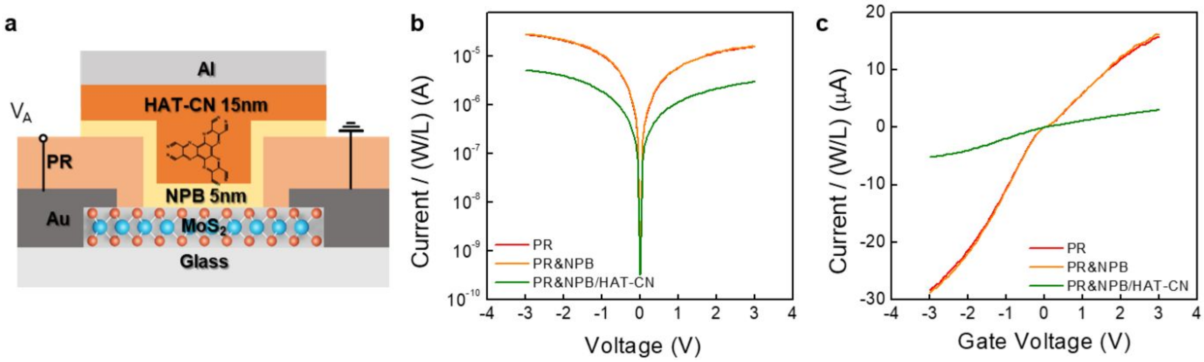

Figure S6. Two terminal I-V measurements for MoS_2 JFET system. (a) Schematic cross section view of our HAT-CN/NPB/MoS_2 JFET. (b) Logarithmic and (c) Linear scale two terminal current-voltage (I-V) measurements of MoS_2 channel in Al/HAT-CN/NPB/MoS_2 JFET. With NPB deposition only, the current is almost the same as that without NPB. With HAT-CN/NPB deposition, the current decreases by an order of magnitude due to HAT-CN/NPB-induced channel depletion. So these results well support our band diagrams in the main manuscript.

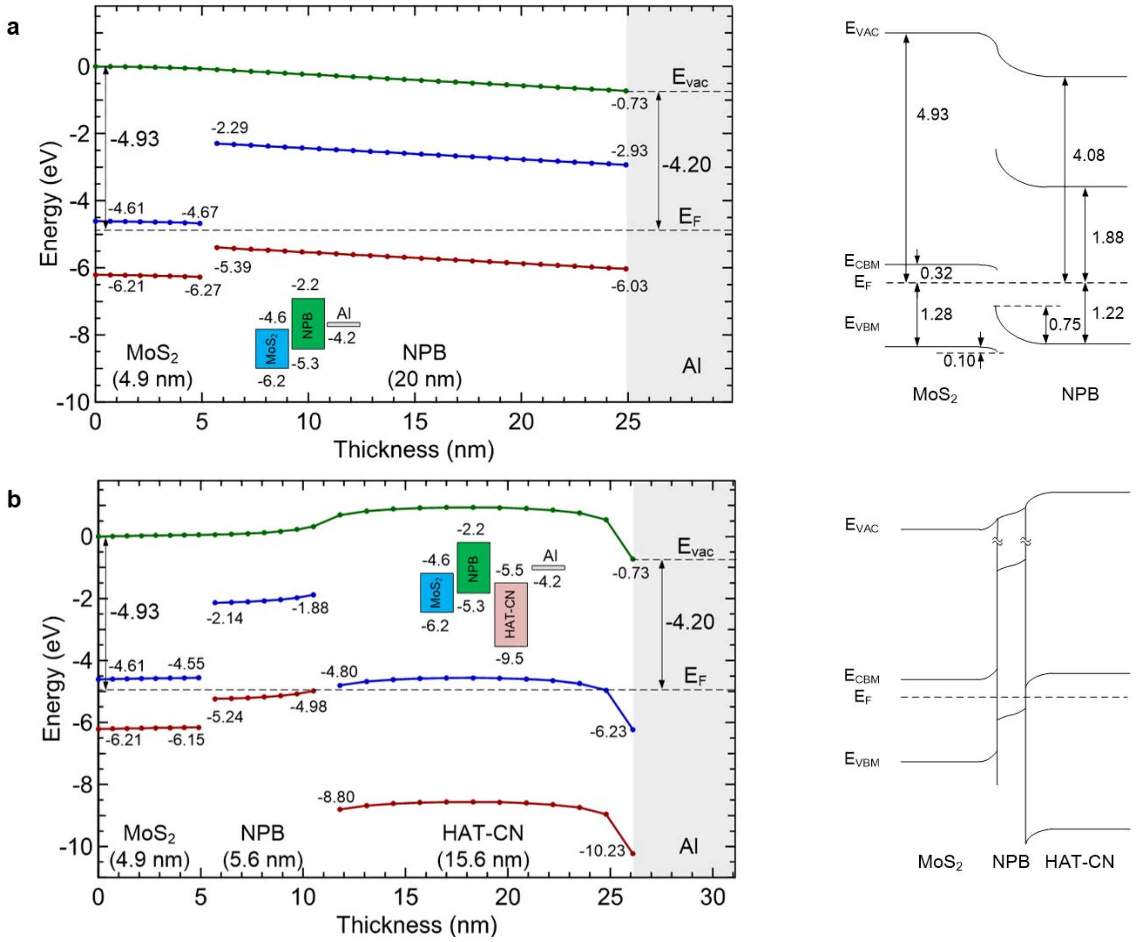

Figure S7. Band diagram simulation for Al/NPB/MoS_2 and Al/HAT-CN/NPB/MoS_2 systems.

(a) Al/NPB/MoS_2 band diagram estimated by simulation that contains Poisson's equations. Results show the charge accumulation (bulk conduction edge E_C *vs*. junction E_C = -4.61: -4.67 eV) at the MoS_2 junction with NPB, which well matches to the expected band diagram in the main manuscript (right). (b) Al/HAT-CN/NPB/MoS_2 band diagram estimated by simulation that contains Poisson's equations. Results show the charge depletion (bulk conduction edge E_C *vs*. junction E_C = -4.61: -4.55 eV) at the MoS_2 junction with NPB due to HAT-CN, which well matches to the expected band diagram in the main manuscript (right).

This numerical electrostatic simulation[2] contains a self-consistent procedure to figure out the actual charge distribution inside the system domain and corresponding electric potential which brings a band bending. For all cases, we assume MoS_2 thickness as ~4.9 nm (7L).

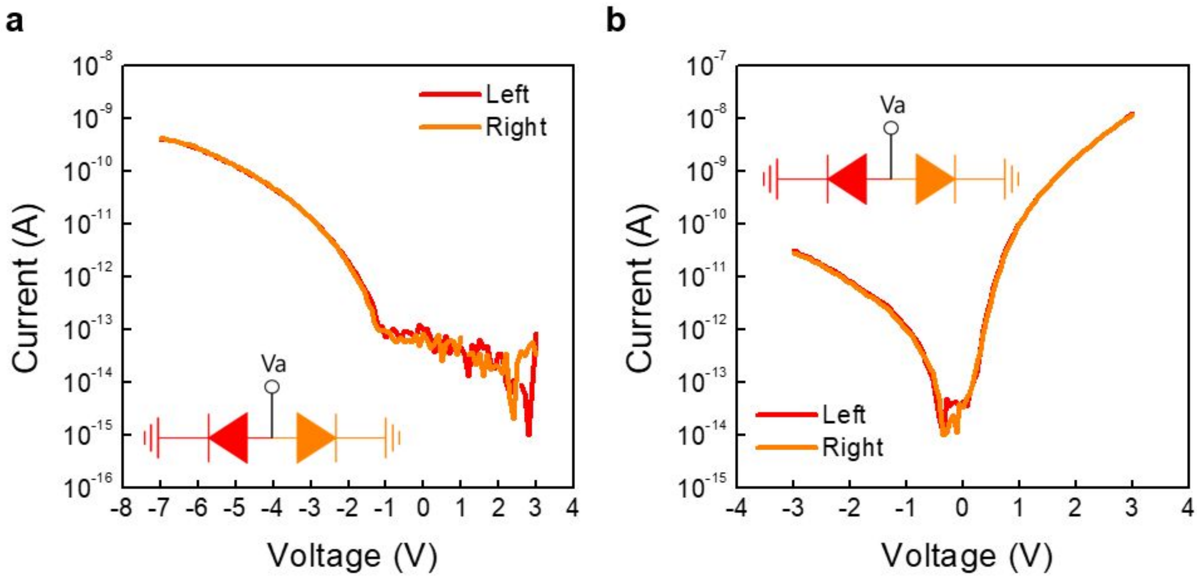

Figure S8. I-V characteristics of Al/*p*-NPB/*n*-MoS_2 and Al/HAT-CN/*p*-NPB/*n*-MoS_2 diodes. (a) I-V characteristics of Al/*p*-NPB/*n*-MoS_2 and (b) Al/HAT-CN/*p*-NPB/*n*-MoS_2 diodes were measured. Here, Al side is biased with respect to *n*-MoS_2 (ground). Basically the current level is low in both cases, but behaves different each other. It is because the second case (b) has thin NPB (~ 5 nm) below thick HAT-CN. NPB is thin enough to allow electron tunneling from *n*-type HAT-CN, causing current increase at (+) bias voltages. (See Figure S7 band diagrams). These vertical PN junction I-V behavior must be similar to those of I_G leakage in each case, which is understandable.

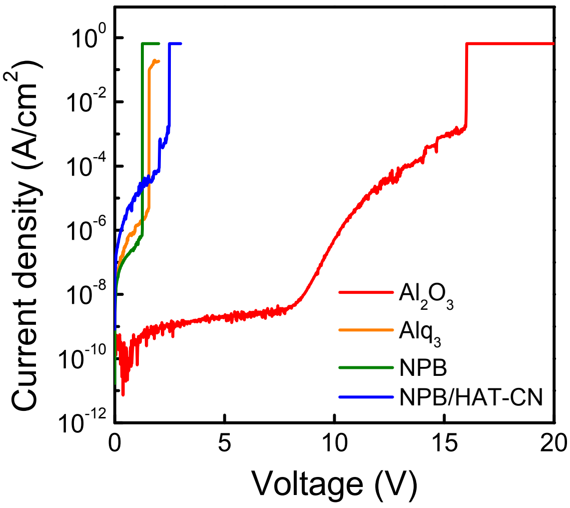

Figure S9. I-V characteristics of organic semiconductor gate layers and reference Al_2O_3 insulator: Au/ 20 nm-thick NPB (or Alq3, NPB/HAT-CN) /Al (metal/organic/metal) diodes and Au/20 nm-thick Al_2O_3/Al diode.

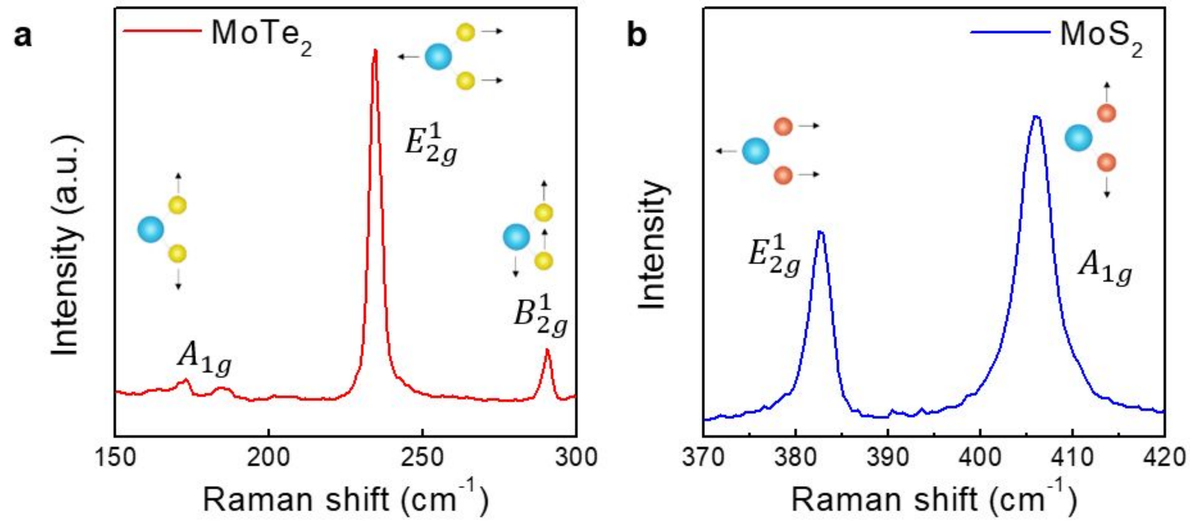

Figure S10. Raman results. Raman spectra obtained from (a) $MoTe_2$ and (b) MoS_2 flakes, which were exfoliated on glass. The spectra appear quite typical for 5-6 nm-thin $MoTe_2$ and MoS_2.

References

(1) Shin, H. G.; Yoon, H. S.; Kim, J. S.; Kim, M.; Lim, J. Y.; Yu, S.; Park, J. H.; Yi, Y.; Kim, T.; Jun, S. C.; Im, S. Vertical and in-Plane Current Devices Using NbS2/*n*-MoS2 van der Waals Schottky Junction and Graphene Contact. *Nano Lett.* **2018**, *18*, 1937–1945.

(2) Oehzelt, M.; Koch, N.; Heimel, G. Organic Semiconductor Density of States Controls the Energy Level Alignment at Electrode Interfaces. *Nat. Commun.* **2014,** *5*, 4174.