Supplementary Information

Two-step synthesis and characterizations of vertically stacked SnS-WS$_2$ and SnS-MoS$_2$ p-n heterojunctions

Adha Sukma Aji, Masanori Izumoto, Kenshiro Suenaga, Keisuke Yamamoto, Hiroshi Nakashima, and Hiroki Ago

$^a$Interdisciplinary Graduate School of Engineering Sciences, Kyushu University, Fukuoka 816-8580, Japan
$^b$Global Innovation Center (GIC), Kyushu University, Fukuoka 816-8580, Japan
Figure S1. Temperature profiles of the first step CVD: (a) WS$_2$ and (b) MoS$_2$. (c) Temperature profile of the second CVD step used for the SnS growth.
Figure S2. Optical microscope images of as-grown WS$_2$ (a) and MoS$_2$ (b) on sapphire substrates.
**Figure S3.** Optical microscope image of as-grown SnS-WS$_2$ at a low growth temperature (175 °C).
Figure S4. (a) Optical micrograph of a SnS-MoS$_2$ heterostructure. (b,c) AFM image and line profile of the SnS-MoS$_2$ heterostructure. (d,e) Raman intensity maps of $A_g$ peak of SnS and $E_{12g}$ peak of MoS$_2$. (f) PL intensity map of MoS$_2$. (g,h) Raman and PL spectra from the three points marked in (d)-(f). Red and green shaded areas indicate the Raman peaks from SnS and MoS$_2$, respectively. All scale bars are 10 µm.
Figure S5. a) Optical micrograph and (b) SEM image of the product obtained by the low-pressure CVD growth of SnS at ~400 Pa. The pre-existing WS$_2$ grains are fully covered with SnS so that only SnS grains are observed on the substrate surface. The SnS grains grew both in lateral and vertical directions. The thickness of the vertically-grown SnS is about 100 nm (b).
Figure S6. Raman spectrum of a SnS grain grown without H_2 gas flow during the CVD. The A_{1g} peak from SnS_2 was detected in addition to the multiple peaks originated in SnS.
Figure S7. (a) Overlap region of SnS on WS$_2$ or MoS$_2$. Distribution of the overlap region in (b) SnS-WS$_2$ and (c) SnS-MoS$_2$ heterostructures.

Overlap region (%) = $\frac{L_{\text{in}}}{L_{\text{in}} + L_{\text{out}}} \times 100\%$
Figure S8. Optical microscope image of a large monolayer WS$_2$ grain with a SnS grain on the top. It is seen that SnS grows at the edge of the small multilayer WS$_2$ grain.
Figure S9. (a) Optical micrograph of the SnS-MoS$_2$ heterostructure FET. Transfer (b) and output (c) curves of MoS$_2$ FET. (d) Output curves of the SnS-MoS$_2$ heterostructure. (e) Transfer curves of the majority (red) and minority (blue) currents of the SnS-MoS$_2$ heterostructure.
Figure S10. (a-d) Fabrication process of the WS\textsubscript{2} device with SnS on top and that without SnS. For comparison SnS device with WS\textsubscript{2} below was also fabricated. The device 1-2 measures the transport property of WS\textsubscript{2} channel with SnS on top. The device 3-4 shows the transport of WS\textsubscript{2} without SnS on top. The device 5-6 shows the transport of SnS with WS\textsubscript{2} underneath. (e) Transfer curves of the devices 1-2 and 3-4. (d) Transfer curve of the device 5-6.
Figure S11. Band diagrams of SnS and WS$_2$. Red and green lines indicate the expected Fermi levels of SnS and WS$_2$, respectively. The band diagrams are taken from refs. 40 and 50.