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Supplementary information for

Large-scale MoS_2 thin films with chemically formed holey structure for enhanced Seebeck thermopower and their anisotropic properties

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1. CVD-grown TMDC films transfer method (PMMA-assisted wet transfer)

In general, it is very difficult to transfer CVD-grown TMDC material such as MoS_2, WSe_2, and WS_2 onto bare SiO_2/Si substrate without etching the bottom SiO_x layer. Therefore, we used the conventional PMMA-assisted wet chemical etching method. First, a thin layer of PMMA (950 K A4) was spin-coated on the CVD-grown MoS_2 film, and the sample was placed on a hot plate at 120 °C for 2 min. The sample was then floated in 5% HF (aq) for 1 min, in which the Si substrate sank owing to etching of SiO_x, which left the PMMA/MoS_2 layers to float on top of the HF solution. Then, the PMMA-coated MoS_2 film was washed with de-ionized water, transferred to target substrates such as SiO_2/Si and Cu/SiO_2/Si layers depending on the in-plane and cross-plane Seebeck coefficient measurements, and dried in ambient conditions for 1 h. For good adhesion to the substrate, further baking was performed at 120 °C for 2 min. Finally, the PMMA layer was removed using warm acetone, followed by rinsing with isopropyl alcohol. The transferred sample was loaded into a furnace to anneal at 250 °C for 2 h to further remove polymer residue. For the Seebeck coefficient measurements of the samples, additional Cu metal layers were deposited on the transferred MoS_2/SiO_2/Si and MoS_2/Cu/SiO_2/Si substrates by radio-frequency sputtering at room temperature. The same method was used for WSe_2 and WS_2 films.

2. AFM measurement of WSe_2 and WS_2 thin films

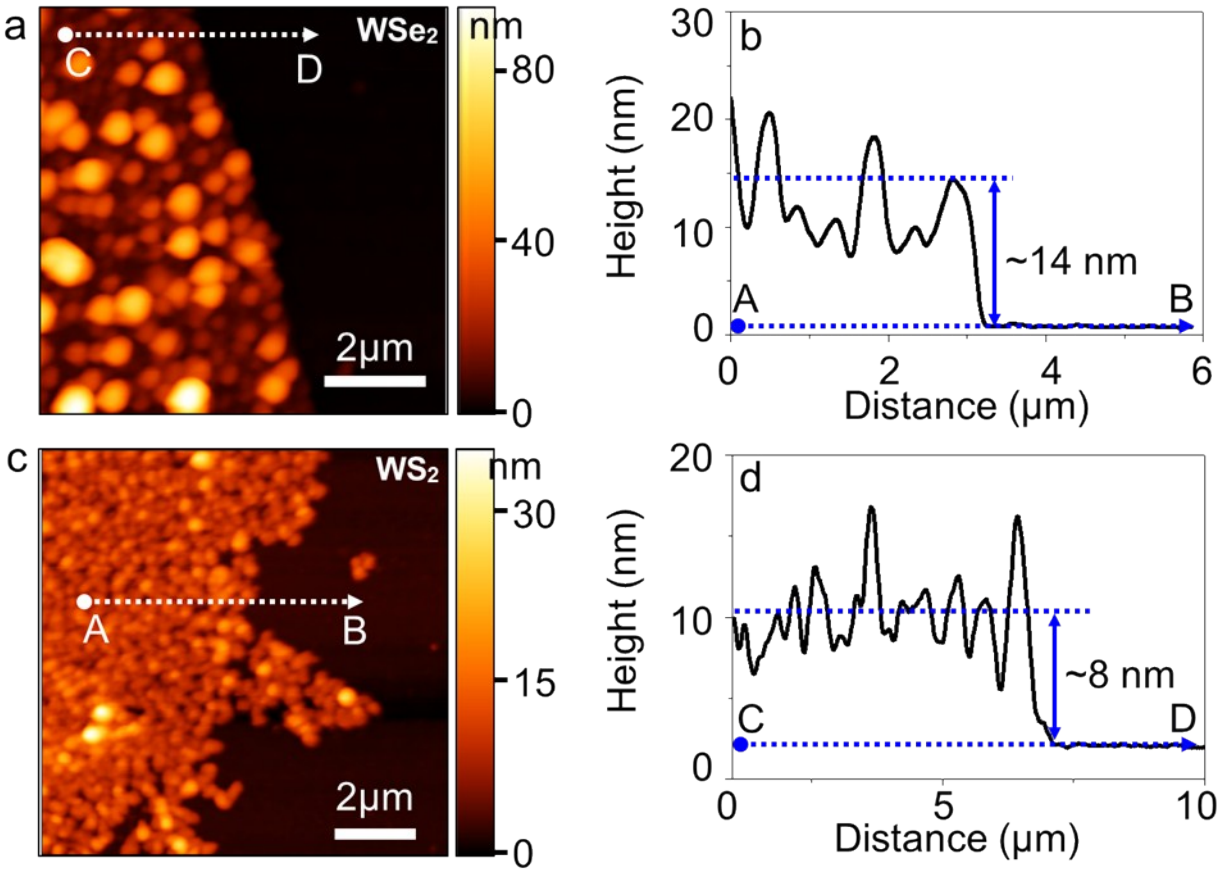

Fig. S1. (a) and (c) AFM mapping images of WSe_2 and WS_2 thin films. (b) and (d) Corresponding AFM height profiles (white lines, A–B and C–D) in (a) and (c). The average thicknesses of the WSe_2 and WS_2 thin films were ~14 and ~8 nm, respectively.

3. Power factors for CVD-grown TMDC films

Fig. S2a shows the in- and cross-plane power factors $(PF = S^2\sigma)$ of CVD-grown MoS_2 films at 300 K with an anisotropic ratio $\left(\frac{PF_{\parallel}}{PF_{\perp}}\right)$ of the power factor. The $PF_{\parallel}$ and $PF_{\perp}$ of the MoS_2 films were determined to be ~432.6 and ~1.8×10^{-6} μW/ m·K^2, respectively, with an extremely large anisotropic ratio of ~2.4×10^8 at 300 K. Fig. S2b shows the cross-plane power factors $(PF_{\perp})$ of MoS_2, WSe_2, and WS_2 films at 300 K measured as ~1.8×10^{-6}, ~2.8×10^{-7}, and ~1.9×10^{-7} μW/ m·K^2.

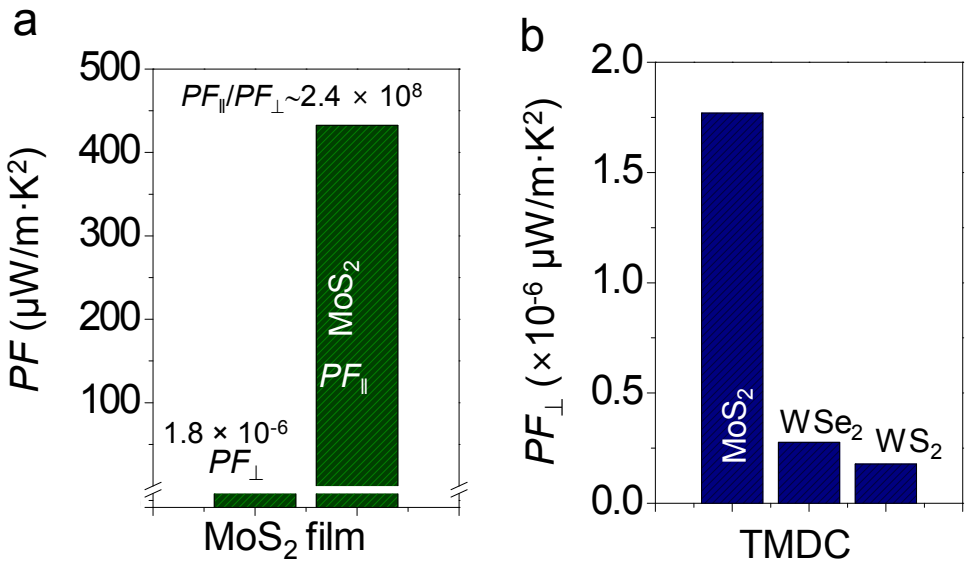

Fig. S2. (a) In- and cross-plane power factors ($PF_{\parallel}$ and $PF_{\perp}$) of CVD-grown MoS_2 films with power factor anisotropy. (b) Cross-plane $PF_{\perp}$ of MoS_2, WSe_2, and WS_2 films at 300 K.

4. Thermoelectric characteristics of various TMDC materials from literatures

Table S1. The comparison of thermoelectric performance of MoS_2 in our work with the thermoelectric performances of TMDCs reported in previous literatures

Materials	Thickness	$\sigma_{\parallel}$ [S/m]	$\sigma_{\perp}$ (S/m)	|S| [uV/K] @ 300 K	Ref
MoS_2	1 – 2 layers	-	-	420 ~ 1 x 10^5	[S1 – S3]
MoS_2	Thin film	17~28.3	-	80~ 93.5	[S4 – S5]
MoS_2	bulk	0.2~1.7	-	100 ~ 150	[S6]
$MoSe_{2.1}$	bulk	8×10^{-6}	3×10^{-6}	~650	[S7]
WSe_2	bulk	0.001~0.19	-	783 ~ 900	[S8 – S9]
WS_2	Thin film	10 ~ 1 × 10^3	9	75~ 700.78	[S10 – S12]
$WS_{2-x}Se_x$	Thin film	0.021 ~ 40	-	300~945	[S13 – S14]
MoS_2	Thin film	~786	~1×10^{-4}	115 ~ 742	This work

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