

Supplementary information for **Precise p -type and n -type doping of two-dimensional semiconductors for monolithic integrated circuits**

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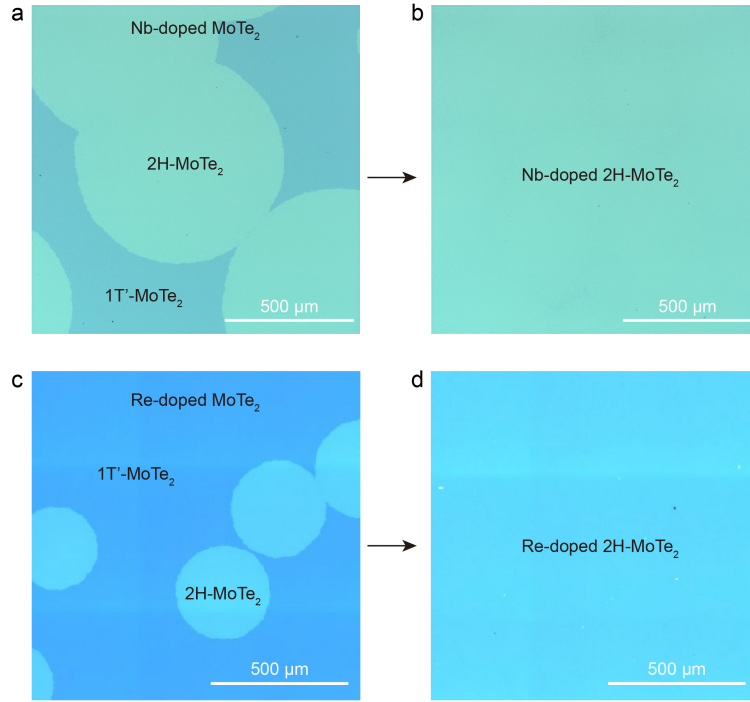
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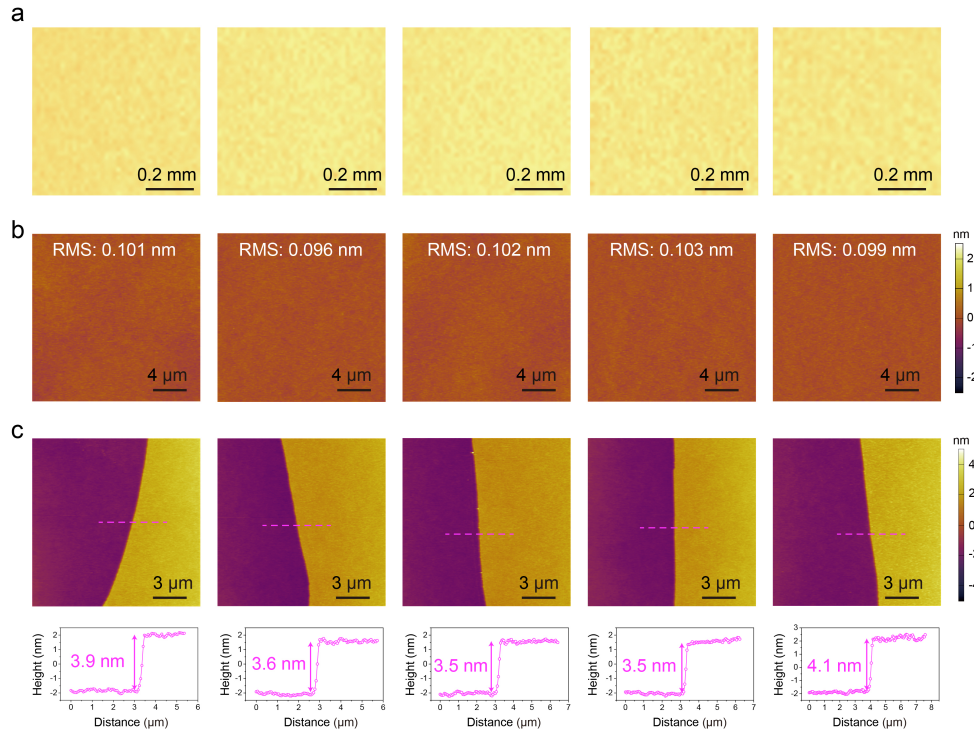
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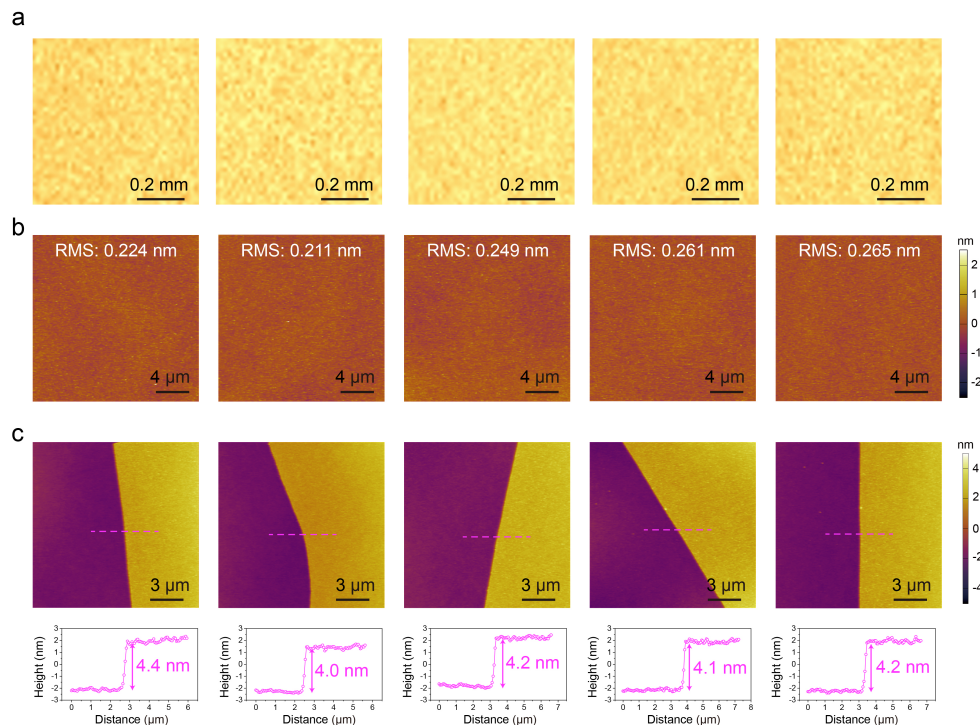
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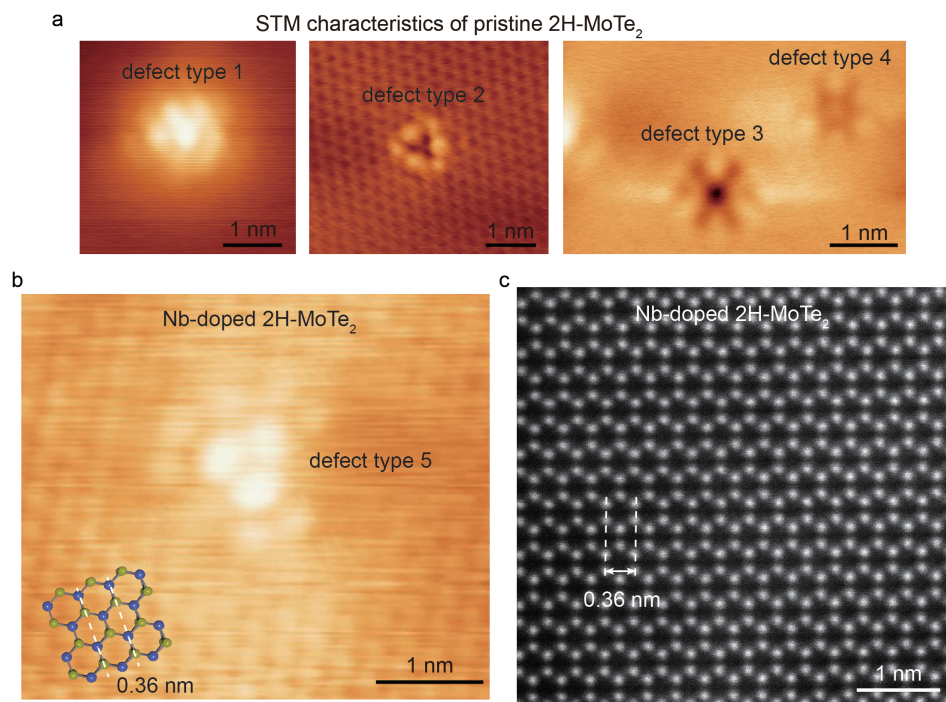
Supplementary Fig. 1 Preparation of the doped 2H-MoTe₂ films. **a, c,** MoTe₂ films doped with Nb or Re grown on Si/SiO₂ substrates for a short period of time. 2H-MoTe₂ circles with a domain size of a few hundred micrometers can be seen. **b, d,** As growth time prolongs, these 2H domains grow gradually and merge together to form a uniform 2H-MoTe₂ film with Nb doping or Re doping.



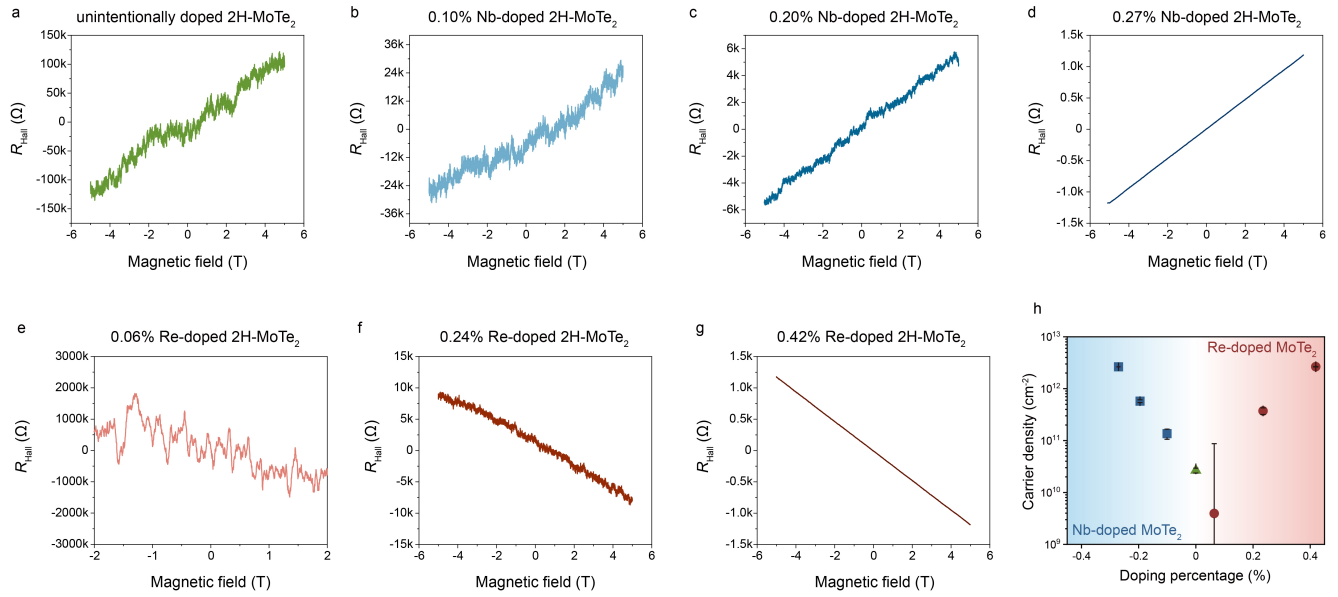
Supplementary Fig. 2 Large-scale uniformity of the as-synthesized Nb-doped 2H-MoTe₂ films. Raman mappings and AFM height images were measured at random 5 different positions across the wafer. **a,** Raman mappings of the representative 2H-MoTe₂ Raman peak intensity (E_{2g}^1 234 cm⁻¹), showing the high uniformity of the Nb-doped 2H-MoTe₂ film. The areas of the Raman mappings are 0.7 mm × 0.7 mm. **b-c,** AFM height images of Nb-doped 2H-MoTe₂ films measured at 5 different positions. The surface roughness (RMS) was in the range of 0.096-0.103 nm, and no holes were observed on the film surface, indicating that the film was continuous and very smooth. Meanwhile, the thickness of the Nb-doped 2H-MoTe₂ films is about 3.5-4.1 nm, corresponding to 5-6 layers of MoTe₂.



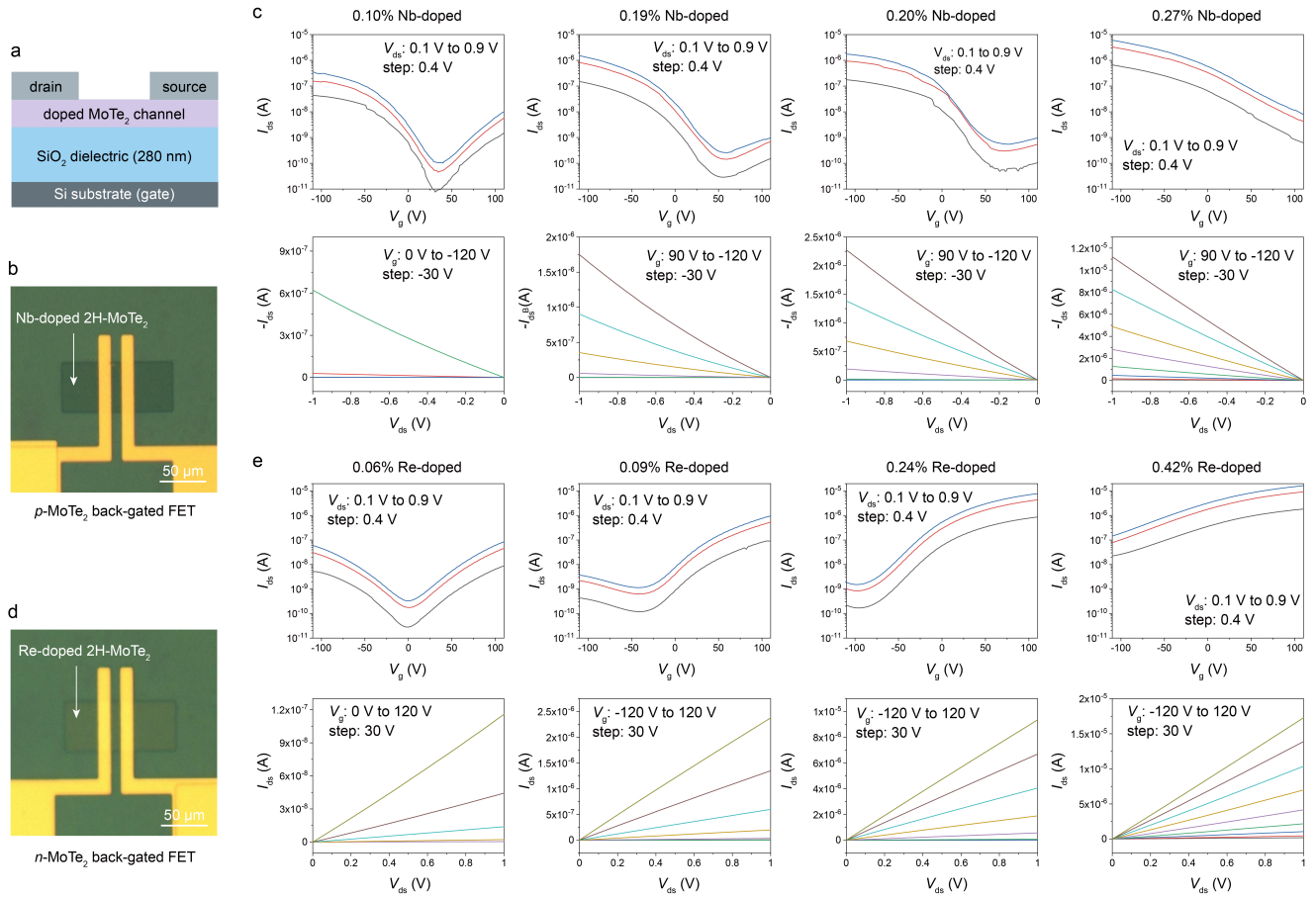
Supplementary Fig. 3 Large-scale uniformity of the as-synthesized Re-doped 2H-MoTe₂ films. Raman mappings and AFM height images were measured at random 5 different positions across the wafer. **a**, Raman mappings of the representative 2H-MoTe₂ Raman peak intensity (E_{2g}^1 234 cm^{-1}), showing the high uniformity of the Re-doped 2H-MoTe₂ film. The areas of the Raman mappings are 0.7 mm $\times$ 0.7 mm. **b-c**, AFM height images of Re-doped 2H-MoTe₂ films measured at 5 different positions. The surface roughness (RMS) was in the range of 0.211-0.265 nm, and no holes were observed on the film surface, indicating that the film was continuous and very smooth. Meanwhile, the thickness of the Re-doped 2H-MoTe₂ films is about 4.0-4.4 nm, corresponding to 6-7 layers of MoTe₂.



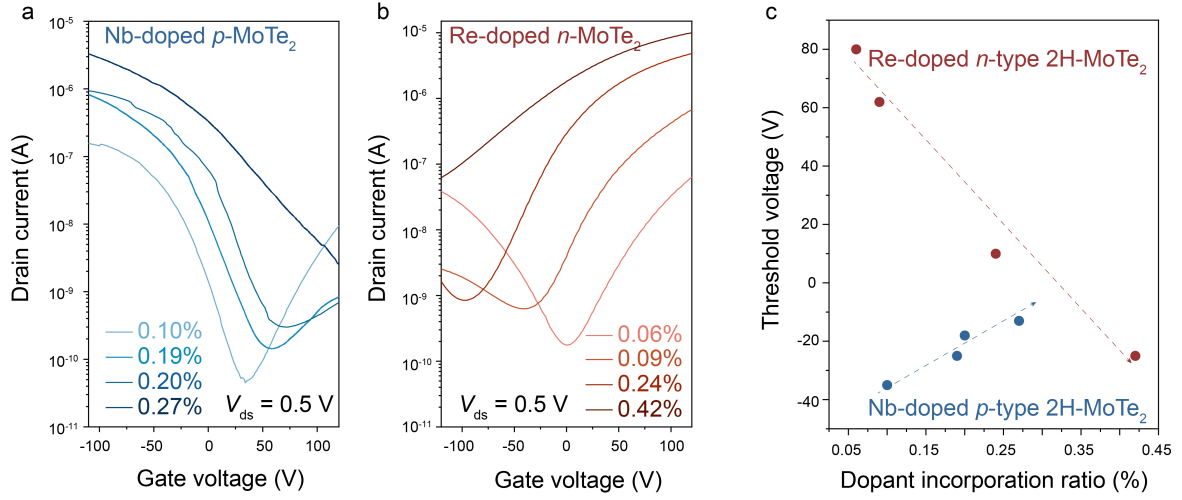
Supplementary Fig. 4 Scanning tunneling microscope (STM) characterizations of 2H-MoTe₂ films. **a** and **b**, STM topography images of the unintentionally doped 2H-MoTe₂ and Nb-doped films grown directly on HOPG substrates. Zoomed-in STM topography of Nb-doped 2H-MoTe₂, with structural model superimposed on top, showing a clear hexagonal 2H-MoTe₂ lattice with a lattice constant of 0.36 nm. The measured lattice constant is consistent with the result from the HAADF-STEM image shown in **c**. **c**, HAADF-STEM image of Nb-doped 2H-MoTe₂, showing a lattice constant of 0.36 nm.



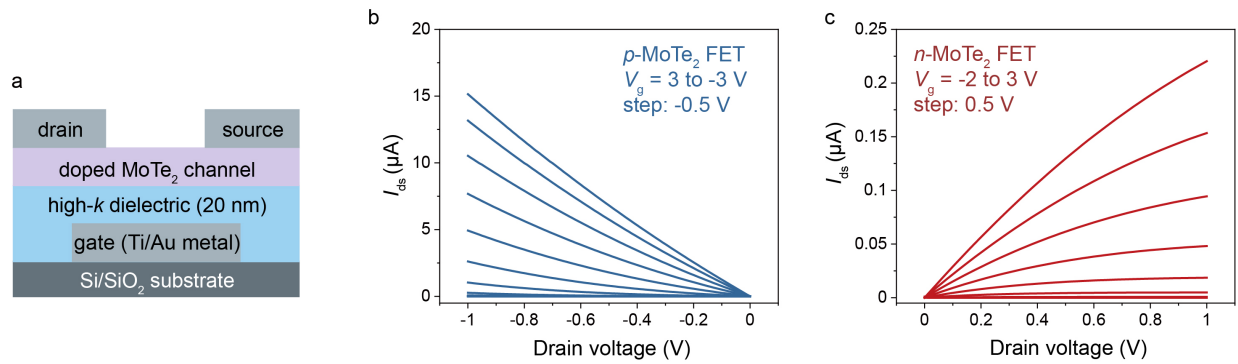
Supplementary Fig. 5 The magnetic field-dependent Hall resistance of the doped 2H-MoTe₂ thin films. **a**, The unintentionally doped case, showing a *p*-type behavior with a low background hole concentration. **b-d**, Nb-doped 2H-MoTe₂ cases with Nb incorporation ratio from 0.10% to 0.27%, showing the conduction hole-conducting *p*-type. **e-g**, Re-doped 2H-MoTe₂ cases with Re incorporation ratio from 0.06% to 0.42%, showing the conduction electron-conducting *p*-type. The inverse of the slope of the R_{Hall} versus B curve determines the carrier concentration. **h**, Hole or electron carrier densities of 2H-MoTe₂ semiconductor films with various Nb or Re incorporation ratios through logarithmic transformation.



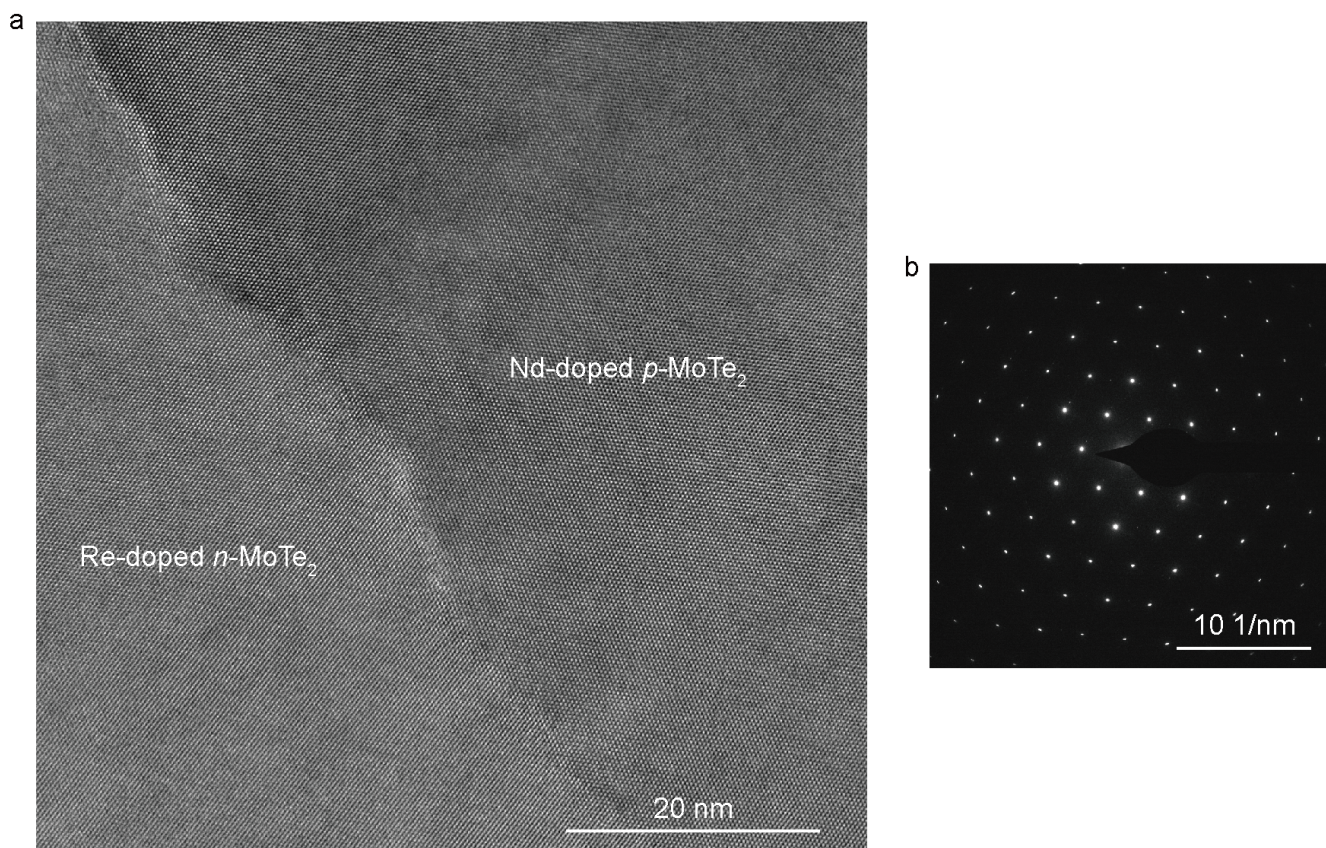
Supplementary Fig. 6 FET characteristics of 2H-MoTe₂ channels with different Nb or Re incorporation ratios. **a**, Schematic diagram of the structure of a doped 2H-MoTe₂ back-gate FET. To minimize process-induced doping effects, we fabricated back-gate FETs on 2H-MoTe₂ thin films with different doping types and carrier concentrations directly grown on commercial Si/SiO₂ substrates. **b**, **d**, Optical images of Nb-doped and Re-doped 2H-MoTe₂ back-gated FETs. These FETs have channels of the same size, with a length of 10 μm, a width of 40 μm, and a thickness of 5 nm. **c**, **e**, Transfer and output characteristics of the back-gated FETs with different incorporation ratios of Nb (**b**) and Re (**d**). The Nb-doped (Re-doped) 2H-MoTe₂ channel exhibits *p*-type (*n*-type) semiconductor transfer characteristic, and its on-current increases with the increase of doping concentration, while the transistor threshold voltage increases towards the positive (negative) voltage direction. The adjustment of transistor threshold voltage with doping concentration is summarized in Supplementary Fig. 7.



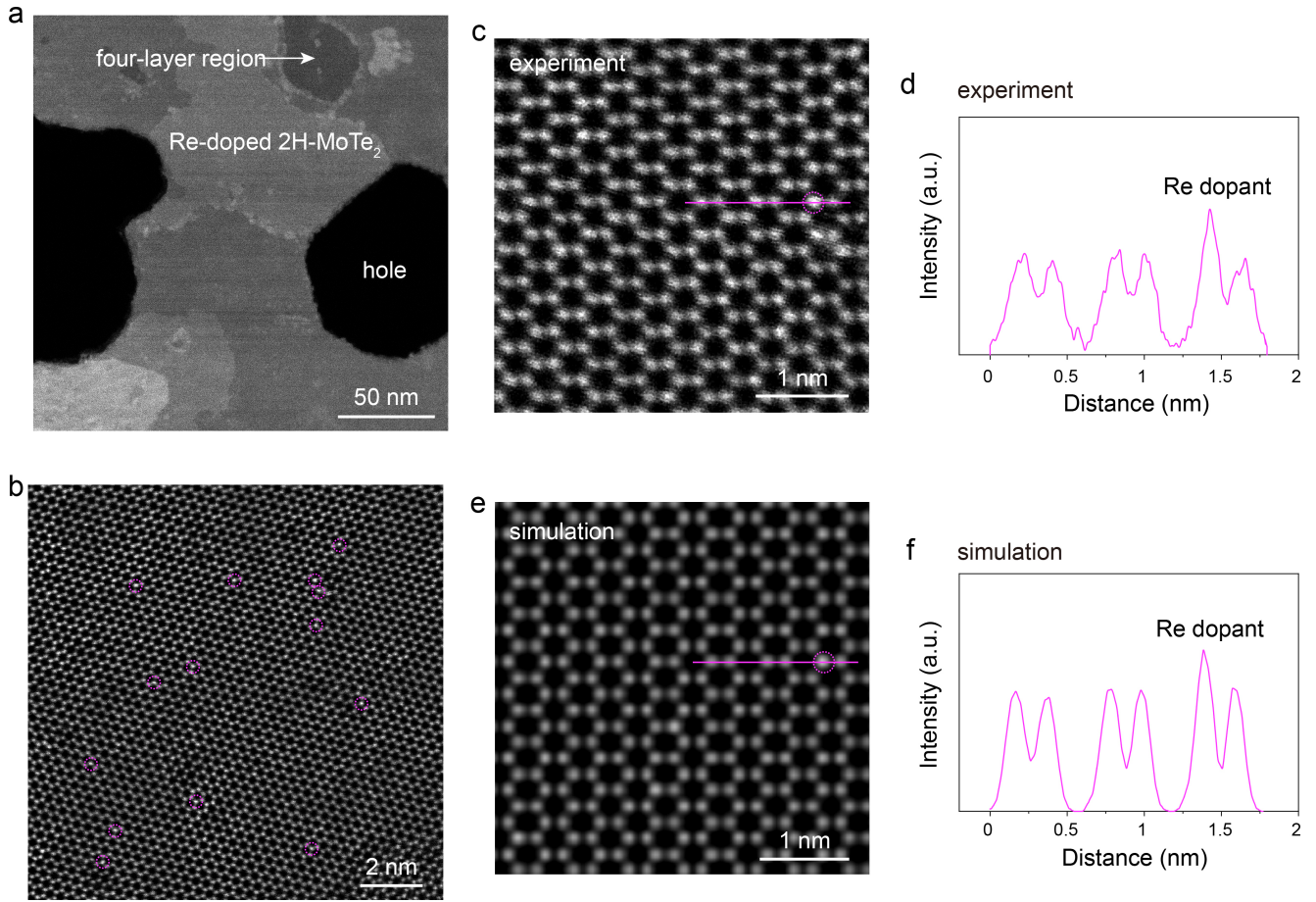
Supplementary Fig. 7 The transfer characteristics of 2H-MoTe₂ FETs as a function of dopant incorporation ratio. **a** and **b**, Transfer characteristics of the *p*- and *n*-type 2H-MoTe₂ FETs with various doping concentrations. **c**, The transistor threshold voltage of *p*-type (*n*-type) 2H-MoTe₂ increases (decreases) with doping, where the dashed lines with arrows indicate the evolution of threshold voltage with doping concentration.



Supplementary Fig. 8 Electrical properties of typical *p*-type (0.20% Nb-doped) and *n*-type (0.24% Re-doped) 2H-MoTe₂ FETs. **a**, Schematic diagram of the structure of a doped 2H-MoTe₂ back-gate FET. The devices utilized a back-gate transistor structure with 20 nm Al₂O₃ dielectric and a channel *W*/*L* of 40/10 μm. The doped 2H-MoTe₂ films were transferred onto the substrate with back-gate electrodes (Ti/Au, 5/20 nm). **b** and **c**, Output characteristics of *p*-type (**b**) and *n*-type (**c**) 2H-MoTe₂ FETs.

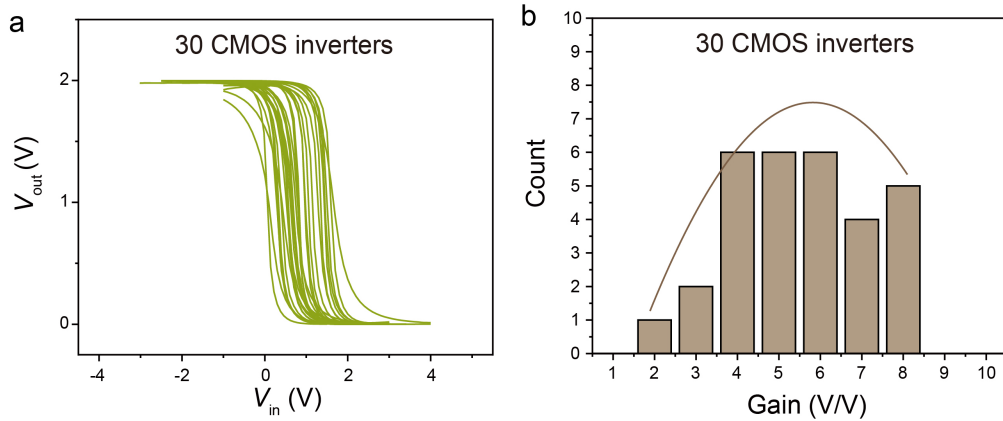


Supplementary Fig. 9 Characterization of atomic structure and crystal orientation at the interface of 2H-MoTe₂ pn structure. **a**, Large field-of-view high-resolution TEM image at the interface of Nb- and Re-doped 2H-MoTe₂ pn structure. **b**, The corresponding SAED pattern at the interface with an aperture size of 800 nm, which shows only one hexagonal lattice of the single crystal.

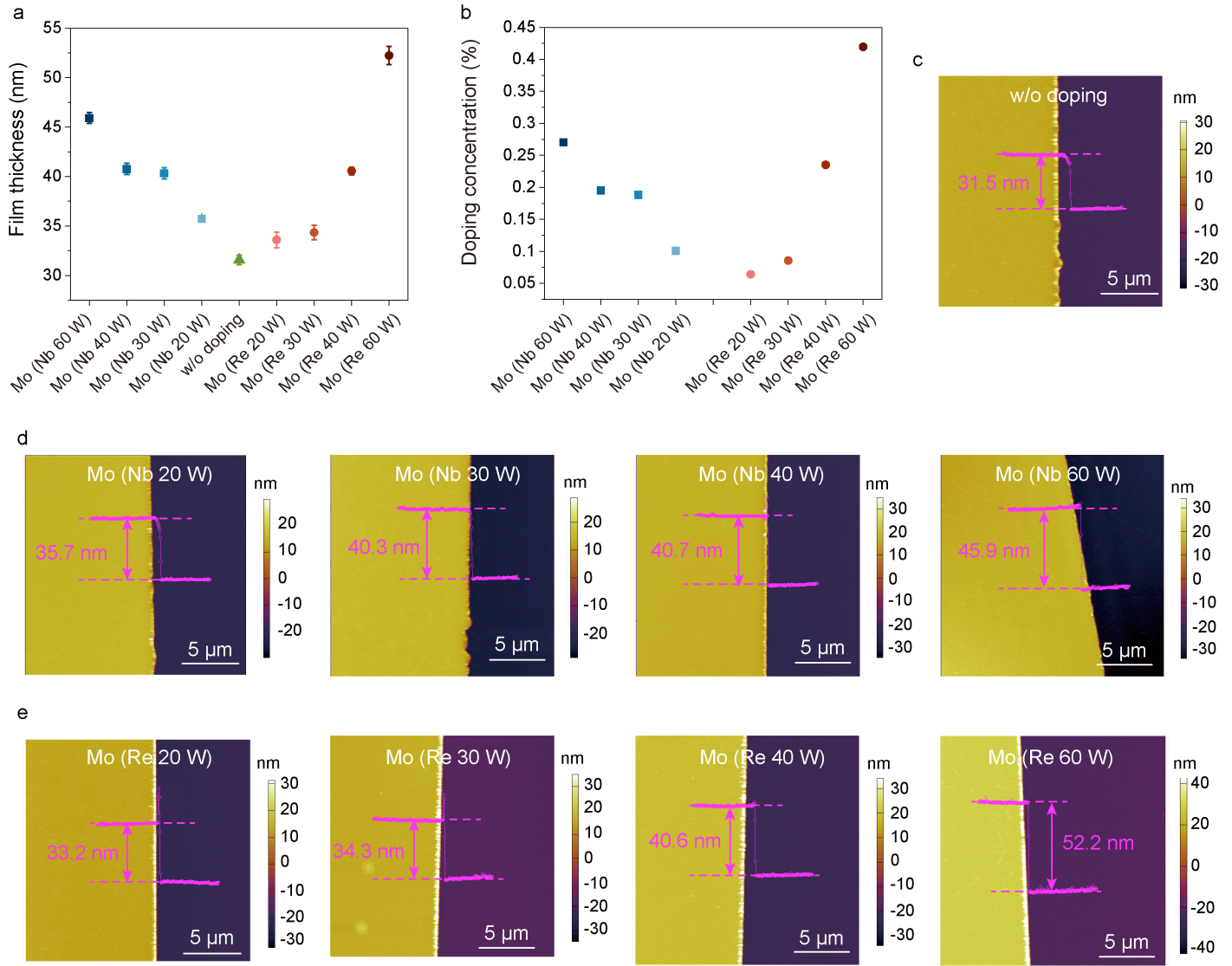


Supplementary Fig. 10 STEM characterizations of the Re-doped thin 2H-MoTe₂ film. **a**, Large field-of-view HAADF-STEM image of a Re-doped 2H-MoTe₂ thin film with some holes and thin four-layer regions. **b**, Zoomed-in HAADF-STEM image of four-layer 2H-MoTe₂ region, showing discernible doping of Re atoms marked by the pink dotted circles. **c, e**, Experimental and simulated HAADF-STEM images of the four-layer 2H-MoTe₂ doped with Re atoms. **d, f**, Line profiles of the image contrast from experimental (**c**) and simulated (**e**) HAADF-STEM images containing one Re dopant. The intensity at the site containing Re is about 30% higher than that of Mo only, and the experimental and simulation results agree very well.

Due to the large atomic number difference between Re and Mo, it is possible to resolve the atomic substitutional doping of Re in the thin Re-doped 2H-MoTe₂. Therefore, we magnetron sputtered a 0.5 nm thick Mo film with a Re incorporation ratio of 0.85% on a SiO₂ substrate. Since the 0.5 nm thick Mo film is discontinuous, we will find a discontinuous 2H-MoTe₂ thin film after tellurization. Under the low-magnification HAADF-STEM image (Supplementary Fig. 10a), we will find some discrete holes, 2H-MoTe₂ of different thicknesses, and occasionally some thinner four-layer regions.



Supplementary Fig. 11 Statistical electrical characterizations of CMOS inverter arrays. **a**, Typical transfer characteristics of 30 inverters composed of *p*-type and *n*-type MoTe₂ transistors. **b**, Statistical histogram distribution of inverter gains in **a** with Gaussian fit.



Supplementary Fig. 12 The thicknesses of samples under different doping target power characterized by AFM to calculate the doping concentration. **a**, Film thicknesses measured by AFM after sputtering for 30 min with sputtering powers of doping targets (Mo target containing Nb or Re) of 0 W, 20 W, 30 W, 40 W, and 60 W, respectively. **b**, The doping concentrations of dopants in Mo films were calculated from these samples with different sputtering powers of the doping targets. **c-e**, AFM height images of the samples with different sputtering powers of doping targets.

In this work, co-sputtering was used to control the doping concentration. The Mo target was connected to a DC power supply with a constant power of 10 W, and the Mo target containing 1% Nb or 1% Re was used as a doping target, and was connected to an AC power supply. The sputtering power of the doping target was used to control the doping concentration of the dopant, while the co-sputtering time was used to control the thickness of the resulting Mo film. To accurately calculate the doping concentration, we prolonged the co-sputtering time to 30 min and measured the thickness of the resulting film using AFM. In the absence of doping, the thickness of the Mo film was measured to be 31.5 ± 0.5 nm (Supplementary Fig. 12a and c). In all AFM measurements, 70 positions were selected to measure the film thickness, and the average value was defined as the Mo film thickness. Assuming the sputtering rate remains constant, we can calculate the sputtering rate of the Mo target is about 1.05 nm/min.

During the co-sputtering deposition process, the Mo target was kept at a constant DC power of 10 W, and the AC sputtering power of the doping target was set to 20 W, 30 W, 40 W, and 60 W, respectively. After a 30 min co-sputtering, the thicknesses of the Nb-incorporated Mo films were 35.7 ± 0.32 nm, 40.3 ± 0.57 nm, 40.7 ± 0.58 nm, and 45.9 ± 0.56 nm, while the thicknesses of the Re-incorporated Mo films were 33.2 ± 0.36 nm, 34.3 ± 0.74 nm, 40.6 ± 0.39 nm and 52.2 ± 0.91 nm (Supplementary Fig. 12a, d and e). The increased thickness compared with the case without doping comes from the sputtered thickness of the doping target. As the dopant (Nb or Re) incorporation concentration of the doping target is known (1%), we can calculate the doping concentration of the dopants in the resulting films with different AC powers:

$$\text{Doping ratio} = \frac{(t - 31.5) \times 1\% \times \rho_{\text{dopant}} \times M_{\text{Mo}}}{t \times \rho_{\text{Mo}} \times M_{\text{dopant}}}$$

where t is the measured film thickness, ρ is the mass density, M is the molar mass. Therefore, at AC powers of 20 W, 30 W, 40 W, and 60 W, the calculated incorporation ratios of Nb were 0.10%, 0.19%, 0.20%, and 0.27%, while those of Re were 0.06%, 0.09%, 0.24% and 0.42% (Supplementary Fig. 12b).