

# High- $\kappa$ perovskite membranes as insulators for two-dimensional transistors

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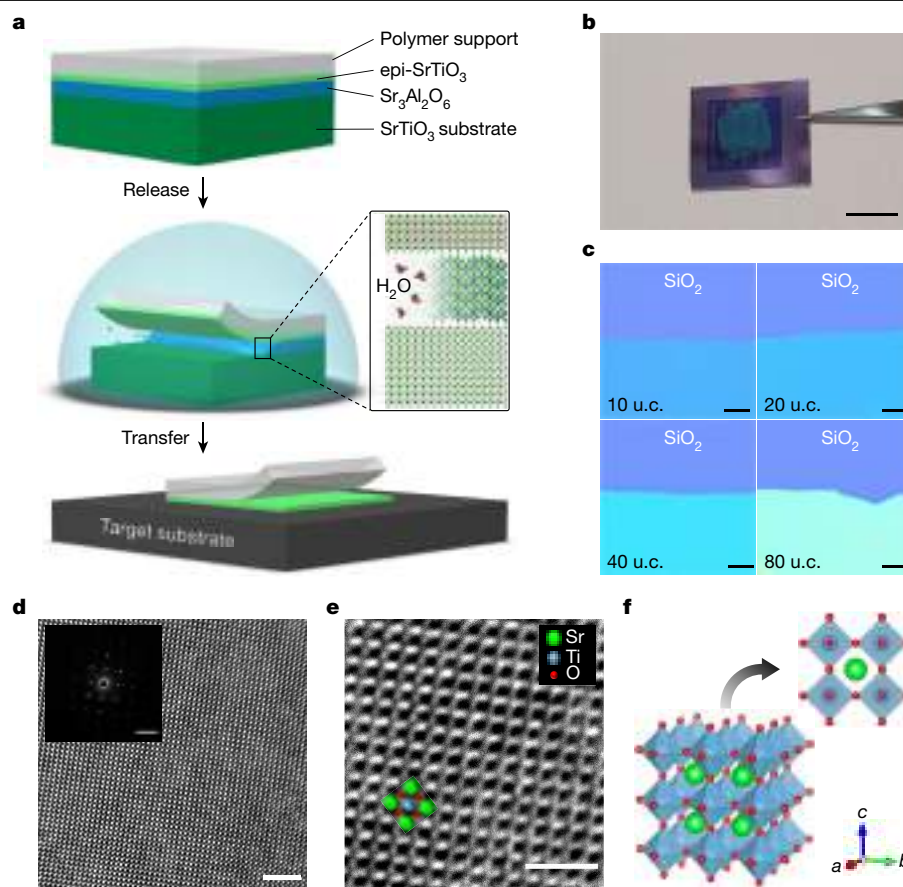
The scaling of silicon metal–oxide–semiconductor field-effect transistors has followed Moore’s law for decades, but the physical thinning of silicon at sub-ten-nanometre technology nodes introduces issues such as leakage currents<sup>1</sup>. Two-dimensional (2D) layered semiconductors, with an atomic thickness that allows superior gate-field penetration, are of interest as channel materials for future transistors<sup>2,3</sup>. However, the integration of high-dielectric-constant ( $\kappa$ ) materials with 2D materials, while scaling their capacitance equivalent thickness (CET), has proved challenging. Here we explore transferrable ultrahigh- $\kappa$  single-crystalline perovskite strontium-titanium-oxide membranes as a gate dielectric for 2D field-effect transistors. Our perovskite membranes exhibit a desirable sub-one-nanometre CET with a low leakage current (less than  $10^{-2}$  amperes per square centimetre at 2.5 megavolts per centimetre). We find that the van der Waals gap between strontium-titanium-oxide dielectrics and 2D semiconductors mitigates the unfavourable fringing-induced barrier-lowering effect resulting from the use of ultrahigh- $\kappa$  dielectrics<sup>4</sup>. Typical short-channel transistors made of scalable molybdenum-disulfide films by chemical vapour deposition and strontium-titanium-oxide dielectrics exhibit steep subthreshold swings down to about 70 millivolts per decade and on/off current ratios up to  $10^7$ , which matches the low-power specifications suggested by the latest International Roadmap for Devices and Systems<sup>5</sup>.

For the sub-10-nm technology nodes in silicon (Si) metal–oxide–semiconductor field-effect transistors (MOSFETs), a subnanometre capacitance equivalent thickness (CET) and flawless interface with the channel are essential for gate dielectrics to maintain the gate controllability<sup>4</sup>. Similarly, the development of reliable high-dielectric-constant ( $\kappa$ ) materials (CET < 1 nm) adaptable to two-dimensional (2D) MOSFETs for future nodes is eagerly awaited. The typically used high- $\kappa$  dielectrics in Si technology (that is, aluminium oxide (Al<sub>2</sub>O<sub>3</sub>) and hafnium oxide (HfO<sub>2</sub>)) have been integrated with 2D transition-metal-dichalcogenide materials<sup>6</sup>. Nevertheless, their amorphous nature and imperfect dielectric/transition-metal-dichalcogenide interfaces make the elimination of charge scatters and traps difficult, in addition to the direct damage of 2D channels caused by dielectric deposition processes<sup>7,8</sup>. Although several interfacial passivation layers have been developed, such as organic molecules and atomically thin hexagonal boron nitride (hBN), these layers reduce the overall gate capacitance<sup>8–11</sup>. Another attractive approach is to adopt crystalline dielectric materials such as multilayer

hBN and epitaxial calcium fluoride (CaF<sub>2</sub>), where the atomically flat surfaces result in smoother dielectric/semiconductor interfaces than conventional amorphous oxides, despite their relatively lower dielectric constant that retards the CET shrinkage<sup>12–14</sup>. The perovskite strontium titanium oxide (SrTiO<sub>3</sub>) exhibits high static permittivity ( $\epsilon_{\text{bulk}} \approx 300$  at room temperature)<sup>15</sup>, which makes it a potential gate dielectric for electrostatic modulation of Si<sup>16,17</sup>, graphene<sup>18,19</sup> or 2D electron gases at complex oxide heterointerfaces<sup>20,21</sup>. Moreover, recent advances in synthesizing single-crystal freestanding perovskite-oxide membranes<sup>22,23</sup> have established a feasible route to integrate the ultrahigh- $\kappa$  crystalline SrTiO<sub>3</sub> films with 2D semiconductors to form high-quality dielectric/channel interfaces to overcome the present limit of gate control.

In this work, the reflection high-energy electron diffraction (RHEED)-assisted pulsed-laser-deposition technique is used to prepare freestanding SrTiO<sub>3</sub> dielectric layers (see Methods for details)<sup>22</sup>. The water-soluble sacrificial strontium aluminate (Sr<sub>3</sub>Al<sub>2</sub>O<sub>6</sub>) layer is epitaxially grown on top of a single-crystalline SrTiO<sub>3</sub> (001) substrate,

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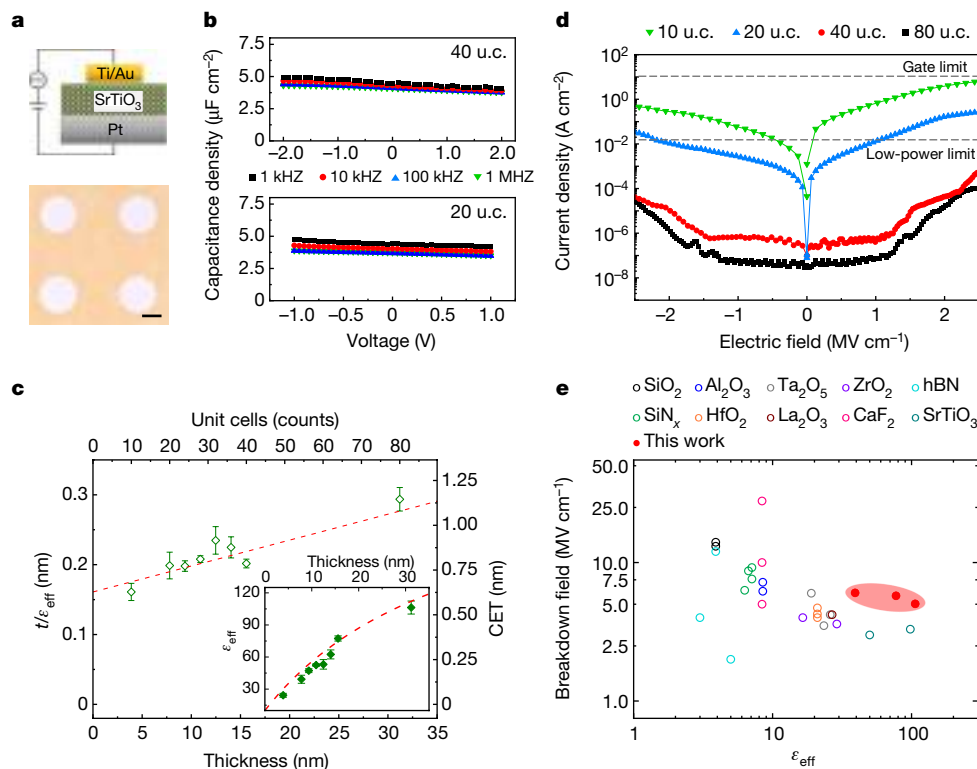
**Fig. 1 | Preparation and characterizations of freestanding single-crystalline SrTiO<sub>3</sub> layers.** **a**, Schematic illustration of the transfer processes of epi-SrTiO<sub>3</sub> thin films onto the target substrate. **b**, Photograph of the transferred millimetre-scale freestanding SrTiO<sub>3</sub> membrane on pre-patterned Si/SiO<sub>2</sub> substrates. Scale bar, 5 mm. **c**, Optical micrographs of SrTiO<sub>3</sub> films with various unit-cell thicknesses on 300-nm Si/SiO<sub>2</sub> wafers.

Violet-blue is Si/SiO<sub>2</sub> substrates and the cyan gradient represents SrTiO<sub>3</sub> films with various thicknesses. Scale bars, 25  $\mu$ m. **d**, **e**, Plan-view dark-field TEM images of 5-u.c.-thick SrTiO<sub>3</sub> membrane. Scale bars, 2 nm (**d**) and 1 nm (**e**). Inset: the corresponding SAED pattern. Scale bar, 5 nm<sup>-1</sup>. **f**, Cubic lattice structure of SrTiO<sub>3</sub>.

which then serves as the template for the subsequent epitaxial growth of single-crystalline SrTiO<sub>3</sub> dielectric membranes. The as-deposited SrTiO<sub>3</sub> films exhibit atomically flat surfaces with clear atomic-step terraces (Extended Data Fig. 1a, b). The X-ray diffraction (XRD) pattern (Extended Data Fig. 1c) and X-ray reflectivity (XRR) analysis (Extended Data Fig. 1d, e) of the as-prepared Sr<sub>3</sub>Al<sub>2</sub>O<sub>6</sub>/SrTiO<sub>3</sub> heterostructure clearly indicate the formation of single-crystalline phases and the atomically sharp interface. Next, the SrTiO<sub>3</sub> layer is coated with a polymer support and delaminated from the substrate after dissolving the Sr<sub>3</sub>Al<sub>2</sub>O<sub>6</sub> sacrificial layer in deionized water. With the polymer support layer, the released single-crystalline freestanding SrTiO<sub>3</sub> membranes are transferred onto the target substrates, as illustrated in Fig. 1a. A photo of the as-released SrTiO<sub>3</sub> layer with a dimension of 5  $\times$  5 mm<sup>2</sup> on the pre-patterned Si/SiO<sub>2</sub> substrate is shown in Fig. 1b. The optical micrographs in Fig. 1c show that the optical contrast gradually increases with increasing SrTiO<sub>3</sub> thickness. The thickness of the SrTiO<sub>3</sub> membranes is determined by atomic force microscopy (AFM) (Extended Data Fig. 2). Representative reciprocal space maps (RSMs) of the transferred SrTiO<sub>3</sub> membrane around the (002), (103) and (013) planes confirm the single crystallinity, and the extracted average in-plane and out-of-plane lattice parameters are 0.3908 nm and 0.3907 nm, respectively (Extended Data Fig. 3a, b), which are within the measurement errors of the theoretical unit-cell (u.c.) value of 0.3905 nm (ref. 22). The plan-view dark-field transmission electron microscopy (TEM) images and the selected-area electron diffraction (SAED) patterns (Fig. 1d, e) of 5-u.c.-thick SrTiO<sub>3</sub> demonstrate its single-crystal nature

with very limited defects in the probed areas (Fig. 1f). Moreover, X-ray photoelectron spectroscopy (XPS) results (Extended Data Fig. 3c, d) suggest the ideal atomic stoichiometry of the SrTiO<sub>3</sub> film.

The metal–insulator–metal (MIM) capacitor structure is used to evaluate the capacitance–voltage (*C*–*V*) characteristics of the as-prepared SrTiO<sub>3</sub> membranes<sup>9,24</sup>, where the SrTiO<sub>3</sub> layer is sandwiched by a platinum (Pt)-coated Si substrate and a top titanium (Ti)/gold (Au) electrode (Fig. 2a). Figure 2b shows that the measured capacitance density moderately decreases with the voltage and frequency, which is ordinarily observed in a paraelectric ceramic and asymmetric electrode configuration<sup>25</sup>. The effective permittivity  $\epsilon_{\text{eff}}$  is related to the capacitance as described by the equation:  $C_{\text{eff}} = A\epsilon_0\epsilon_{\text{eff}}/t$ , where  $C_{\text{eff}}$  is the measured capacitance,  $A$  is the area of the top electrodes,  $t$  is the thickness of the SrTiO<sub>3</sub> layer and  $\epsilon_0$  is the vacuum permittivity<sup>24</sup>. The inset of Fig. 2c shows the thickness dependence of  $\epsilon_{\text{eff}}$ , where the result for a particular thickness is obtained from the measurements conducted on at least 17 individual devices made of SrTiO<sub>3</sub> membranes. The  $\epsilon_{\text{eff}}$  as a function of thickness can be well described by the typical ‘dead layer’ model emerging in a high- $\kappa$  nanocapacitor<sup>24</sup> (details in Supplementary Note 1) as follows:  $t/\epsilon_{\text{eff}} = t/\epsilon_{\text{bulk}} + D$ , where  $\epsilon_{\text{bulk}}$  is the bulk permittivity of the dielectric material and  $D$  is the derived constant resulting from the interfacial dead layers. Figure 2c shows that the CET exhibits a near-linear relationship with the SrTiO<sub>3</sub> thickness, implying that a sub-1-nm CET can be achieved with an oxide thickness thinner than 26 nm (about 65 u.c.). The CET is calculated as  $3.9t/\epsilon_{\text{eff}}$ , where 3.9 is the dielectric constant of silicon oxide<sup>4</sup>. In addition, the constant



**Fig. 2 | Dielectric properties of single-crystalline SrTiO<sub>3</sub> membranes.**

**a**, The structure and optical micrograph of the MIM device. Scale bar, 100  $\mu\text{m}$ . **b**, Voltage-dependent capacitance density ( $C$ – $V$ ) for MIM devices of 40-u.c.-thick and 20-u.c.-thick SrTiO<sub>3</sub> thin films at four different frequencies. **c**,  $t/\epsilon_{\text{eff}}$  and CET as a function of various SrTiO<sub>3</sub> thicknesses measured from MIM capacitors. The inset shows  $\epsilon_{\text{eff}}$  as a function of SrTiO<sub>3</sub> thickness. The red

dashed lines represent the fitted curves using the ‘dead layer’ model. The error bars represent the standard deviation. **d**, Electric-field-dependent leakage current density of SrTiO<sub>3</sub> films with various unit-cell thicknesses. The grey dashed lines mark the limits for relative applications. **e**, The breakdown field versus effective dielectric constant of our SrTiO<sub>3</sub> membranes, comparison with various dielectric materials<sup>25,26,28–32</sup>.

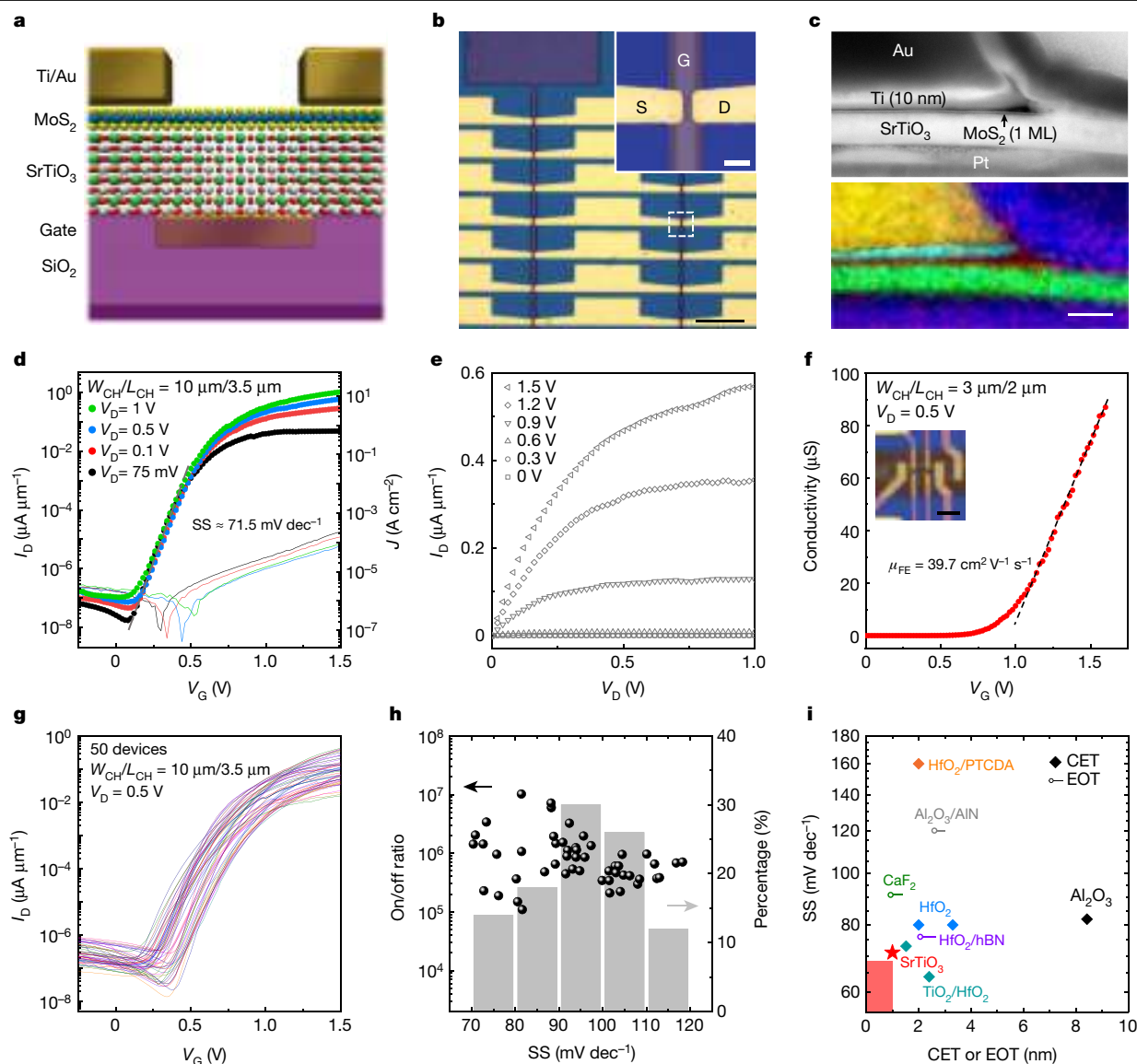
$D \approx 0.161$  and  $\epsilon_{\text{bulk}} \approx 270$  are extracted from the linear fit, where  $\epsilon_{\text{bulk}}$  approaches the ideal value ( $\epsilon_{\text{bulk}} \approx 300$ )<sup>15</sup> of a bulk SrTiO<sub>3</sub> single crystal, which indicates the excellent quality and ultrahigh- $\kappa$  nature of the SrTiO<sub>3</sub> membranes.

By contrast, a low leakage current and high breakdown strength of dielectric materials are the essential performance parameters that are related to the device power consumption and reliability<sup>4,8,26</sup>. Figure 2d shows the leakage current characteristics of the as-prepared SrTiO<sub>3</sub> membranes with thicknesses of 10 u.c., 20 u.c., 40 u.c. and 80 u.c., respectively. For the applied field of 2.5 MV cm<sup>−1</sup>, the leakage currents of the 40-u.c.-thick and 80-u.c.-thick SrTiO<sub>3</sub> membranes are far below the low-power limit (leakage current  $J_{\text{leak}} < 1.5 \times 10^{-2} \text{ A cm}^{-2}$ ), and all investigated membranes meet the MOSFET gate-limit requirement ( $< 10 \text{ A cm}^{-2}$ )<sup>27</sup>. Moreover, Fig. 2e compares the breakdown field ( $E_{\text{bd}}$ ) versus  $\epsilon_{\text{eff}}$  for the dielectrics used in the state-of-art Si and 2D semiconductor technologies<sup>25,26,28–32</sup>. Our optimized SrTiO<sub>3</sub> membranes (20 u.c., 40 u.c. and 80 u.c.; Supplementary Note 2) have comparable  $E_{\text{bd}}$  values to widely used Al<sub>2</sub>O<sub>3</sub> and HfO<sub>2</sub> while possessing much higher effective permittivities. In clear contrast to the high- $\kappa$  dielectrics produced by deposition processes, the freestanding SrTiO<sub>3</sub> membrane that possesses smooth and well defined surfaces and an elastic nature behaves as a quasi-2D layer to interface with adjacent materials using a van der Waals (vdW) gap (see cross-sectional scanning tunnelling microscope measurements in Supplementary Note 3 as illustration), greatly diminishing the interfacial imperfections and reinforcing the dielectric strength<sup>6,25,33</sup>. Many ultrahigh- $\kappa$  dielectrics have bandgaps below 4 eV, which is considered to be not large enough for good insulators; however, as demonstrated in our experiments, single-crystal and transferrable SrTiO<sub>3</sub> performs well as a gate dielectric via vdW integration in the 2D transistors. We believe a clean vdW gap is an effective

tunnelling barrier that reduces the carrier tunnelling probability<sup>6,8</sup> and thus suppresses the gate leakage currents. Hence, proper integration of the vdW gap in devices seems to provide an opportunity to enhance device performance in short-channel FETs.

To examine the gate dielectric performance, a SrTiO<sub>3</sub> membrane with a thickness of 40 u.c. is transferred onto a SiO<sub>2</sub> substrate with pre-defined gate metals. A monolayer molybdenum disulfide (MoS<sub>2</sub>) thin film grown by chemical vapour deposition (CVD) is then transferred on top of the SrTiO<sub>3</sub>, and the quality of MoS<sub>2</sub> is verified by Raman and photoluminescence (PL) spectroscopies (Extended Data Fig. 4). Finally, the MoS<sub>2</sub> channels are patterned to form local back-gate MoS<sub>2</sub> FETs (Fig. 3a), and a photo of the FET arrays is shown in Fig. 3b. The cross-sectional structure of the FET is revealed by scanning transmission electron microscopy (STEM) and energy-dispersive X-ray spectroscopy (EDS) mapping (Fig. 3c). The transfer (drain current versus gate voltage,  $I_{\text{D}}-V_{\text{G}}$ ) and output (drain current versus drain voltage,  $I_{\text{D}}-V_{\text{D}}$ ) characteristics of a long-channel monolayer MoS<sub>2</sub> FET (channel width/channel length,  $W_{\text{CH}}/L_{\text{CH}} = 10 \mu\text{m}/3.5 \mu\text{m}$ ) are plotted in Fig. 3d and Fig. 3e, respectively. The drain current of transfer curves shows a steep increase at the subthreshold region with a subthreshold swing (SS) value of about 71.5 mV dec<sup>−1</sup> and low gate leakage current. Typical output characteristics also demonstrate promising current control and saturation. Besides, the extracted four-probe field-effect mobility is 39.7 cm<sup>2</sup> V<sup>−1</sup> s<sup>−1</sup>, which is comparable to the mobilities of FETs constructed with conventional gate dielectrics in the literature (Fig. 3f, Supplementary Table 1). Figure 3g further presents the  $I_{\text{D}}-V_{\text{G}}$  curves for 50 MoS<sub>2</sub> FETs, and Fig. 3h shows the correlation between the on/off current ratio and the SS value from these devices. A number of devices approach an on/off current ratio of 10<sup>7</sup>; meanwhile, the best SS values are close to 70 mV dec<sup>−1</sup>, which is among the best values





**Fig. 3 | Local back-gated MoS<sub>2</sub> FETs with ultrahigh- $\kappa$  SrTiO<sub>3</sub> dielectrics.**

**a**, Structure of the local back-gated FETs, where a clean vdW interface exists in between SrTiO<sub>3</sub> and MoS<sub>2</sub>. **b**, Optical micrograph of batch-fabricated FET arrays. Inset: a magnified image of the white square, where G, S, and D represent gate, source, and drain respectively. Scale bars, 100  $\mu$ m and 10  $\mu$ m (inset). **c**, Cross-sectional STEM image and corresponding EDS mapping obtained in the contact-to-channel area. ML: monolayer. The colours represent atoms as follows: blue, Pt; yellow, Au; cyan, Ti; red, S; green, Sr. Scale bar, 25 nm. **d**, Transfer characteristics ( $I_D$ - $V_G$ ) of monolayer MoS<sub>2</sub> FET, showing steep subthreshold slopes. **e**, Output characteristics ( $I_D$ - $V_D$ ) of the same device. **f**, Four-probe conductivity as a function of  $V_G$  for MoS<sub>2</sub> monolayer device with a

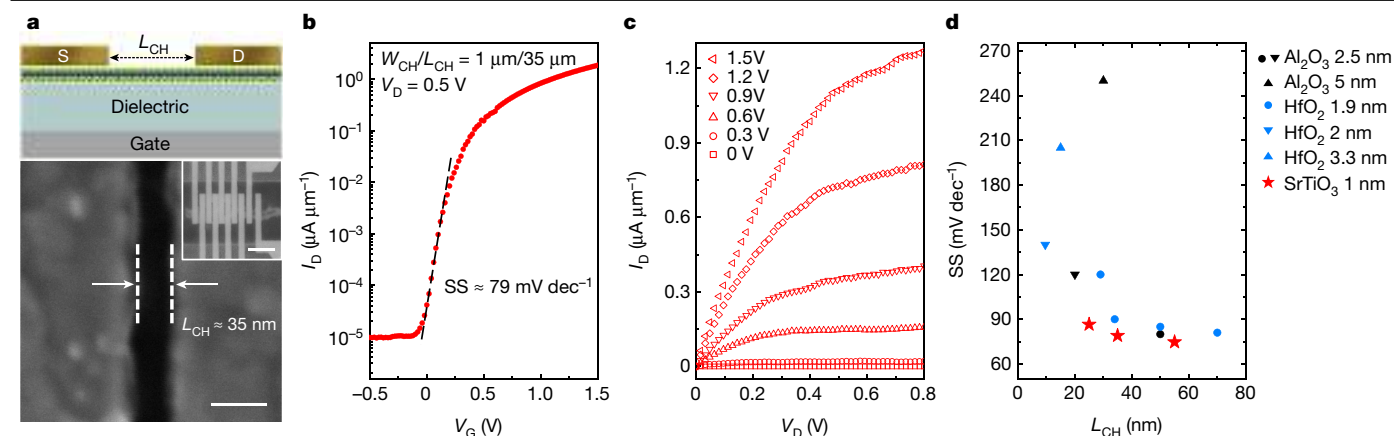
40-u.c.-thick SrTiO<sub>3</sub> gate dielectric.  $\mu_{FE}$ , field-effect mobility. Inset: the device structure. Scale bar, 5  $\mu$ m. **g**,  $I_D$ - $V_G$  curves of 50 SrTiO<sub>3</sub>/MoS<sub>2</sub> FETs. **h**, Scatter distribution (black) of recorded on/off current ratios and SS values, and statistical histogram (grey) of SS from 50 devices. **i**, Comparison of SS values achieved by state-of-the-art CVD-prepared MoS<sub>2</sub> FETs with the sub-10-nm CET<sup>8,34,36–38</sup> or EOT<sup>9,14,39</sup> technologies. The ball-and-stick symbol represents the EOT, where the ball is the theoretical value and the stick is the EOT range extracted from practical permittivity in general nanocapacitors. SS values were extracted within the  $V_D$  range of about 0.1–1 V and an  $L_{CH}$  range of about 0.1–5  $\mu$ m. The shaded red corner is the IRDS low-power specification for 2028.

attained by MoS<sub>2</sub> FETs. Figure 3i summarizes the SS values achieved by the reported CVD-grown MoS<sub>2</sub> long-channel FETs coupled with sub-10-nm CET high- $\kappa$  dielectrics, where the crystalline dielectrics generally exhibit better switching behaviours than the amorphous dielectrics (see Supplementary Table 2 for details). Note that some reports estimate the gate capacitance only by adopting ideal dielectric permittivity without performing  $C_{eff}$  measurements, which may underestimate the equivalent oxide thickness (EOT). Thus, we re-estimated these EOTs with the scope of both ideal and practical permittivities derived from the nanocapacitors to ensure an impartial judgement. Notably, the SrTiO<sub>3</sub> dielectric yields extremely low SS values that are among the lowest published results based on CVD-grown MoS<sub>2</sub>, and

already meet the microelectronic technology metrics 2028 (shaded red corner) projected by the International Roadmap for Devices and Systems (IRDS)<sup>5</sup>. To further probe the interface properties, the trap density ( $D_{it}$ ) at the SrTiO<sub>3</sub>/MoS<sub>2</sub> interface is estimated using the expression<sup>6</sup>:

$$SS = \ln(10) \frac{k_B T}{q} \left( 1 + \frac{q D_{it}}{C_G} \right)$$

where  $k_B$  is Boltzmann constant,  $T$  is absolute temperature,  $q$  is the elementary charge and  $C_G$  is the gate capacitance obtained from MOS capacitance measurements (Extended Data Fig. 5a). The extracted  $D_{it} \approx 4.3 \times 10^{12} \text{ cm}^{-2} \text{ eV}^{-1}$  is lower than the values typically obtained from



**Fig. 4 | Electrostatics of short-channel MoS<sub>2</sub> FETs based on ultrahigh- $\kappa$  SrTiO<sub>3</sub> dielectrics.** **a**, Schematic illustration of the structure and SEM image of the device with a channel length of 35 nm. Inset: an optical micrograph of the associated devices. Scale bars, 50 nm and 1  $\mu m$  (inset). **b**,  $I_D$ – $V_G$  characterization of the device shown in **a**. **c**,  $I_D$ – $V_D$  output curves of the same device.

CVD-grown MoS<sub>2</sub> channels (Supplementary Table 3), consistent with the low SS values from our devices. Also, the small hysteresis of the transfer curve is in agreement with the presence of limited border traps and interface states<sup>6</sup> (Supplementary Note 4). We anticipate that further optimization of the CVD growth of 2D materials, nanofabrication processes and interface engineering could lead to improvements in SS values and hysteresis. The as-fabricated devices show excellent stability under cyclic tests and maintain good switching after one month of storage (Extended Data Fig. 5b). In addition, the constant-voltage stress time-dependent dielectric breakdown analysis projects that the proposed devices could reach a 10-year lifetime under an operation voltage of < 2.5 V (Extended Data Fig. 6).

With the scaling of channel length, the drain-induced barrier lowering becomes pronounced; consequently, the electrostatic control of the gate degrades, leading to the higher subthreshold current and poor SS values. Such a phenomenon can be mitigated by reducing the CET<sup>4,34</sup>. Hence, we fabricated short-channel MoS<sub>2</sub> FETs ( $L_{CH} \approx 25$  nm to  $L_{CH} \approx 55$  nm; Fig. 4a, Extended Data Fig. 7) to demonstrate the gate-to-channel control with the ultrahigh- $\kappa$  SrTiO<sub>3</sub> dielectrics. The transfer characteristics of the 35-nm short-channel MoS<sub>2</sub> FET (Fig. 4b) show a near-10<sup>6</sup> on/off current ratio and a steep SS value of 79 mV dec $^{-1}$ , and the output curves (Fig. 4c) show promising current control and saturation. The results of the SS values for the reported short-channel FETs fabricated with different gate dielectrics are summarized in Fig. 4d (see Supplementary Table 2 for details), where the SS values achieved by the SrTiO<sub>3</sub> gate dielectrics are among the lowest reported for CVD-prepared MoS<sub>2</sub> FETs with similar device geometries. Despite these achievements, we are aware that there is an acknowledged permittivity range of gate dielectrics for further scaling MOSFETs, where the ‘proper’ range is estimated by considering the effects of both drain-induced barrier lowering and fringing-induced barrier lowering (FIBL)<sup>4</sup>. In a conventional Si MOSFET, the benefits of using a very high- $\kappa$  material to slim the CET are limited. As the fringing field originating from the drain penetrates into the channel through the physically thicker gate dielectrics, the source-to-channel potential barrier gets lowered, which degrades the subthreshold characteristics of MOSFETs. Hence, the accredited upper limit of the high- $\kappa$  thickness ( $t_{high-\kappa}$ ) for a sub-10-nm node Si transistor (towards sub-1-nm CET) is regarded as 7 nm, whereas the lower limit is 4 nm for preventing direct tunnelling. In consequence, a proper permittivity range can be determined by  $\epsilon_{SiO_2} \times t_{high-\kappa}$ , which approximates as 20 to 30. However, dissimilar to the conventional fabrication of 3D dielectrics, the vdW integration leads to the presence of a few-ångström-thick vdW gap between 2D semiconductors and

**d**, Benchmark SS values for reported short-channel ( $L_{CH} \leq 100$  nm) 2D FETs coupled with different sub-5-nm CET gate dielectrics<sup>37,40–43</sup>. For a fair comparison, only the devices with a CVD-prepared channel and single-gate geometry are shown.

dielectrics, which has an important role in suppressing FIBL. To elaborate this, we performed a technology computer-aided design (TCAD) simulation, where Extended Data Fig. 8a, b shows the simulated equipotential contours of a 10-nm short-channel MoS<sub>2</sub> FET without and with the involvement of a 3-Å vdW (vacuum) gap, respectively. The presence of the vdW gap redirects most of the fringing field lines through itself, which greatly restrains the drain-to-channel coupling. The calculated conduction band diagrams (Extended Data Fig. 8c) also imply that FIBL is apparently moderated in the SrTiO<sub>3</sub> dielectric with the vdW gap. Correspondingly, the simulated transfer characteristics reflect vast differences between the interfaces with and without the vdW gaps ranging from 3 Å to 5 Å, especially in the ultrahigh- $\kappa$  SrTiO<sub>3</sub> dielectric (Extended Data Fig. 8d, e). Note that the same approach is not applicable to conventional SiO<sub>2</sub> with a relatively low dielectric constant, as introducing a vdW gap with a comparable thickness level to 1-nm-thick SiO<sub>2</sub> (EOT = 1 nm) will largely degrade the overall gate capacitance. The simulation results reveal the concealed advantage of vdW integration of 2D semiconductors and further expand the selection criteria of high- $\kappa$  gate dielectrics for the ultra-scaling 2D electronics.

In brief, the crystalline surface and well defined vdW interfaces with 2D semiconductors exhibit scaling potential for future transistor technologies. As shown in Extended Data Fig. 9, the elastic and slim nature of perovskite membranes have also enabled the accomplishment of flexible and transparent electronics using MoS<sub>2</sub> as the transistor channel<sup>35</sup>. Meanwhile, considering the exquisitely controlled and scalable growth techniques already established in the perovskite oxide field<sup>17,22,23</sup>, the capability to freely integrate functional perovskite oxide membranes with 2D materials offers a route to laminate assembly for monolithic 3D integration<sup>1,2,33</sup>.

## Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at <https://doi.org/10.1038/s41586-022-04588-2>.

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## Methods

### Preparation of freestanding SrTiO<sub>3</sub> layer

The TiO<sub>2</sub>-terminated surface of single-crystalline SrTiO<sub>3</sub> (001) substrates was prepared by annealing the substrates at 1,000 °C in an oxygen atmosphere. The atomically sharp step and terraces of the substrates were examined by AFM. Both the Sr<sub>3</sub>Al<sub>2</sub>O<sub>6</sub> and SrTiO<sub>3</sub> films were grown by a pulsed-laser-deposition technique using a 248-nm krypton-fluoride excimer laser. The water-soluble Sr<sub>3</sub>Al<sub>2</sub>O<sub>6</sub> layer was deposited first on the substrate at 950 °C with an oxygen partial pressure ( $p_{O_2}$ ) of  $1 \times 10^{-4}$  torr. The four intensity oscillation periods indicated the epitaxial growth of a 1-u.c.-thick Sr<sub>3</sub>Al<sub>2</sub>O<sub>6</sub> layer (Extended Data Fig. 10)<sup>22</sup>. Subsequently, the epitaxial growth of a SrTiO<sub>3</sub> layer on the Sr<sub>3</sub>Al<sub>2</sub>O<sub>6</sub> thin film was conducted at 700 °C and  $p_{O_2} = 1 \times 10^{-4}$  torr. All the deposition was monitored by RHEED oscillations to control the as-deposited film thickness and ensure the film growth in a layer-by-layer mode. On the completion of epitaxy, the sample was naturally cooled to room temperature under  $p_{O_2} > 10^{-4}$  torr. A polydimethylsiloxane (PDMS) support was coated on the as-prepared sample and then placed into the deionized water at room temperature until the water-soluble Sr<sub>3</sub>Al<sub>2</sub>O<sub>6</sub> layer was fully dissolved. Afterwards, the PDMS-supported SrTiO<sub>3</sub> membrane was transferred on the target substrate, followed by peeling off the PDMS support. Furthermore, the post-oxygen-annealing (about 350–400 °C,  $p_{O_2} > 760$  torr, 3 h) was applied to prevent non-stoichiometry in the SrTiO<sub>3</sub> membrane before device fabrication.

### CVD synthesis and transfer of MoS<sub>2</sub>

Molybdenum trioxide (100 mg) was placed in a ceramic boat located at the heating zone centre of the main furnace. The sulfur powder was placed in a separate ceramic boat at the upper stream side maintained at 155 °C by a separate heating tap. The c-sapphire substrates for depositing MoS<sub>2</sub> were put next to the metal oxide boat at the downstream side, where the vapours of both precursors were carried to the targeting substrates by an argon flow (100 sccm, 50 torr). The centre heating zone was heated to about 700–800 °C at a ramping rate of 25 °C min<sup>-1</sup>. After reaching the growing temperature, the heating zone was kept isothermal for 15 min, and the furnace was then naturally cooled to room temperature. The transfer procedure follows the common polymethyl-methacrylate-assisted method.

### Material characterization

Crystal structure-related analyses, that is, XRD, XRR and RSM, were conducted using a Rigaku SmartLab thin-film XRD system with a rotating copper anode X-ray source. The surface morphologies and membrane thickness were examined on a commercial multifunction AFM instrument (Bruker Dimension Icon SPM). The Raman and PL spectra of MoS<sub>2</sub> were collected in a confocal Raman microscopy system (WITec alpha 300R) with a 532-nm laser. The Si peak at 520 cm<sup>-1</sup> was used as the reference for Raman calibration. X-ray photoelectron spectra and ultraviolet photoelectron spectra of SrTiO<sub>3</sub> and MoS<sub>2</sub> were obtained with a Thermo Scientific ESCALAB250i high-resolution X-ray Photoelectron Spectrometer (monochromatic Al K $\alpha$  line and He I source). A Lambda 1050 UV/Vis/NIR spectrophotometer was used to record the absorption spectra of the SrTiO<sub>3</sub> membrane. The cross-sectional STM measurement was conducted with an in situ cleaved Si (100)/SrTiO<sub>3</sub> sample under ultrahigh pressure ( $5 \times 10^{-11}$  torr) and a temperature of 4.3 K.

### TEM characterization

Plan-view dark-field TEM images and SAED patterns of freestanding SrTiO<sub>3</sub> membranes were acquired using a JEOL JEM-F200 S/TEM at 200 kV from a top-view sample suspended on a holey carbon grid. A cross-section lamella of the FETs was prepared with a focused-ion-beam system (FEI Nova Nanolab 200). A Pt protection layer was first deposited on top of a device, followed by milling the surrounding channel area

with the layered structure. Cross-section STEM images and related EDS mapping were recorded by the same STEM system at 200 kV.

### Device fabrication

For the MIM capacitors, the freestanding single-crystalline SrTiO<sub>3</sub> membranes were transferred onto Pt-coated Si substrates, followed by depositing 10-nm Ti and 90-nm Au electrodes with the shadow mask. FETs were framed on the pre-patterned Si/SiO<sub>2</sub> (300 nm) substrates or hard-coated polyester films with Pt gate electrodes. After transferring the SrTiO<sub>3</sub> membranes and MoS<sub>2</sub> layers, the source and drain areas were align-patterned accordingly. Subsequently, the channels were contacted by Au/Ti (90 nm/10 nm) or Au (35 nm) electrodes on top of MoS<sub>2</sub>. Note that all the metallization was done by an electron-beam evaporator under about  $10^{-7}$  torr. The submicrometre patterns were carried out using electron-beam lithography (Elionix ELS-F125), and the other large patterns were conducted by a direct-write lithographer (Heidelberg MLA100). The channel width and channel length ( $W_{CH}$  and  $L_{CH}$ ) are specified in the figures. Before the encapsulation, the devices were annealed at 150 °C in an argon environment for 30 min. Polymethyl methacrylate (molecular weight: 950K, dilution in anisole) was applied to encapsulate the devices (5,000 rpm, 30 s).

### Electrical measurements

Electrical characterization of the FETs was performed using a Keithley 4200 semiconductor parameter analyser and a Keithley 2450 source meter. Capacitance measurements were conducted using a Keithley 4210-CVU module and an Agilent E4980A LCR meter. All the devices were measured on a Cascade MPS150 probe station under ambient conditions with a shielded enclosure.

### TCAD simulations

For the simulations of the device characteristics, we used the Sentaurus TCAD software package, where the material parameters for MoS<sub>2</sub> and SrTiO<sub>3</sub> were extracted from refs.<sup>44–47</sup>. In the simulation, we considered a multivalley band structure model for the MoS<sub>2</sub> layer. In addition, we assumed that there was no bandgap narrowing because the device works at room temperature, and there was no intended doping. For the gate-stack leakage, we introduced the Fowler–Nordheim tunnelling model, the direct tunnelling model and the thermal emission model. Furthermore, to establish an ideal situation in the low-field direct-current simulation, no charge traps or fixed charges were considered in the simulation. This simplification is also helpful for improving the convergence of the simulation.

### Data availability

The data needed to evaluate the conclusions in this work are publicly available online at <https://doi.org/10.5281/zenodo.6245863>. Additional data related to this paper may be requested from the corresponding authors upon reasonable request.

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**Author contributions** S.L. and L.-J.L. supervised the project. J.-K.H. conceived and directed the project. J.Z. led W.W., N.Y. and Y.L. to perform the synthesis and characterization of the perovskite oxide heterostructures. J.-K.H. and Y.W. synthesized the MoS<sub>2</sub> monolayer. J.S., X.G. and L.H. carried out microscopy characterization. J.-K.H., C.-H.L. and T.W. performed the Raman and PL analyses. Z.-L.Y., B.-C.H. and Y.-P.C. the executed cross-sectional STM measurements. J.S. and J.-K.H. contributed to the device fabrication. J.-K.H. conducted electrical measurements and analyses of devices. Y.W. and L.-J.L. proposed the device model.

Z.W. contributed to the TCAD simulation. J.Y., D.W., V.T., K.K.-Z., X.Z. and L.Q. provided constructive opinions and suggestions. All the authors discussed and contributed to the results. J.-K.H., L.-J.L. and S.L. drafted the manuscript.

**Competing interests** J.-K.H., J.S., J.Z. and S.L. are co-inventors on a patent application (Australian provisional filing numbers 2021902514 and 2022900344) related to the research presented in this paper.

**Additional information**

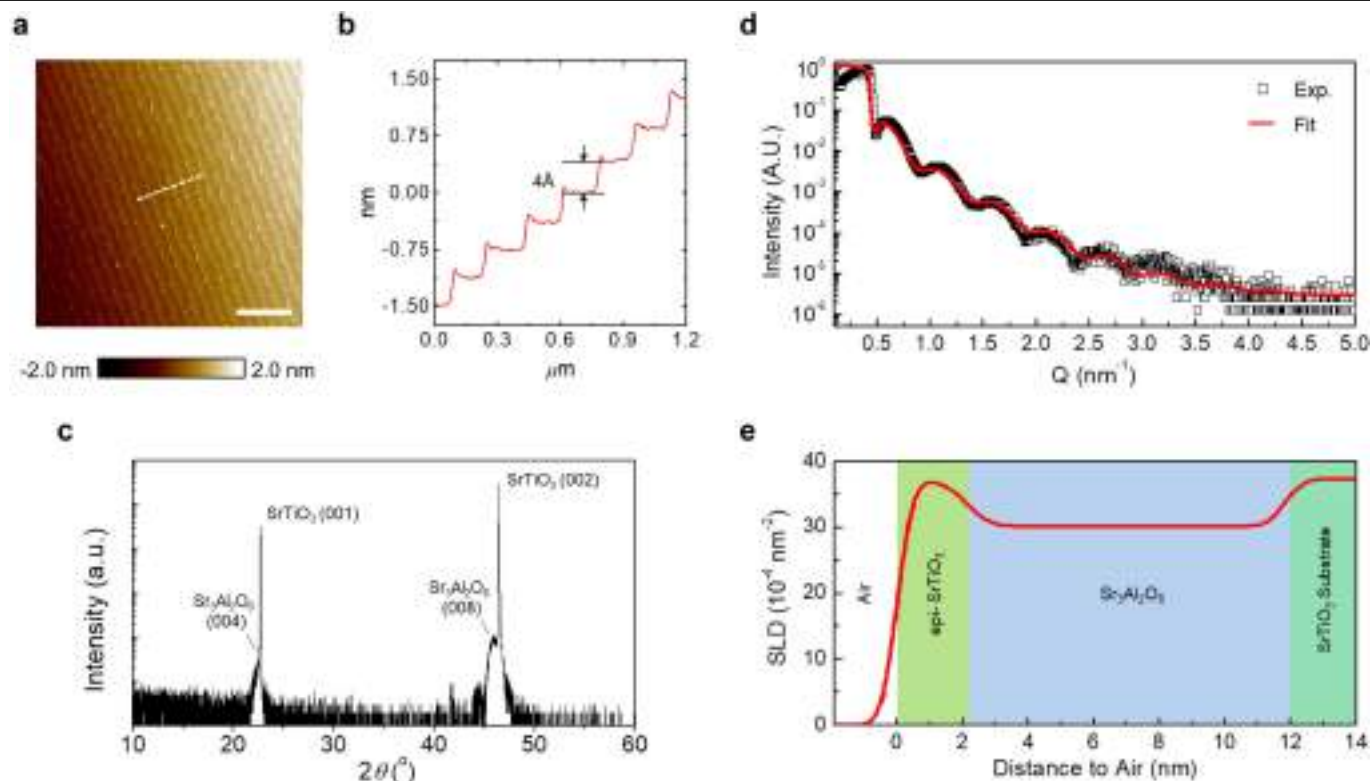
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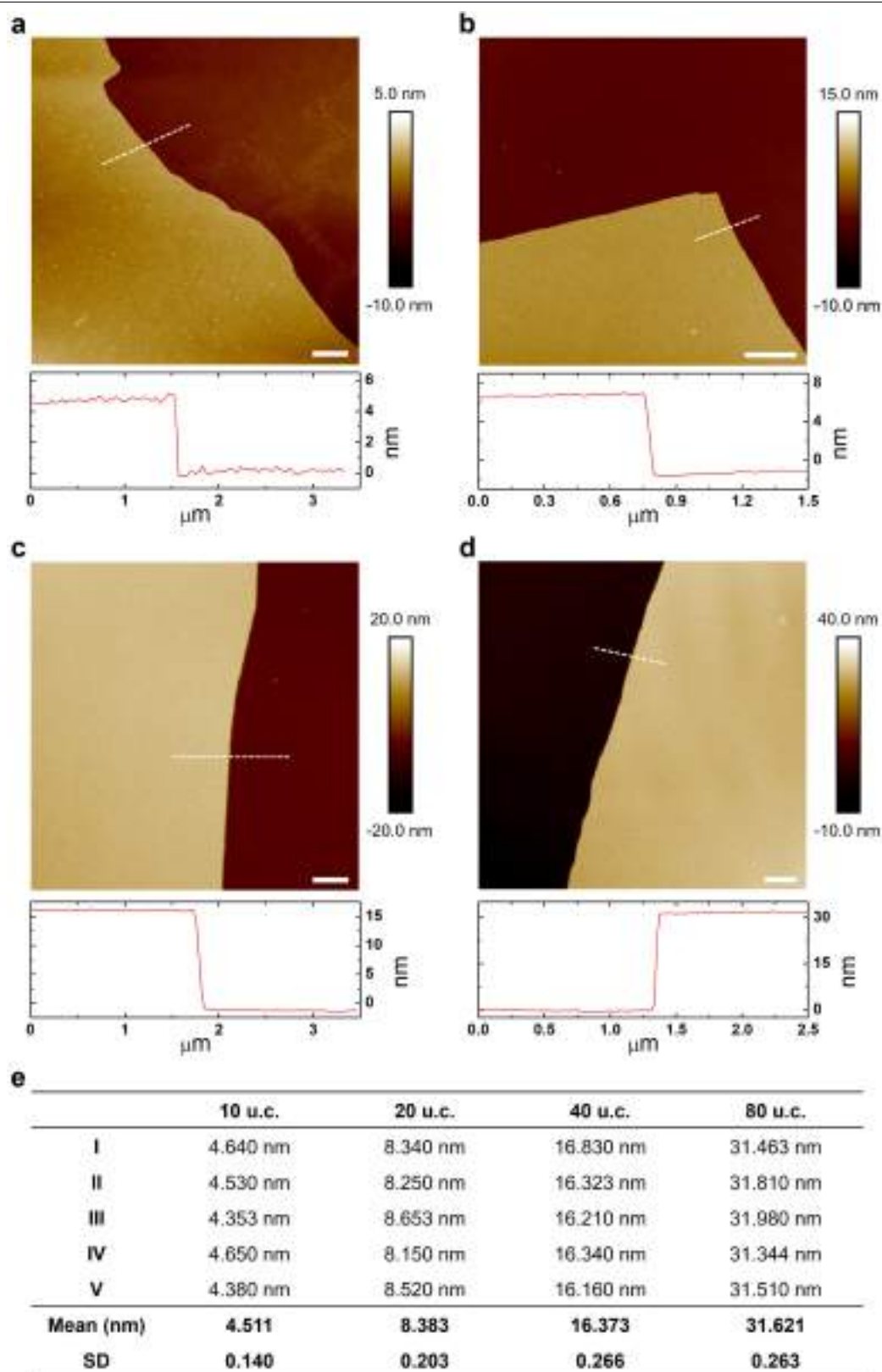
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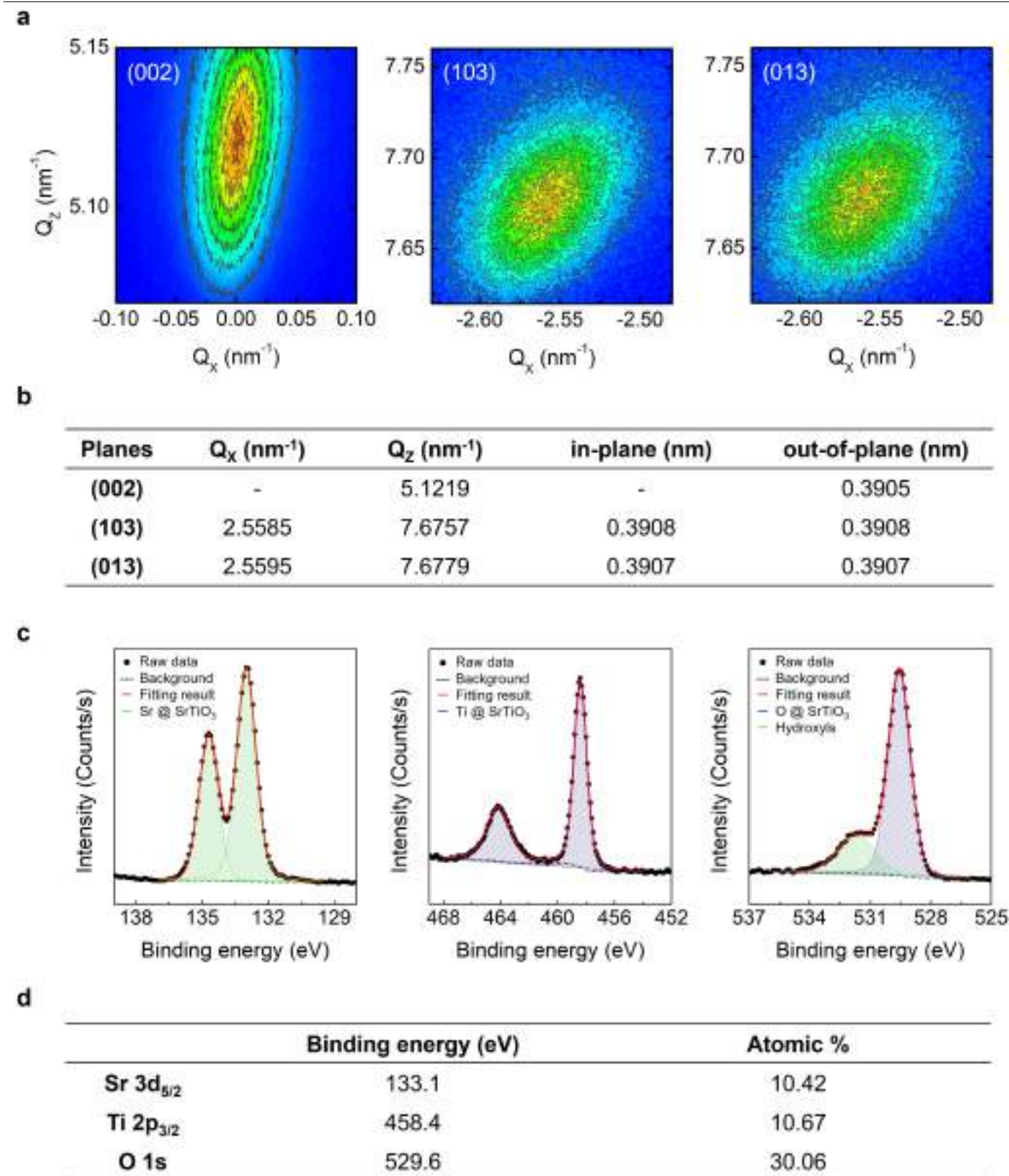
**Extended Data Fig. 1 | Crystalline structure of epitaxial membranes before release.** **a, b**, AFM topography and cross-sectional height profile of an as-grown  $\text{SrTiO}_3$  film indicating an atomically smooth surface with unit cell step terraces. The scale bar is 1  $\mu\text{m}$ . **c**,  $2\theta$ - $\omega$  X-ray diffraction the scan of  $\text{SrTiO}_3$ /

$\text{Sr}_3\text{Al}_2\text{O}_6$ /epi- $\text{SrTiO}_3$  heterostructure. **d**, The experimental and fitted XRR curves, where the fitting adequately describes the true structure ( $\text{SrTiO}_3$ /  $\text{Sr}_3\text{Al}_2\text{O}_6$ /epi- $\text{SrTiO}_3$ ). **e**, X-ray SLD model used to fit the experimental data. The model represents  $\text{SrTiO}_3$ / $\text{Sr}_3\text{Al}_2\text{O}_6$  (9.9 nm)/epi- $\text{SrTiO}_3$  (2.1 nm).



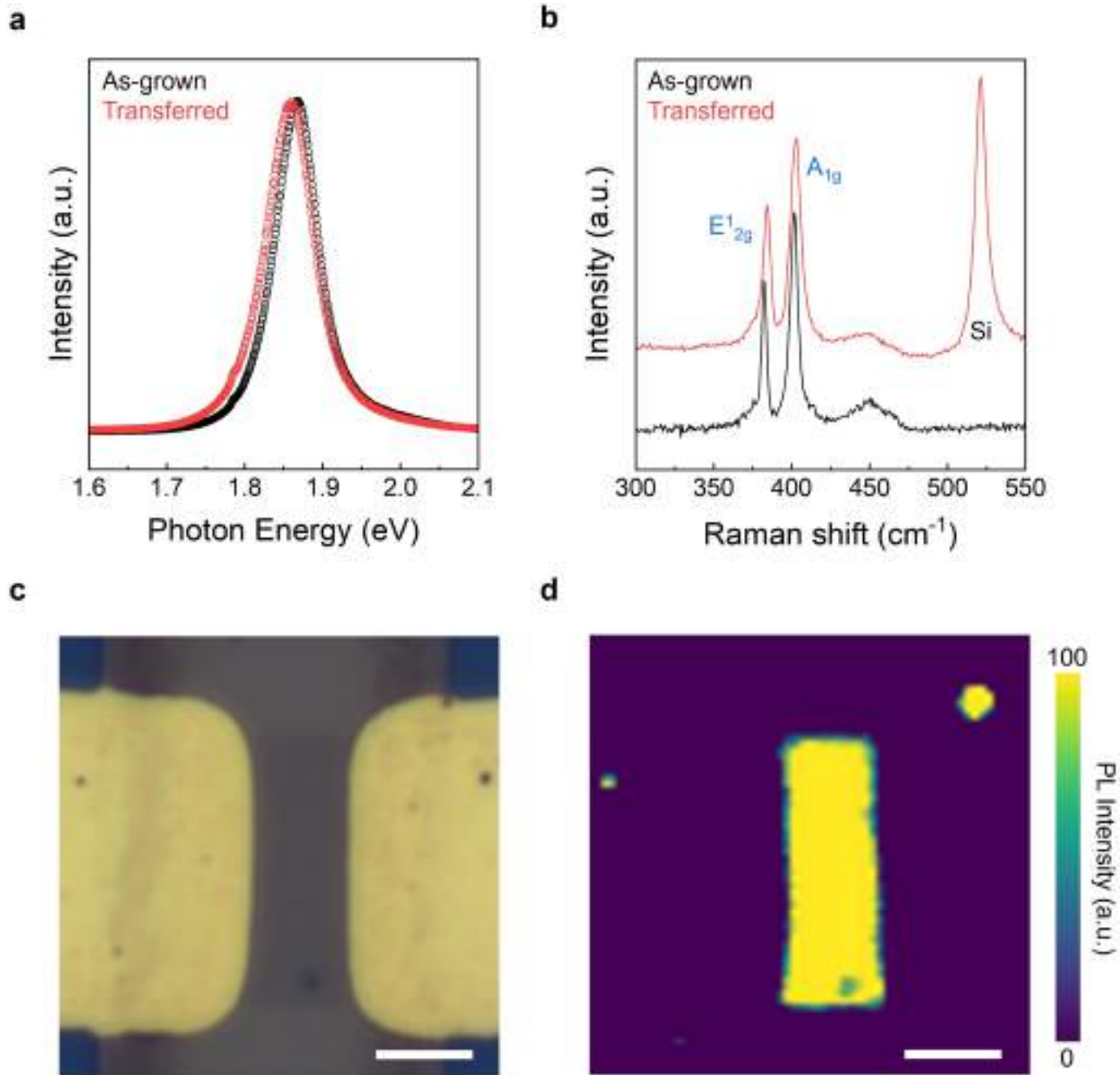
**Extended Data Fig. 2 | AFM measurements of transferred SrTiO<sub>3</sub> membranes. a-d**, 10 u.c., 20 u.c., 40 u.c., and 80 u.c. thick SrTiO<sub>3</sub> thin films and selected cross-sectional height profiles showing the thickness, respectively.

The scale bar indicates 1  $\mu\text{m}$ . **e**, Thicknesses extracted from various AFM measurements of the transferred thin film. Note that the theoretical thickness of one u.c. thick SrTiO<sub>3</sub> is 0.3905 nm (ref. <sup>23</sup>).



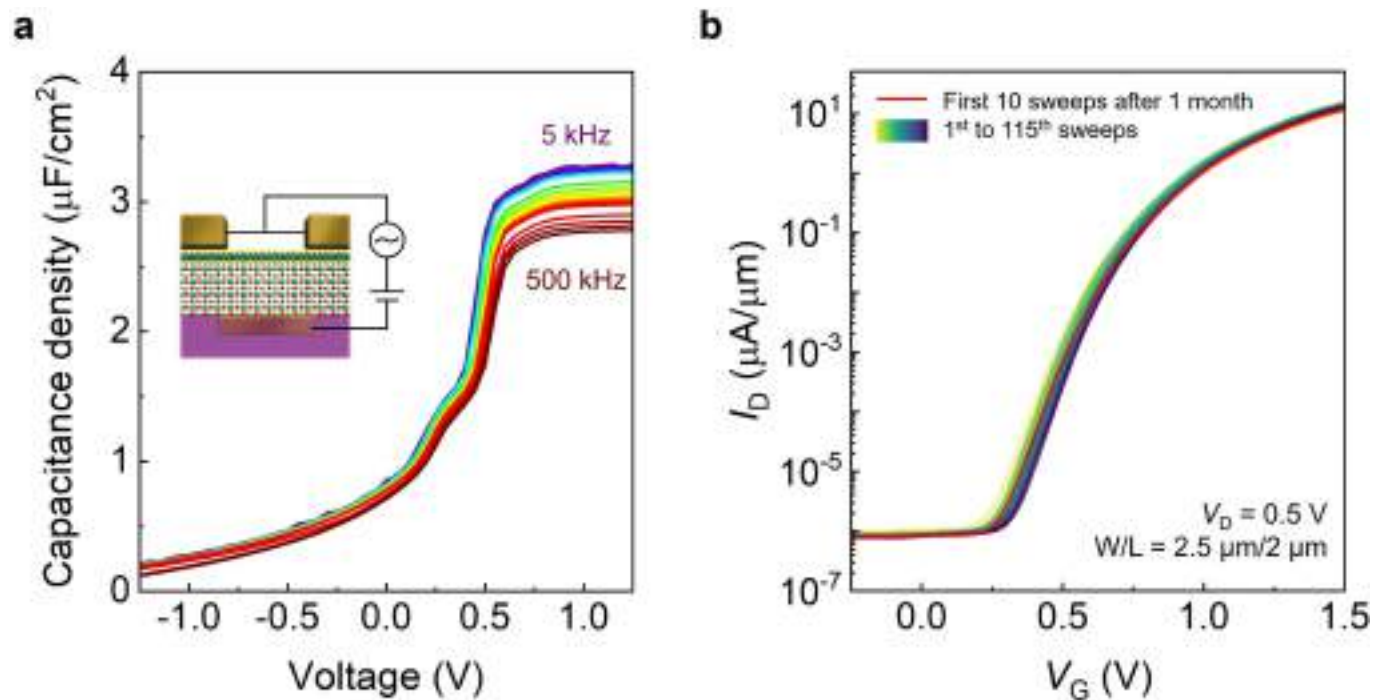
**Extended Data Fig. 3 | Structural and stoichiometric characterizations of transferred SrTiO<sub>3</sub> films. a**, RSM of transferred 80 u.c. SrTiO<sub>3</sub> film around (002), (103), and (013) planes. **b**, Relative values shown in (a) and extracted lattice parameters.  $Q_x$ ,  $Q_z$ : reciprocal space coordinates. **c**, High-resolution XPS spectra and analysis of the released SrTiO<sub>3</sub> film at Sr 3d, Ti 2p, and O 1s regions.

The fitted Ti 2p and Sr 3d spectra indicate that there is no second phase presenting in the membranes while the O 1s spectra were extracted to both intrinsic O in the SrTiO<sub>3</sub> and extrinsic O in hydroxyls absorbed on the surface<sup>48</sup>. **d**, Binding energy and stoichiometric analysis of XPS results<sup>49</sup>.



**Extended Data Fig. 4 | Characterization of MoS<sub>2</sub> monolayers.** **a**, PL spectra and **b**, Raman spectrum of the as-grown CVD MoS<sub>2</sub> monolayer before and after transferring on SrTiO<sub>3</sub>. Note the Raman frequency differences between  $E'_{2g}$  and

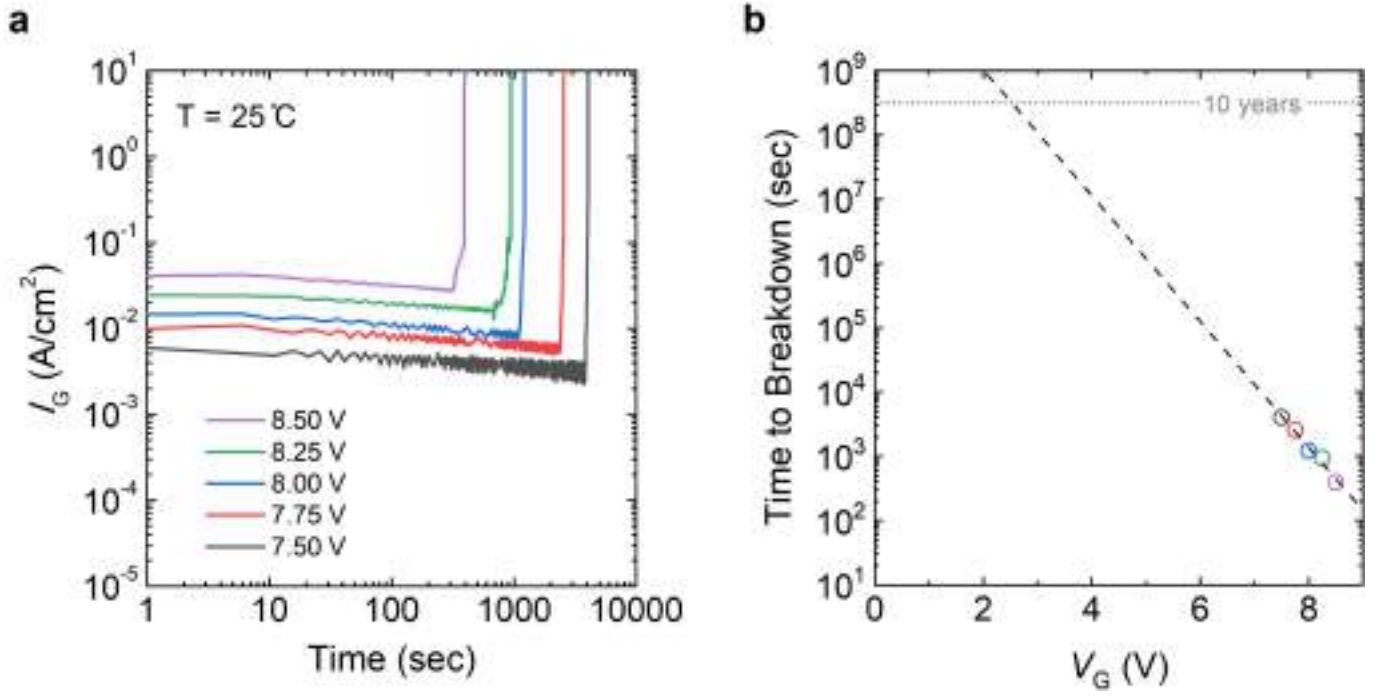
$A_{1g}$  are less than  $20 \text{ cm}^{-1}$ , indicating the monolayer nature<sup>50</sup>. **c**, Optical micrograph of the MoS<sub>2</sub> FET at channel region. **d**, PL intensity mapping of the corresponding area. The scale bars indicate  $4 \mu\text{m}$ .



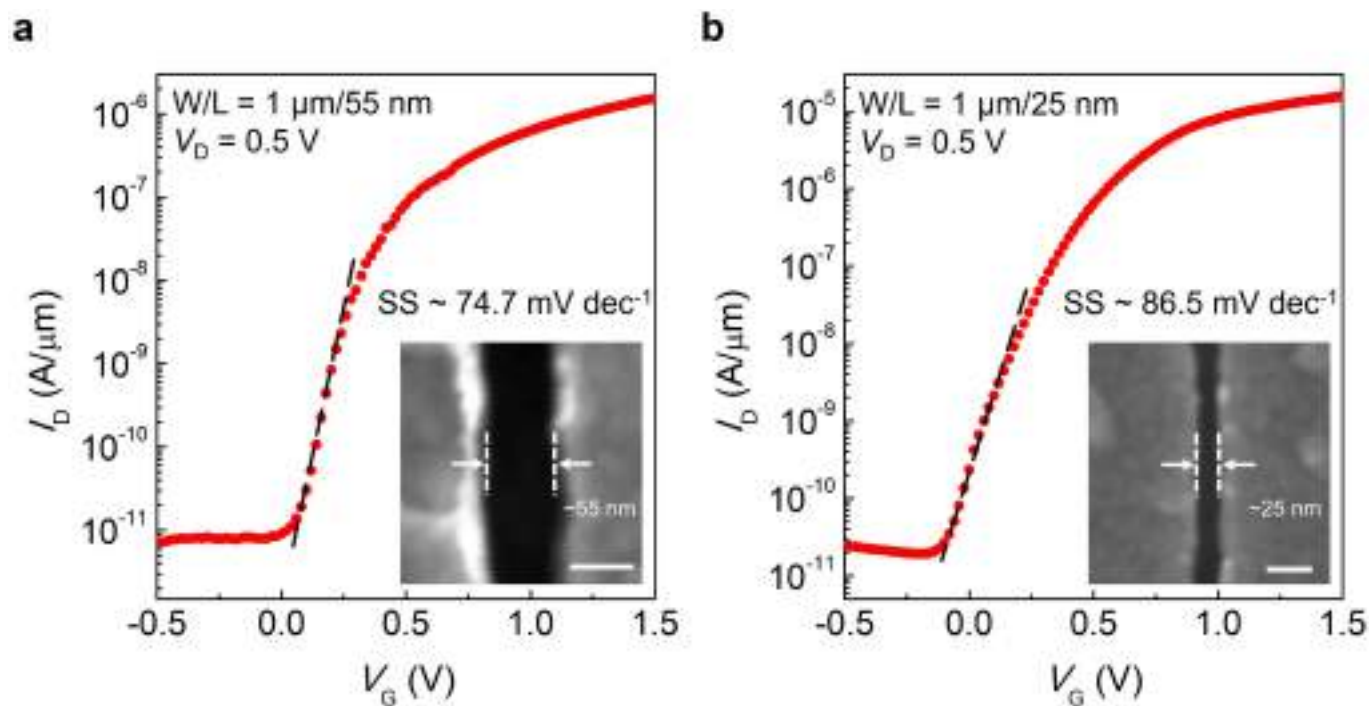
**Extended Data Fig. 5 | MOS capacitance and long-term stability. a,**  $\text{MoS}_2$  MOS capacitor measurements on 16.4 nm (40 u.c.) thick  $\text{SrTiO}_3$  layer. The accumulation capacitance of  $3.45 \mu\text{F}/\text{cm}^2$  corresponds to  $\sim 1 \text{ nm}$  CET. **b,** Transfer curves of  $\text{MoS}_2$  transistor with a 40 u.c. thick  $\text{SrTiO}_3$  gate dielectric. The

sequential color column displays the results of the device swept from 1<sup>st</sup> to 115<sup>th</sup> times, and the red lines show the results of the first 10 sweeps from the same device after one-month storage.

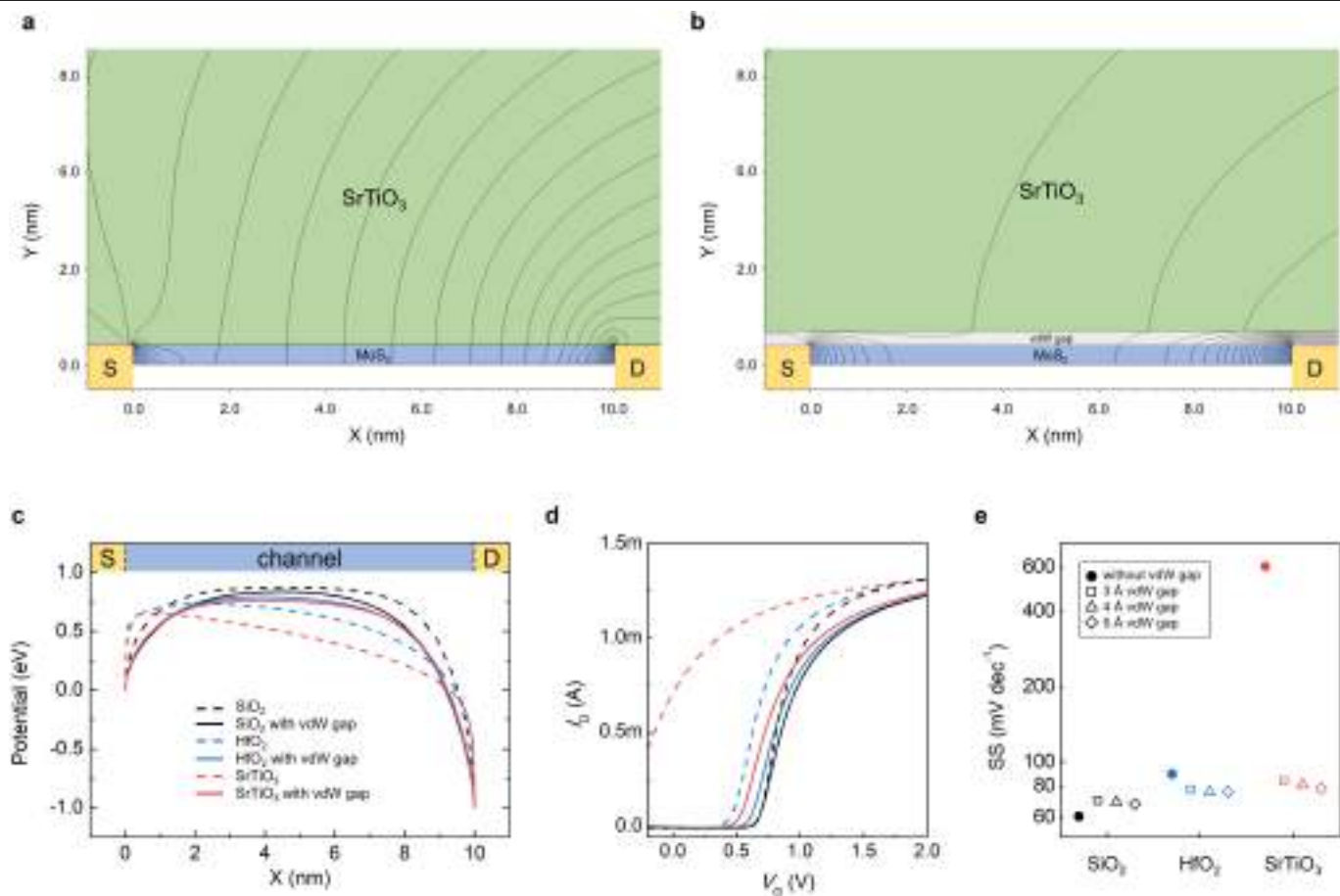




**Extended Data Fig. 6 | Lifetime projection.** **a**, Time to breakdown ( $T_{\text{BD}}$ ) of MOS devices with a 40 u.c. thick  $\text{SrTiO}_3$  dielectric at various voltage stresses. **b**,  $T_{\text{BD}}$  versus stress voltage characteristics, where the black dash-line represents the E model fitting for lifetime prediction.



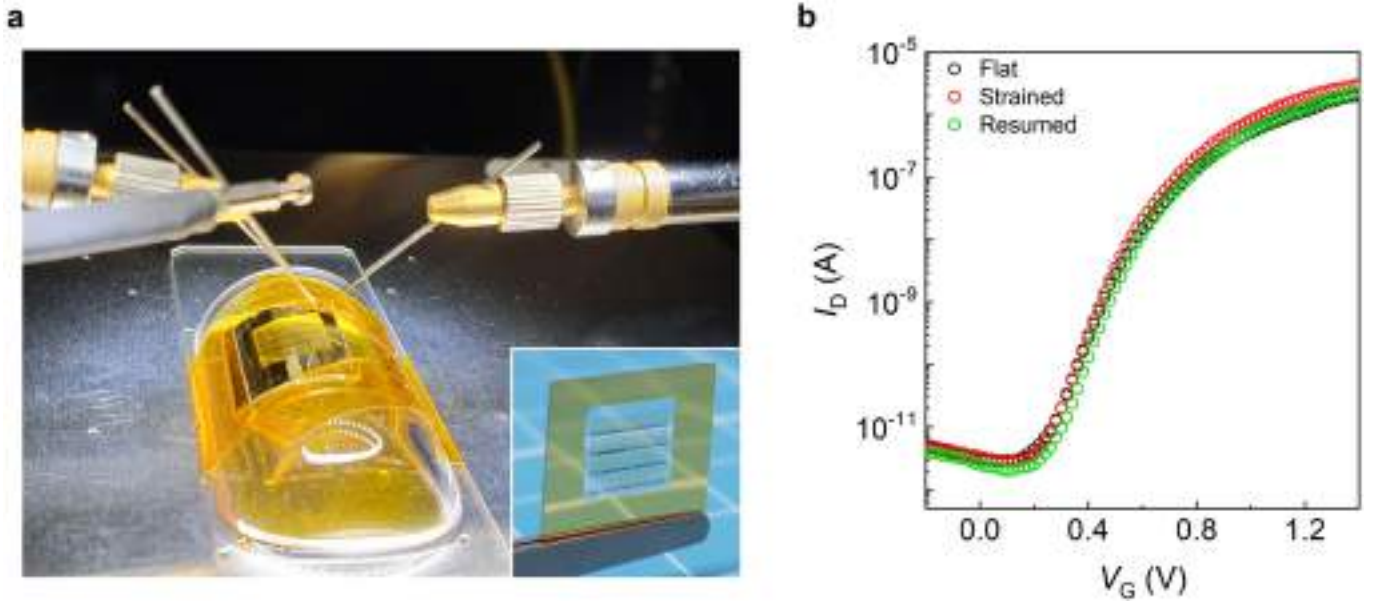
**Extended Data Fig. 7 | Short-channel performance.** **a, b**, Transfer characteristic of the short-channel device with  $L_{CH} = 55$  nm and 25 nm, respectively. The scale bars indicate 50 nm.



**Extended Data Fig. 8 | Impact of the vdW gap on moderation of FIBL effect.**

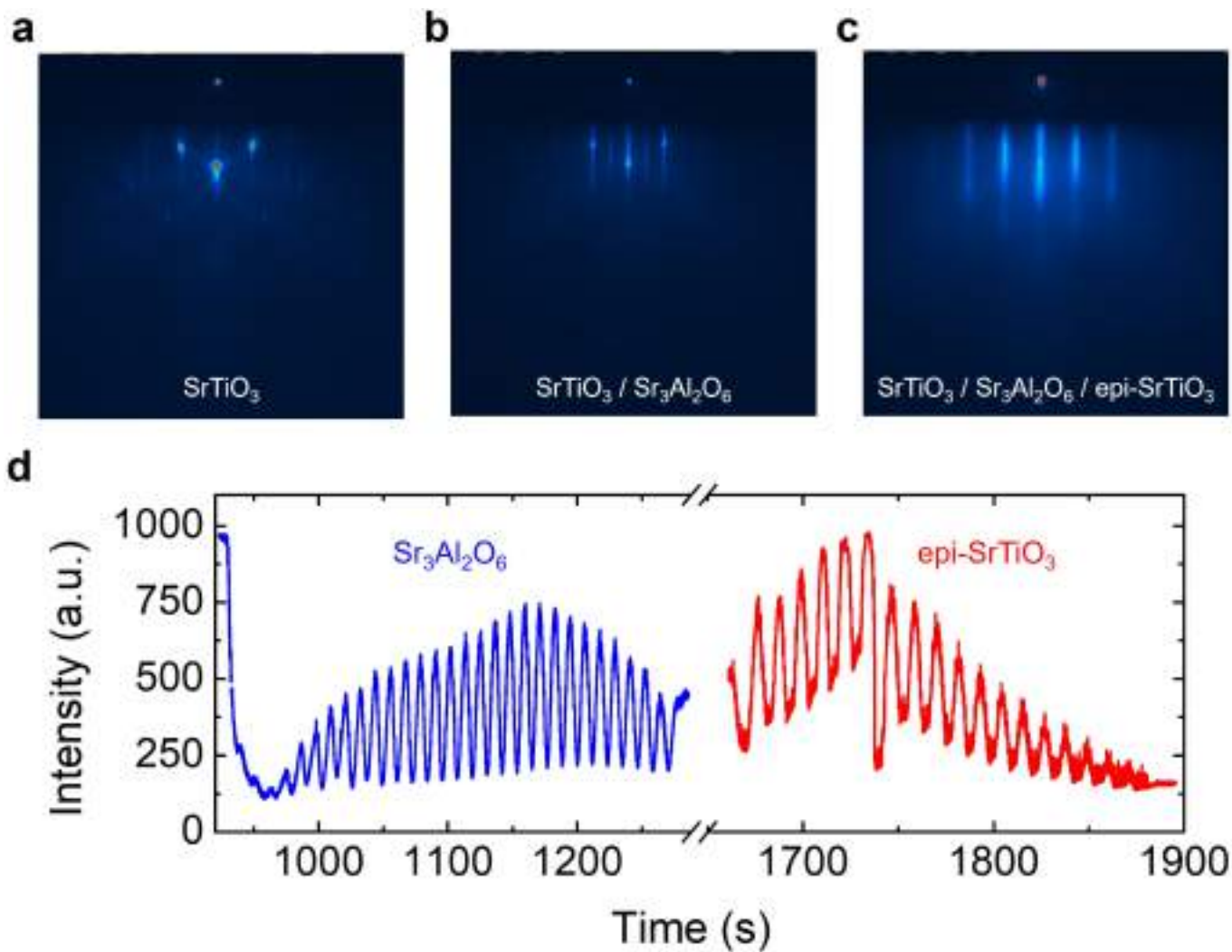
**a, b**, Simulated equipotential contours of short-channel (10 nm) MoS<sub>2</sub> FETs without vdW gap (a) and with 3 Å vdW gap (b), where source and gate are grounded,  $V_D = 1V$ , and SrTiO<sub>3</sub> dielectric is set as 1 nm EOT according to our experimental results. **c**, Calculated conduction band diagrams of

short-channel MoS<sub>2</sub> FETs with 1 nm EOT gate dielectrics of SiO<sub>2</sub> ( $\kappa = 3.9$ ), HfO<sub>2</sub> ( $\kappa = 22$ ), and SrTiO<sub>3</sub> ( $\kappa = 75$ ), where 3 Å vdW gap was used. **d**, Corresponding transfer characteristics of simulated short-channel MoS<sub>2</sub> FETs. **e**, Extracted SS values of the transfer characteristics with various vdW gap thicknesses.



**Extended Data Fig. 9 | Flexible and transparent MoS<sub>2</sub> FETs with SrTiO<sub>3</sub> gate dielectric. **a**, Photograph of the experiment setup for mechanical bending of MoS<sub>2</sub> FETs, where the samples are bent to a tensile strain of 0.54%. The strain at a given bending radius can be approximated by  $strain = d/2r$  where  $d$  is**

substrate thickness (0.125 mm) and  $r$  is the radius of curvature (11.5 mm)<sup>51</sup>. Inset is the photograph of MoS<sub>2</sub> FET arrays on flexible and transparent PET substrate. **b**, Measured transfer characteristics of the device under flat, strained, and resumed conditions.



**Extended Data Fig. 10 | Growth of freestanding  $\text{SrTiO}_3$  membranes.** **a-c**, Evolution of RHEED patterns during the preparation of  $\text{SrTiO}_3/\text{Sr}_3\text{Al}_2\text{O}_6/\text{epi-SrTiO}_3$  heterostructure. **d**, RHEED intensity oscillations in the deposition process accordingly, which indicate the typical layer-by-layer 2D growth mode.