Ledge-directed epitaxy of continuously self-aligned single-crystalline nanoribbons of transition metal dichalcogenides

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Two-dimensional (2D) transition metal dichalcogenides (TMDs) nanoribbons are touted as the future extreme device downscaling for advanced logic and memory devices but remain a formidable synthetic challenge. Here, we demonstrate a ledge-directed epitaxy (LDE) of dense arrays of continuous, self-aligned, monolayer and single-crystalline MoS₂ nanoribbons on β -gallium (III) oxide (β -Ga₂O₃) (100) substrates. LDE MoS₂ nanoribbons have spatial uniformity over a long-range and transport characteristics on par with those seen in exfoliated benchmarks. Prototype MoS₂ nanoribbon-based field-effect transistors exhibit high on/off ratios of 10⁸ and an averaged room temperature electron

mobility of $65 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$. The MoS_2 nanoribbons can be readily transferred to arbitrary substrates while the underlying $\beta\text{-Ga}_2\text{O}_3$ can be re-used after mechanical exfoliation. We further demonstrate LDE as a versatile epitaxy platform for the growth of p-type WSe_2 nanoribbons and lateral heterostructures made of p- WSe_2 and n- MoS_2 nanoribbons for futuristic electronics applications.

Planar transistors have been used for myriad generations with size and voltage scaling to enhance performance and save cost, following the well-known Moore's Law¹. Innovation of Fin-field effect transistor (Fin-FET) architecture was the solution and rendered the further device scaling possible. Unfortunately, short channel effect shall ultimately limit the Fin-FET scaling. A wave of revolutionary design in FET architecture with a superior gate control over the channel begins to take hold. This emerging stacked sheet architecture typically consists of multi-stacked semiconducting nanosheets with surrounding gate metals, which demonstrates better short-channel control and thus holds promise to extend the Moore's Law². Aligned arrays of single-crystal, monolayer 2D TMDs nanoribbons with high aspect-ratios, which represent the ultimate limit of miniaturization in the vertical dimension, are therefore very attractive in this context. Specifically, the ability to achieve single crystallinity and electrical uniformity throughout the entirety of the 2D TMDs nanoribbons, the key metrics of enabling batch production FET arrays, would allow a very high degree of electrostatic control at very low-power consumption. Synthetic strategies, toward TMDs nanoribbons have been reported to individually achieve control of layer number, single-crystallinity, self-alignment, and dimensionalities³⁻⁵. However, the dearth of a manufacturing route toward TMDs nanoribbons that synergistically combine all the aforementioned properties remains a major challenge.

It is known that the lattice orientation of 2D TMDs can be guided by substrates through lifting the energy degeneracy of the 2D TMD-substrate van der Waals (vdW) system⁶. We have further revealed that the lateral docking of 2D hexagonal boron nitride (*h*BN) seeds to the atomic step edges of Cu (111) substrates predominates over the vertical vdW registry of *h*BN on Cu, ensuring the mono-orientated nucleation and thus achieving the growth of single-crystal 2D *h*BN film⁷. These neat demonstrations of

synthesizing the uniform monolayer 2D TMD films with single crystallinity highlight that the selection of substrate (e.g., thermodynamics) and the growth parameter control (e.g., kinetics) are critically important. Here, we explore the epitaxial growth of single crystalline and aligned TMDs nanoribbons via LDE assisted chemical vapor deposition (CVD), that hinges on the thermodynamic control of TMD seeding orientation in conjunction with the kinetic control of growth direction. **Figure 1a** illustrates the LDE growth scheme for MoS₂ nanoribbons: (I) A single-crystal β -Ga₂O₃ (100) substrate with exposed ledges is used; (II) Nucleation of MoS₂ seeds with preferred orientation takes place at the ledges of β -Ga₂O₃; (III) Aligned MoS₂ domains merge into continuous nanoribbons; (IV) MoS₂ nanoribbons can be easily peeled off from the β -Ga₂O₃ (100) substrate and readily transferred to arbitrary substrates via a polydimethylsiloxane (PDMS)-assisted process; and (V) Exfoliated β -Ga₂O₃ substrate can be re-used for another round of growth.

Intrinsically, the (100) plane of the freshly exfoliated β -Ga₂O₃ substrate exhibits atomically sharp steps with a step height of ~ 6 Å (half unit cell). These steps trend up and down across the entire β -Ga₂O₃ substrate, resulting in two sets of structurally equivalent but crystallographically inverted ledges namely (001) and (-201)_{8,9} (**Fig 1b**). As featured in **Figs. 1c-e**, various stages in the growth of MoS₂ nanoribbons are revealed by atomic force microscopy (AFM). **Figure 1c** clearly suggests that unidirectional nucleation of four MoS₂ seeds carries out at the ledges. The edges of these triangular MoS₂ seeds stay parallel to the well-defined step edge whereas the vertices point toward the lower terrace. Meanwhile, we observed that the nucleation density of oriented MoS₂ seeds along both (001) and (-201) ledges is overwhelmingly higher than that of flat terraces where only sporadic distribution of randomly oriented MoS₂ flakes (orientation varies between 0°, 90°, 180° and 270° owing to the symmetry of β -Ga₂O₃ substrate, which is monoclinic in nature) can be spotted. The observation of unidirectional MoS₂ seeding flakes on the atomically-textured, single crystalline β -Ga₂O₃ (100) substrate clearly indicates the existence of an energetically minimized MoS₂— β -Ga₂O₃ ledge configuration, thus forming the basis for subsequent coalesce into continuous nanoribbons with single crystallinity. Indeed, aligned and mono-oriented MoS₂ seeds grow by successive addition from the surrounding precursors and

ultimately merge into a MoS₂ nanoribbon as LDE approaches completion (**Fig. 1e**). The resulting MoS₂ nanoribbons exhibit a uniform step height of ~ 8 Å, characteristic of monolayer MoS₂. Another unique capability of LDE is the controlled nucleation and unidirectional growth of ordered arrays of MoS₂ nanoribbons at the atomic scale in previously unachievable quantities and scales (up to centimeter long and aspect ratios > 5,000). Images of AFM and scanning electron microscopy (SEM) collectively demonstrated the growth of dense arrays of globally aligned, continuous MoS₂ nanoribbons enabled by LDE over the entire β -Ga₂O₃ (100) substrate (**Figs. 1f-g**).

In parallel, the innate step edges which are naturally present on the monolithic β -Ga₂O₃ (100) crystals¹⁰, have a propensity to cleave parallel to (100) and (001) planes by half unit cell. This is the result of unique octahedral arrangements of the Ga atoms (see **Supplementary Fig.1**) which are parallel to the (010) plane¹¹. Consequently, the newly exfoliated (100) plane of β -Ga₂O₃ retains atomically clean, ordered and spatially distributed step edges with half-unit cell ledges as shown in **Fig. 1b**. Photoluminescence (PL) measurement taken on different batches of MoS₂ nanoribbons grown on the repeatedly exfoliated β -Ga₂O₃ (100) substrate reveals neither changes in full width at half maximum (FWHM) nor the shift in PL peaks (see **Supplementary Fig. 2**), making it possible for the continuous and reliable batch production of high-quality MoS₂ nanoribbons. This peeling feature is particularly appealing as the ability to re-use β -Ga₂O₃ substrates eliminates the needs for a time-consuming and often laborious lithography process.

While all the aligned MoS₂ flakes interlock in the same way and have the identical orientation, the atomic structures of β -Ga₂O₃ (100) has a profound implication on the geometric shapes of edges of MoS₂ nanoribbons. Unlike the MoS₂ flakes grown on a symmetrical substrate, MoS₂ flakes grown on β -Ga₂O₃ (100) exhibit asymmetrically shaped edges, i.e., smooth and zigzag-shaped edges. Away from the well-defined ledges, the extremities of the merged MoS₂ flakes are permitted to grow without any external constraint. The one edge of the single crystalline MoS₂ nanoribbons assumes a regular zig-zag shape. High-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) Images in **Supplementary Fig. 3** confirm the zigzag-

shaped edges of the nanoribbons. Occasionally, we observed the formation of bilayer MoS₂ nanoribbons. High-resolution (HR) HAADF-STEM images near the edges of the bilayer regions reveal the absence of Moiré patterns, indicating predominantly 2H stacking orders. Moreover, by controlling the growth temperature and nucleation density, the width of the MoS₂ nanoribbons can be systematically varied between 70 nm to 600 nm (see **Supplementary Fig. 4**), for which the width likely can meet the requirement for stacked sheet transistor applications. Further decrease in width for which fundamental confinement effects may arise, such as changes in bandgap and presence of 1D metallicity (**Supplementary Discussion 1, and Fig. 5 and Fig. 6**), is possible experimentally. Yet, the LDE TMD nanoribbons may not be continuous in the millimeter to centimeter length scale.

To verify the orientation of individual flakes and the associated crystallinity of the MoS₂ nanoribbons, the LDE MoS₂ nanoribbons were characterized by second harmonic generation (SHG) micro-spectroscopy and dark-field (DF) STEM. It is known that polarization-resolved SHG is sensitive to the crystal orientation and the intensity profile map can be used as a descriptor for verifying spatial orientations of the merged flakes within the coalesced nanoribbons¹²⁻¹⁴. **Figure 2a** displays the SHG intensity map taken from three horizontally aligned MoS₂ nanoribbons with perpendicular polarization. All three MoS₂ nanoribbons demonstrate homogenous SHG intensities except for a few nodes along the direction of laser irradiation. The discontinuity of SHG intensity is the result of rarely observed multilayer MoS₂ seeds interspersed between the continuous MoS₂ nanoribbons by comparison of AFM images (see **Supplementary Fig. 3 and Fig. 7**). The homogeneity of the SHG intensity proves that each nanoribbon indeed comprises MoS₂ flakes with a single orientation. Furthermore, we deduced the angles between the laser polarization direction and the nearest armchair direction via the equation: $\theta = (1/3)\tan^{-1}\sqrt{I_x/I_y}$ ¹²⁻¹⁴. In this light, **Fig. 2b** features the intensity map that spatially resolves the angle distribution derived from compiling the simultaneously detected I_x and I_y SHG intensity, revealing uniform yet narrow angular distribution of $\sim 2^\circ$. The orientation of the zigzag direction is further confirmed by drawing comparisons of polarization resolved SHG intensity between the MoS₂ nanoribbons and the reflected

laser from the substrate. As indicated in the polar plot of **Fig. 2c**, MoS₂ flakes with mirror domains of 0° and 180° orientations are angularly equivalent in terms of SHG intensity. The SHG can help to characterize the nanoribbons in a large area but the further distinguishing such mirror domains requires other methodologies.

It is known that the variation in crystallographic orientations disturbs the structural continuity, i.e., the formation of grain boundaries. This disruption manifests signs of polycrystalline in annular dark-field (ADF)-STEM on the nanometer length scale¹⁵ as shown in **Fig. 2d** (see **Supplementary discussion 2**). Mirror domains of 0° (color in blue) and 180° (color in yellowish gold) can therefore be clearly determined on the basis of convergent beam electron diffraction patterns (**Fig. 2e**). ADF-STEM images shown in **Figs. 2f, g** confirm the absence of mirror domains and thus the existence of crystallographic continuity of our LDE MoS₂ nanoribbons on the micrometer length scale. This nanoribbon consists of more than twenty mono-oriented flakes and all ADF-STEM images exhibit crystallographically coherent domains with no visible grain boundaries, confirming the single-crystal nature of LDE MoS₂ nanoribbons. Other characterizations, including SEM image, corresponding PL mapping of the characteristic excitonic direct gap emission of monolayer MoS₂, and signatures from Raman spectroscopy, again prove the structure continuity and crystallographic coherence of chemical states of MoS₂ nanoribbons (**Supplementary Fig. 8**).

To understand the preferred nucleation at the ledge and the controlled growth along the step of β -Ga₂O₃, we conduct the cross-sectional HAADF HR-STEM (**Figs. 3a, b**) to provide the atomically resolved structures of both MoS₂ nanoribbon and the underlying β -Ga₂O₃ (100) substrate. Focus ion beam (FIB) was performed in the transverse direction of the MoS₂ nanoribbon (perpendicular to the [010] of β -Ga₂O₃). The atomic structures of MoS₂ nanoribbons are divided into three regions based on the locations: namely, (I) bottom terrace (left), (II) ledge (center), and (III) top terrace (right), respectively, allowing us to elucidate the relationship between epilayer and growth substrate. In agreement with the AFM image (**Fig. 1d**), region (II), the center segment of the nanoribbons where the nucleation of aligned, triangular seeds takes place, is found to lay above the (-201) ledge of β -Ga₂O₃ (**Fig. 3b**). This preferred alignment of triangular

seeds reveals that (-201) ledge may represent the preferential nucleation site with the local energetic minimum. With this assumption, we first examine the effect of preferred nucleation sites along the (-201) edges through constructing a cross-sectional atomic model for β -Ga₂O₃ (100). Here, the β -Ga₂O₃ (100) substrate has a monoclinic structure with lattice constants of $\mathbf{a} = 3.037 \text{ \AA}$, $\mathbf{b} = 5.798 \text{ \AA}$, and $\beta = 103.8^\circ$ ¹¹. Two possible nucleation cases are proposed and their binding energies are calculated: (1) case A, where a Ga atom is notably missing from the vicinal (-201) ledge (**Figs. 3b,c**); and (2) case B, whereas Ga atoms remain intact near the (-201) ledge (**Supplementary Fig. 9**). In case A, MoS₂ molecules with 0° and 180° orientations are used as nuclei and are intentionally placed in the vicinity of (-201) edges on β -Ga₂O₃ (100) as schematically represented in **Figs. 3c,d**. After relaxation, we find that MoS₂ molecule with 0° orientation predominately dock at the binding sites of (-201) edges.

Meanwhile, first-principles calculations revealed a drastic difference in binding energy of ~2 eV relative to that of inversely orientated MoS₂ molecule (180°), thus favoring the mono-oriented growth and therefore the unidirectional alignment. An opposite trend is observed in case B (**Supplementary Fig. 9**), but there exhibits an energy difference of only ~0.535 eV when the MoS₂ molecules dock to the oxygen at the bottom of (-201) ledge. Unlike case A where 0° is the preferred orientation, the preferred orientation in case B is 180° which will lead to mirror grain boundaries in the ribbons (see **Supplementary Discussion 3 and Fig. 10**). Certainly, the mono-oriented seeds in our nanoribbons are nucleated following the favorable nucleation case (case A) due to the fact that Ga vacancies are naturally present near the edge of the steps (see **Supplementary Fig. 11**).

The proposed mechanism toward unidirectional nucleation is similar to the recently reported defect-enhanced degeneracy-breaking of TMDs¹⁶, but is quite independent due to the difference in spatial arrangement of docking sites—randomly distributed and disorganized defect sites vs. spatially ordered and aligned ledge sites. Nevertheless, in this work, they observed the reversal of triangle orientation (i.e., 0° becomes 180°) of MoS₂ flakes across a step edge in the *h*BN substrate under the assumption of a change in layer polarity of AA'-stacked *h*BN. On the contrary, the two

energetically equivalent but crystallographically inverted ledges, (-201) vs. (001) revealed by DF-STEM and atomic models, across the step edges of β -Ga₂O₃ (100), thus guiding the alignment of MoS₂ nuclei in 0° and 180° orientations respectively (see **Supplementary Discussion 3 and Fig. 11**). Once the mono-oriented nucleation approaches completion, the rich sulfur (S) environment not only helps to break the vdW interaction between the aligned MoS₂ seeds and the ledges, but also facilitates the growth of single crystalline domains extended beyond both ends of the step edge, ultimately merging together into a continuous nanoribbon.

We note that growth of individual domains which strongly depends on the diffusion path, seems to be confined and directed along the ledges of β -Ga₂O₃ (100). This is very intriguing as the growth of TMDs on highly symmetric substrates by means of a CVD typically results in the omnidirectional diffusion of precursor vapors to the local environment. To verify the origin of this directional diffusion pathway, we performed a potential energy surface (PES) mapping of the (-201) plane of β -Ga₂O₃ (100) via density function theory (DFT) calculations. As shown in **Fig. 3e**, the surface diffusion kinetics along the [010] energetically confine the growth of MoS₂, thus driving the energetically favorable and directionally modulated growth of aligned domains into single crystalline nanoribbons. These findings collectively point toward an entirely novel strategy to synthesize dense arrays of single crystalline, and globally aligned TMD monolayer nanoribbons for device applications.

The success of creating extended, single-crystal MoS₂ nanoribbons is manifested in the uninterrupted, homogenous yet narrow distribution of signature PL wavelength across the aligned domains, indicating the lack of atomic misfits between merged domains as shown in **Fig. 4a**, and **Supplementary Fig. 12a**. Meanwhile, hyperspectral PL mapping, which provides a fast, global mapping with high spatial and spectral resolution, does not reveal any sign of PL quenching typically associated with grain boundaries. Results from conductive (C-) AFM on the MoS₂ nanoribbons directly grown on a semiconducting β -Ga₂O₃ substrate show the similar trend in the representative topography and corresponding current maps are shown in **Fig. 4b**. The local point current-voltage (I-Vs, vertical transport) and current mapping were done by

applying a positive bias to β -Ga₂O₃ substrate while the conductive tip (Pt-Ir) was held at ground (**Supplementary Figs. 12b,c**). The MoS₂ nanoribbons appear highly conducting relative to that of the underlying β -Ga₂O₃ substrate, making them clearly visible in the current map. The average current flowing throughout the MoS₂ nanoribbons in the vertical direction is 18 (± 2) nA. The point I-V curve measured along the MoS₂ nanoribbons exhibits non-ohmic characteristics that appear symmetric. These measurements provide direct experimental evidence of the undisruptive conductive path throughout the entirety of MoS₂ nanoribbons.

Furthermore, we verified the quality of MoS₂ nanoribbons by evaluating the field-effect carrier mobility in a bottom-gate transistor configuration as illustrated in **Fig. 4c** and **Supplementary 13a-d**. To reduce the screening effect from the HfO₂, while eliminating the charge scattering and trap sites, a single-crystal *h*BN monolayer film is embedded as an interface layer between HfO₂ and MoS₂ nanoribbons⁷. Noted that measurements were performed at room temperature. The electrical properties of our LDE MoS₂ nanoribbons have two important features: the spatial uniformity over a long range similar to those wafer-scale films synthesized by CVD/MOCVD and transport characteristics on par with those seen in exfoliated counterparts. Top panel of **Supplementary 13e** plots field-effect mobility and on/off ratios measured from five devices, fabricated on the same MoS₂ nanoribbon and separated by up to 20 μ m on a single chip. All five FETs exhibit nearly identical behaviors. These include an averaged field-effect mobility of 65 cm²/V-s and on/off ratios near $\sim 10^8$ independent of channel length and location of MoS₂ nanoribbons, suggesting the spatial homogeneity of the electrical properties of the MoS₂ nanoribbons across various length scales. Specifically, both values are comparable to the performance of the mechanically exfoliated benchmarks as shown in **Fig. 4d**¹⁷. In parallel, bottom panel of the **Supplementary 13e** provides the histogram of field-effect mobility and on/off ratios measured from 100 FETs made of different batches of MoS₂ nanoribbons. Evidently, single crystallinity throughout the entirety of MoS₂ nanoribbons is attested by the very narrow distributions of both field-effect mobility and on/off ratios. Occasionally, we find that field-effect mobility of MoS₂ nanoribbons FETs exceeds 100 cm²V⁻¹s⁻¹, with the highest value of 109 cm²V⁻¹s⁻¹¹⁸. The mobility is enhanced due to the synergistic effect between *h*BN layer and MoS₂

nanoribbons which are both single crystal in nature, ensuring the smooth charge transport along the heterointerface channel (**Supplementary Fig. 14**).

The location-selective hyperspectral PL included in **Supplementary Fig. 15a-c** revealed that both PL peak positions and FWHM did not vary significantly when the focus of laser spot was moved across the LDE MoS₂ nanoribbon, characteristic of the uniform quality and continuous crystallinity of LDE MoS₂ nanoribbons. In parallel, the low temperature PL (**Supplementary Figs. 15d,e**, excitation: 532 nm, power: 200 μ W) shows characteristics unique to exfoliated monolayer MoS₂ benchmarks, including comparable PL intensity, a similar level of defects, neutral exciton and trion emission peaks, further confirming the high quality of LDE MoS₂ nanoribbons. CVD grown-MoS₂ typically exhibits a high density of defects even though these specimens are characterized as high crystallinity. As a consequence, PL induced from defects of CVD-synthesized TMD emerges and outweighs the intrinsic PL at 4K unless treated chemically or doped electrostatically^{19,20}. The result is the impaired transport property and decreased mobility²¹. This finding of containing a low-level defect density and preserving the high level of single crystallinity simultaneously during the growth stage has not been reported or achieved elsewhere and thus distinguishes the LDE from the other epitaxy approaches.

Because our LDE growth is directed by the combination of ledge sites and surface diffusion limited pathway, intrinsic to the Ga₂O₃ substrates, its use is not limited to the MoS₂-Ga₂O₃ combination reported here. Instead, it could be generalized for producing various TMD nanoribbons, including n-(MoS₂), p-(WSe₂) and even lateral n-(MoS₂)-p(WSe₂)-n(MoS₂) junctions with precise single-crystallinity, alignment and monolayer controls over a micro-to-centimeter scale (**Supplementary Fig. 16**). While TMD nanoribbons with lateral heterostructures have been recently reported by a vapor-liquid-solid growth⁴, such a process only allows the growth of heterostructures with either different metals or chalcogen atoms, thus making it challenging for the creation of p-n heterostructures or even n-p-n multi-heterostructures. LDE WSe₂-MoS₂ lateral n-p-n multi-heterojunctions are achieved by growing WSe₂ nanoribbons first on β -Ga₂O₃ (100) substrates, followed by the edge epitaxy of MoS₂ nanoribbons on both sides as shown

in **Supplementary Fig. 17a**. Hyperspectral PL mapping of relevant PL characteristics, including MoS₂ in red, and WSe₂ in blue, in tandem with Raman and PL spectra, evidentially proves the successful in-plane growth of n-type MoS₂ at both edges of p-type WSe₂ (**Supplementary Fig. 17b-d**).

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Author statements:

A.A., L.J.L. and V.T. conceived the project. A.A., J.-H.F., Y.W., M.H., and R.A. performed the synthesis of TMDs nanoribbons and heterostructures, and carried out Raman, PL, and AFM characterizations. C.C.H., T.A.C., M.-Y.L., and J.-H.F. conducted fabrication of field-effect transistors and associated calculations. D.R.N., E.Y. and T.A. performed and analyzed C-AFM and Hyper PL spectra. S.B. synthesized and provided the single-crystal Cu (111) for *h*BN. S.-H.B. and J.K. transferred 2D TMD and heterostructures. C.P.C. and Z.C. performed the first-principles calculations. A.A., S.L. and J.-H.F. accomplished the DF-STEM, and cross-sectional HAADF-STEM. C.-J.L., W.-T.H. and W.-H.C. executed the SHG analysis. All of the authors discussed and contributed to the results. A.A., L.J.L. and V.T. wrote the paper.

Competing interests:

The authors declare no competing interests.

Methods:

Growth and Transfer of MoS₂ and WSe₂ nanoribbons. Single-crystal MoS₂ and WSe₂ monolayer nanoribbons were grown on the β -Ga₂O₃ (100) substrate by the conventional chemical vapor deposition (CVD) in a horizontal hot-wall 2" furnace tube with two heating zones. High purity of the S (Sigma-Aldrich, 99.99%), Se (Sigma-Aldrich, 99.99%), MoO₃ (Sigma-Aldrich, 99.9%) and WO₃ (Sigma-Aldrich, 99.9%) powders were used as the reaction precursors. MoO₃ (WO₃) powder typically was placed in a ceramic boat and was put in the heating zone centre of the furnace. S (Se) powder was placed in a separate quartz boat at the upper stream side maintained at 140°C (270°C) during the reaction. The single-crystal β -Ga₂O₃ (100) substrate was placed at the downstream side, where the precursor vapours were brought to the substrates by an argon (Ar) flowing gas at 30 torr for MoS₂ and Ar/hydrogen (H₂) mixture gas at 10 torr for WSe₂. The centre heating zone was heated to 800°C and kept for 10 min for the growth of MoS₂ nanoribbons. On the other hand, for the growth of WSe₂ nanoribbons, the furnace was heated to 900°C and was held for 15 min. Upon completion of growth, the furnace was naturally cooled down to room temperature.

Transfer of Monolayer MoS₂ nanoribbons. After the CVD growth, the resulting MoS₂ nanoribbons on β -Ga₂O₃ (100) were transferred onto a substrate of interest via a polydimethylsiloxane (PDMS)-assisted approach. In brief, a thin PDMS film was placed on top of MoS₂/ β -Ga₂O₃. Note that it is very critical to ensure conformal contact between PDMS and MoS₂/ β -Ga₂O₃. Next, the PDMS/MoS₂/ β -Ga₂O₃ stacked film was soaked in a 1M KOH for 5 min at room temperature, followed by rinsing the sample with a copious amount of deionized (DI) water, PDMS/MoS₂ stacked film was slowly peeled-off from β -Ga₂O₃ and then placed on a target substrate. The sample was kept in a vacuum for 30 min to make sure the adhesion between MoS₂ and the target substrate. Residual water droplets were dried under a constant nitrogen (N₂) flow. Finally, PDMS was peeled off, leaving behind the MoS₂ nanoribbons on the target substrate.

Characterizations. A FEI Quanta 600 scanning electron microscopy (SEM) was utilized to provide morphological views operating at 5 kV. Raman and photoluminescence (PL) spectra on MoS₂, WSe₂ nanoribbons and the lateral heterostructures of both were

collected using a Witec alpha 300 confocal Raman microscope equipped with a RayShield coupler. A 532-nm solid state laser as the excitation source. The excitation light with a power of 2.5 mW was focused onto the sample by a 100X objective lens (N.A. = 0.9). The signal was collected by the same objective lens, analyzed by a 0.75-m monochromator and detected by a liquid-nitrogen-cooled CCD camera. The atomic force microscopy (AFM) characterizations were conducted with Olympus (OMCLAC240TS) Al-coated silicon cantilevers. The resonance frequency was ~ 70 kHz, the spring constant was ~ 2 N/m, and the tip curvature radius was ~ 7 nm. The C-AFM measurements were conducted using Pt-Ir-coated conductive probes (SCM-PIT, Bruker) with a spring constant in the range of 0.5 to 4 N/m. Contact mode was utilized for imaging. A constant tip to sample bias of +3V was applied during all the C-AFM scans. AFM raw images were processed using a Gwydion 2.51 software. The cross-sectional scanning transmission electron microscopy (STEM) samples were prepared in a Helios NanoLab 660 DualBeam focused ion beam (FIB) system. Cross-sectional HR-STEM imaging was conducted using a Thermofisher USA (former FEI) Titan Themis Z transmission electron microscope (TEM) equipped with a double Cs (spherical aberration) corrector operating at 300 kV. Dark field (DF)-STEM with High-angle annular dark-field (HAADF) imaging was done in a scanning mode using an acceleration voltage of 80 kV with a column (at the sample) vacuum of about $2\text{--}4 \times 10^{-7}$ Torr at room temperature. Hyper-spectral photoluminescence (PL) measurements and mapping were taken with an IMA^{TM} hyperspectral microscope from Photon Inc. The samples were excited from a 532 nm laser with the intensity of $6.4 \mu\text{W}/\mu\text{m}^2$ and spectra were collected from an area of $90 \mu\text{m} \times 65 \mu\text{m}$ with 1 nm resolution. Exposure time was chosen as 120 sec for each wavelength.

Second Harmonic Generation (SHG) measurements. A home-built microscope arranged in the backscattering set-up was utilized to measure the SHG signals. Laser pulses generated from a mode-locked titanium (Ti)-sapphire laser with a wavelength of 850 nm, a pulse width of ~ 150 fs and a repetition rate of 80 MHz were used as the fundamental laser field. The polarization was selected by a linear polarizer and a half waveplate. The laser beam which illuminates normally onto the sample was focused by a 100X objective lens with a N.A. of 0.9. The generated SHG signals were collected by

the same objective and sent to a 0.75 m spectrometer equipped with a liquid N₂ cooled CCD camera. The scattered fundamental laser field was blocked by a 450 nm short pass filter. For the measurement of polarization resolved SHG images, the sample was mounted on a motorized x-y stage and scanned with a step size of 0.2 μm . The SHG signals were sent through a polarizing beam-splitter to separate the SHG intensity with polarizations that are parallel and perpendicular to the laser polarization. The two components were sent into the spectrometer and detected by the CCD camera simultaneously. For the polarization resolved SHG, the orientation of MoS₂ nanoribbons was aligned closely to the polarization direction of the excitation laser. The linear polarization of the excitation laser and the SHG signals were selected and analyzed by the combination of a half waveplate and a linear polarizer. The polarization of the generated signals was rotated by a half waveplate mounted on a motorized rotational stage with a step of 4° and analyzed by the polarizer.

First-principles Calculation. The first-principles calculations were carried out based on the density function theory (DFT) as implemented in the Vienna Ab-initio Simulation Package (VASP)²² within the MedeA²³. The exchange-correlation potential described by the PBE-GGA²⁴, and the van der Waals (vdW) correction vdW-DF (optB86b) functional²⁵ is used to calculate the binding energy difference between MoS₂ molecules and β -Ga₂O₃ substrates. The β -Ga₂O₃ has a monoclinic structure with lattice constants of $a = 3.037 \text{ \AA}$, $b = 5.798 \text{ \AA}$, and $\beta = 103.8^\circ$ ¹¹. A k-grid of $8 \times 1 \times 1$ and an energy cut-off 400 eV were used for the system of 5MoS₂/1 $\times$ 5 β -Ga₂O₃, with a vacuum of 20 $\text{\AA}$ along c and at least 10 $\text{\AA}$ along b between MoS₂ molecule and the other side of ledge in a periodic cell to eliminate spurious interaction.

Field-Effect Transistor Fabrication and Measurement. Monolayer MoS₂ nanoribbons grown on β -Ga₂O₃ (100) were transferred on 15 nm HfO₂, which was deposited on heavily doped silicon (Si) via the atomic layer deposition (ALD) as a gate insulator. A single crystalline hBN monolayer was detached from the Cu (111)/sapphire substrate by electrochemical delamination and then transferred onto HfO₂/Si via a combination of thermal release tape (TRT; #3195M) and poly (methyl methacrylate) (PMMA). The TRT can be released by annealing the TRT/PMMA/hBN/HfO₂/Si stacked films on a hotplate

at 180°C. The PMMA film was thoroughly removed via iteratively immersing the sample in a hot acetone bath for 40 min, leaving behind a *h*BN/HfO₂/Si stacked substrate. After transferring the MoS₂ nanoribbons, the resulting MoS₂ nanoribbons/*h*BN/HfO₂/Si were placed in a vacuum chamber under a pressure of 10⁻⁶ torr for 12h. Owing to the global alignment of LDE grown MoS₂ nanoribbons that provides far fewer constraints for the effective fabrication of field-effect transistors, electron-beam lithography emerges as the reliable method for producing the patterns of metal electrodes comprised of nickel (Ni, 20 nm) and gold (Au, 50 nm) necessary for electrical testing. More than one hundred single-nanoribbon field-effect transistors were reliably produced, and all tested to confirm the electrical output performance. This is due to the uniform, self-aligned, and tunable distribution of MoS₂ nanoribbons over the entire area of the β-Ga₂O₃ (100) substrate (~1 cm x 1.5 cm). All measurements were carried out under ambient conditions using a Keithley 4200 semiconductor analyzer.

Data availability:

The data from this study are available from the corresponding author on reasonable request.

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Fig. 1 | LDE growth of wafer-scale, globally aligned monolayer MoS_2 nanoribbons. **a**, Schematic illustration depicts the sequential growth of the monolayer MoS_2 nanoribbons along the intrinsically aligned ledges on $\beta\text{-Ga}_2\text{O}_3$ (100) substrate, which can be reused after a facile mechanical exfoliation. **b**, Computer generated crystal

structures provide a cross-sectional view of both (-201) and (001) ledges on the (100) β -Ga₂O₃. **c**, Height profile along the well-defined atomic step on pristine β -Ga₂O₃ substrate helps determine the step height of ~ 6 Å. **d**, Nucleation of unidirectional MoS₂ seeds along the ledge. **e**, A continuous MoS₂ nanoribbon with asymmetric edges. **f**, Dense arrays of aligned MoS₂ nanoribbons. **g**, SEM image showcases the continuous growth of globally aligned MoS₂ nanoribbons beyond millimeter-scale.

Fig. 2 | Polarization-resolved SHG measurement and DF-STEM characterization. **a**, The intensity map of the parallel component of SHG. The black arrow indicates the direction of incident laser polarization. **b**, Map of the angle (θ) between the direction of laser polarization and the armchair direction of the MoS₂ nanoribbons. **c**, Polar plot of the polarization-resolved SHG intensity and the backscattered laser light as a function of detection angles. **d**, (left) MoS₂ mirror domains of 0° and 180° orientations can be clearly juxtaposed with the differential diffraction filtered STEM mapping of domain orientations (right). **e**, Corresponding convergent beam diffraction patterns of the oppositely oriented domains in (d). **f**, ADF-STEM image together with (g) differential diffraction filtered STEM mapping of the MoS₂ nanoribbon highlighted in the red dotted rectangle collectively attests to the absence of grain boundaries which in turn confirms the single crystallinity of the LDE MoS₂ nanoribbons.

Fig. 3 | Atomically resolved imaging and proposed mechanism for LDE growth of MoS₂ nanoribbons on the ledge of β -Ga₂O₃ (100). **a**, Cross-sectional HAADF STEM image of a MoS₂ nanoribbon grown on the β -Ga₂O₃ (100) substrate. The insets provide atomically resolved views of different sections of the MoS₂ nanoribbons on: (I) bottom terrace (left); (II) ledge (center); and (III) top terrace (right), respectively. **b**, Cross-sectional HAADF HR-STEM image of β -Ga₂O₃ (100) substrate taken normal to the [010] direction reveals a missing Ga atom from the ledge. Computer-generated atomic models suggest two possible nucleation events on (-201) ledges with orientations toward **(c)** 0°, and **(d)** 180°, respectively. The presence of the ledge provides an energetically favorable docking site that breaks the energetic degeneracy by ~2 eV (0°), thus unidirectionally orienting the seeding flakes. **e**, Potential energy surface (PES) mapping derived from the DFT calculations sheds light on the diffusion limited pathway to direct the growth of aligned MoS₂ flakes into single-crystal nanoribbons.

Fig. 4 | Optical, electrical characterizations and batch fabrication of MoS₂ nanoribbon field-effect transistor. **a**, Hyper-spectral PL mapping displays a uniform wavelength distribution along the two parallelly aligned MoS₂ nanoribbons. **b**, Topological (top) and the corresponding current mapping images (bottom) show the spatially uniform current profile of a MoS₂ nanoribbon at a constant tip voltage. **c**, Schematic illustration shows the device architecture comprised of a monolayer MoS₂ nanoribbon stacked on top of a single crystalline hBN interface layer. **d**, Transfer characteristic of the MoS₂ nanoribbon/hBN field-effect transistor (FET, Inset shows the SEM of a MoS₂ nanoribbon sandwiched between source and drain electrodes); the length and width of the device are 1 μm and 0.39 μm, respectively, giving rise to an averaged electron mobility $\mu = 65 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ at a drain voltage V_{ds} of 0.5 V.