

Single-crystal rhombohedral boron nitride wafers for integrated sliding ferroelectric memory

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Two-dimensional rhombohedral boron nitride (rBN) has emerged as a promising sliding ferroelectric material for high-density, fast-speed and non-volatile memories. However, the batch production of ferroelectric rBN wafers has remained a critical challenge due to the thermodynamic instability of the rhombohedral phase and the inherent difficulties in maintaining consistent growth kinetics. Here we report a step-templated interfacial epitaxy strategy that enables the reproducible fabrication of 4-inch rBN wafers from nickel–boron films sputtered on the stepped sapphires. Comprehensive structural characterizations across multiple scales confirm the high phase purity and batch-to-batch consistent quality of produced rBN wafers. The intrinsic sliding ferroelectricity is verified in a unified ferroelectric field-effect transistor (FeFET), which exhibits ultrafast switching speed (at the nanosecond scale), high fatigue resistance ($>2 \times 10^9$ cycles) and long-term non-volatility (10 years). FeFET arrays are also constructed based on rBN wafers, featuring a high integration density with significant on/off ratios (10^6). Remarkably, the rBN FeFETs can maintain large memory windows (4 V) and excellent thermal stability (>470 K), even at an ultrashort channel length of 30 nm. These results establish rBN as a scalable platform for next-generation sliding ferroelectric memories, facilitating their applications in high-density memory chips and artificial intelligence accelerators.

Two-dimensional (2D) sliding ferroelectrics, covering all electronic components including metals, semiconductors as well as insulators, and featuring robust ferroelectricity persisting down to the nanometre scale^{1–5}, have recently triggered broad interest in high-density non-volatile memory technologies^{6–9}. The polarization of 2D sliding ferroelectrics, unlike that of the traditional ferroelectric materials, emerges and accumulates at the 2D interfaces^{10–12}. And the polarization switching is achieved through the relative in-plane sliding of adjacent

atomic layers^{13,14}, exhibiting the extremely high structural stability for robust storage capability^{15–17}. This unique mechanical–electrical coupling also enables the ultrafast switching dynamics (the speed calculated to be $1,000 \text{ m s}^{-1}$ in the AB-stacked boron nitride bilayers¹⁶) and the low power consumption (the sliding barrier estimated to be only a few milli-electronvolts per atom¹⁸). Additionally, 2D sliding ferroelectrics are naturally free of surface dangling bonds, making them highly compatible with on-chip integration processes.

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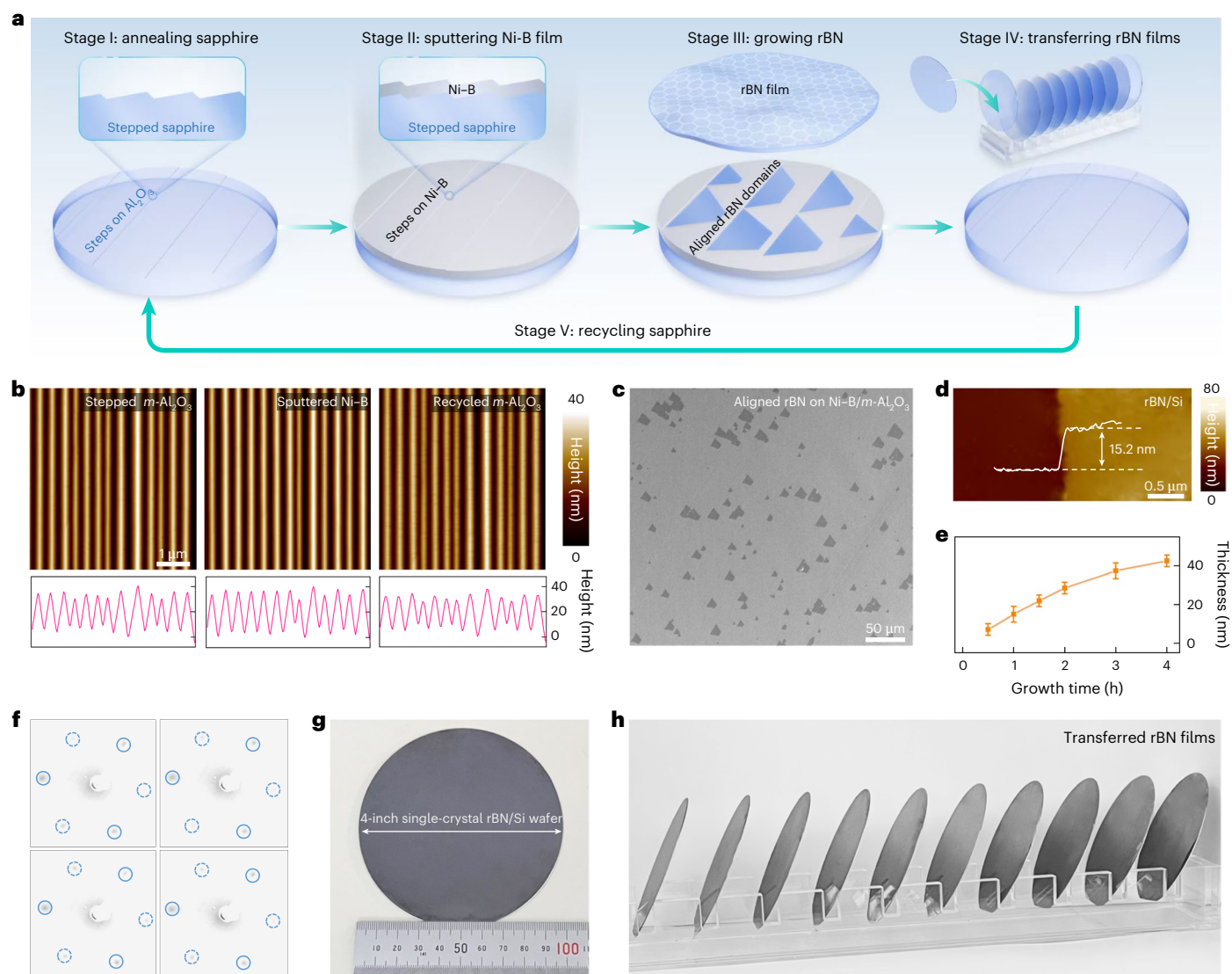


Fig. 1 | Step-templated interfacial epitaxy of 4-inch rBN wafers. **a**, Diagram of the batch growth process of single-crystal rBN wafers. **b**, Typical AFM topographic maps (top) and corresponding height profiles (bottom) of the original stepped *m*-plane Al_2O_3 , a sputtered Ni–B alloy and recycled *m*-plane Al_2O_3 , respectively. **c**, Representative scanning electron microscopy (SEM) image of the aligned rBN domains on the Ni–B/ Al_2O_3 surface. **d**, **e**, AFM height mapping

(**d**) and thickness evolution versus the growth time (**e**) of a rBN wafer. The thickness variation in **e** arises from ten different positions on the transferred rBN film. **f**, LEED patterns obtained at randomly selected areas of the rBN wafer. **g**, **h**, Photographs of an as-grown 4-inch rBN film transferred onto a Si wafer (**g**) and batches of transferred rBN wafers (**h**).

Within the 2D family, rhombohedral boron nitride (rBN) with a parallel ABC-stacking sequence serves as the outstanding sliding ferroelectric dielectric material. This is because rBN not only retains the full potential of AB-stacked BN bilayers with the record-high polarization switching performance^{16,19,20}, but also possesses the additional electrical insulation (large bandgap of ~6 eV and high breakdown strength of 10–20 MV cm^{−1}), thermal management ability (in-plane thermal conductivity of ~350 W m^{−1}K^{−1}) and extreme material stability (1,100 °C stable in air)^{21–24}. Therefore, a high-quality rBN single-crystal wafer is imperative for the integrated sliding ferroelectric memories; however, it remains elusive.

Currently, the batch production of rBN wafers for on-chip integrated memory technologies faces three essential challenges. (1) The growth of rBN crystals is thermodynamically unfavourable, as the formation energy of the rhombohedral phase is ~2 meV per atom greater than that of the stable hexagonal phase (hBN)²⁵. (2) Achieving high levels of within-wafer uniformity and batch-to-batch consistency requires strict control over the growth conditions,

which is extremely difficult to guarantee. (3) The viability for the large-scale integration of rBN ferroelectric field-effect transistors (FeFETs) remains unproven, particularly regarding the ultimate device performances.

Previously, several attempts have been made to regulate the in-plane lattice orientation and stabilize the interlayer rBN phase, such as through growth precursor regulation^{21,26}, surface lattice engineering^{27,28} and substrate step modulation^{18,29–31}. Among these methods, to realize the large-scale preparation of rBN, single-crystal metal substrates with controlled surface steps are typically needed. However, it is quite difficult to reproduce the precise treatments at wafer scale across multiple batches, and the challenges posed by the mass production of rBN single crystals persist. As a result, the sliding ferroelectricity of rBN layers has thus far been demonstrated at a proof-of-principle level. Practical applications necessitate the precise and consistent growth of rBN wafers, as well as the large-scale construction of ferroelectric devices without polarization pinning^{4,13}.

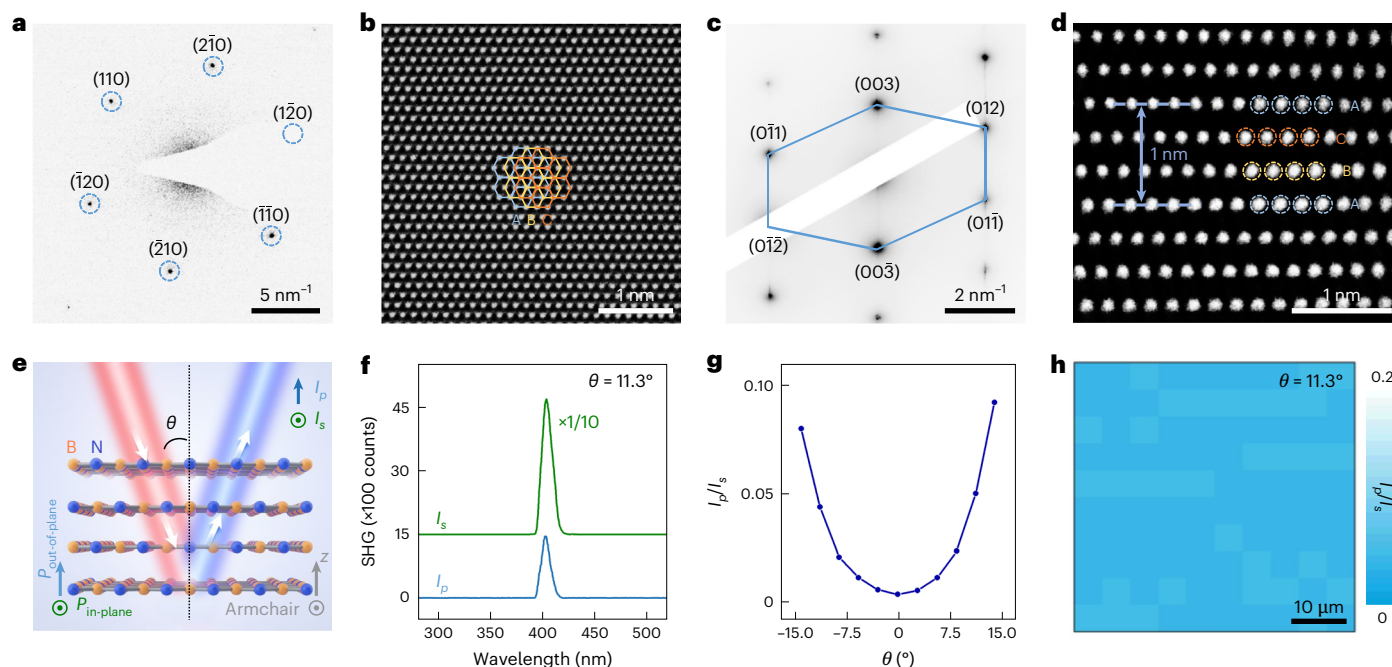


Fig. 2 | Structural characterizations of rBN wafers. **a**, Planar SAED pattern of the rBN wafer with the absent first-order diffraction spots. **b**, Characteristic planar HRTEM image of the rBN film. The blue, yellow and orange hexagons correspond to the different layer lattices. **c**, Cross-sectional SAED pattern of the rBN sample. **d**, Typical cross-sectional STEM image, showing the consistent sliding of multiple rBN layers. The blue, yellow and orange dashed circles indicate the B–N pairs in the different layers. **e**, Schematic of tilted-incidence SHG measurements.

The orange and blue spheres indicate B and N atoms, respectively. The initial polarization direction of the excited light was set along the in-plane zigzag direction of the rBN films. **f**, I_p and I_s intensities collected at the fixed tilted angle ($\theta = 11.3^\circ$) of the incident light. **g**, θ -dependent I_p/I_s ratio (**g**) and corresponding I_p/I_s mapping obtained with a fixed θ of 11.3° (**h**), demonstrating the uniform out-of-plane polarization of rBN at a large scale.

Batch production of crystalline rBN wafers

Here we propose a step-templated interfacial epitaxy strategy to realize the batch production of 4-inch rBN wafers. In our design, pre-treated *m*-plane sapphire (*m*-Al₂O₃) with parallel high steps is employed as the template (Fig. 1a, stage I). Then, a nickel–boron (Ni–B) film is uniformly sputtered to achieve the perfect conformal replication of surface steps (Fig. 1a, stage II, and Extended Data Fig. 1). As the N-terminated zigzag edges of BN layers preferentially attach to the solid-state step edges of metal substrates¹⁸, the parallel high steps could effectively ensure the unidirectional growth of BN domains (Extended Data Fig. 2a,b). Besides, the B source is supplied from the Ni–B film, thus the growth of each new layer takes place at the interface between the as-grown BN layers and the Ni–B substrate via the incessant nitrogen diffusion. This enables the Ni–B steps to continuously guide the unidirectional orientation of each BN layer and its parallel interlayer stacking order, ensuring the prepared samples with a consistent rhombohedral phase structure throughout the films (Fig. 1a, stage III, and Extended Data Fig. 2c,d). Moreover, the uniformly sputtered B source (Ni–B films) and homogeneously dispersed N source (N₂) guarantee a consistent feedstock supply³², facilitating the high uniformity across the 4-inch rBN wafers. Finally, the as-grown rBN films can be transferred onto arbitrary substrates (Fig. 1a, stage IV) and the stepped *m*-Al₂O₃ templates can be reused for the next batch (Fig. 1a, stage V).

The key of this growth strategy lies in the step structures of the sputtered Ni–B film/*m*-Al₂O₃, which could be readily verified through atomic force microscopy (AFM) (Fig. 1b and Methods). In a control experiment, when the stepped structure is absent or insufficiently defined (such as on *c*-plane or *a*-plane Al₂O₃ templates), the growth primarily tends to produce the hBN films (Extended Data Fig. 3a–h). Also, if the growth temperature is approaching the melting point of the sputtered Ni–B alloy film, the solid-state surface step structure vanishes, resulting in the formation of hBN layers (Extended Data Fig. 3i–k).

Utilizing the step-templated interfacial epitaxy method, unidirectionally aligned rBN domains were nucleated and grown at the initial

stage (Fig. 1c), then merged into a 4-inch wafer with a thickness of up to 45 nm (Fig. 1d,e and Extended Data Figs. 3l and 4a–c). The saturation of rBN thickness with the prolonged growth time is attributed to the depletion of boron content within the sputtered Ni–B film. Low-energy electron diffraction (LEED) patterns with typical alternating bright and dark points offered strong evidence of the single crystallinity of the prepared rBN wafers (Fig. 1f and Extended Data Fig. 4d). Given that the penetration depth of LEED is limited to a few atomic layers, X-ray diffraction (XRD) ϕ -scan measurements were also performed, and three main peaks at intervals of 120° confirmed the high crystallinity of rBN samples (Extended Data Fig. 4e). Furthermore, second harmonic generation (SHG) was used to reveal the stacking order of the thorough layers. The SHG signal of 15-nm-thick rBN layers was enhanced ~2,000-fold over that of monolayers, conforming the quadratic scaling of the SHG intensity with the number of layers (Extended Data Fig. 4f). The homogenous SHG intensity mapping and the identical polarized SHG patterns affirmed a uniform in-plane lattice configuration of the as-prepared rBN single crystals at a large scale (Extended Data Fig. 4g,h). By sequentially reusing the same stepped template, batch production of 4-inch rBN wafers with high crystal quality was thereby realized (Fig. 1g,h).

Characterizations of single-crystal rBN layers

Combined characterizations of AFM, ultraviolet–visible absorption spectroscopy (UV–vis), X-ray photoelectron spectroscopy (XPS), Raman spectroscopy and SHG were conducted on the different batches to verify the high crystal quality and the batch-to-batch consistency of the rBN wafers (Extended Data Fig. 5a–f). In addition, the superior crystalline quality of the rBN batches was also reflected by an exceptional laser-induced damage threshold of up to 490 GW cm^{−2} (Extended Data Fig. 5g), a large electric breakdown field of 10 MV cm^{−1} (Extended Data Fig. 5h,i) and a high in-plane thermal conductivity of 320 W m^{−1}K^{−1} (Extended Data Fig. 5j,k).

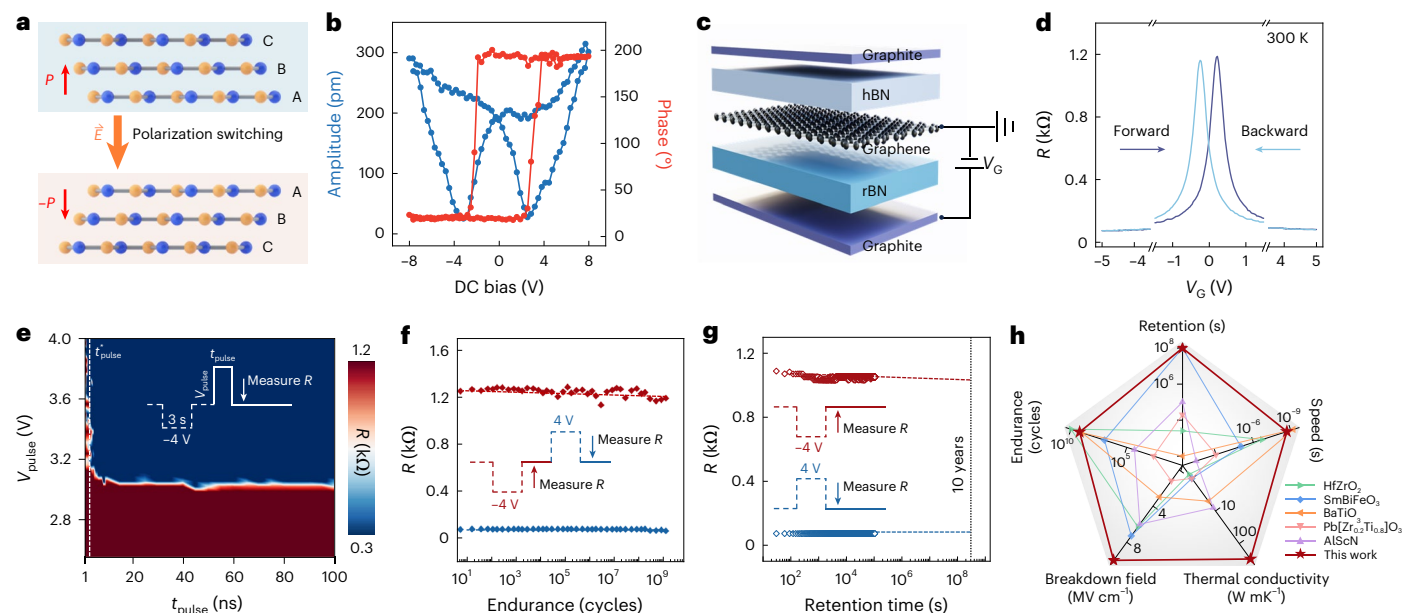


Fig. 3 | Sliding ferroelectric properties of rBN layers. **a**, Schematic of the polarization switching of rBN layers induced by the external electric field. **b**, Typical PFM signal of the rBN wafers, where blue and red data correspond to the amplitude and phase results, respectively. **c**, Diagram of a rBN/graphene FeFET device, in which the rBN multilayer film is used as the dielectric layer and the monolayer graphene is used as the channel layer. **d**, Resistance of graphene versus the gate voltage at room temperature. The dark and light curves

correspond to forward and backward sweeps, respectively. **e**, Resistance state reversal of graphene by implementing the voltage pulses. The minimum switching pulse width (t_{pulse}^*) was evaluated to be at the nanosecond timescale. **f, g**, Endurance (**f**) and retention (**g**) measurements of the unified rBN FeFETs, verifying the high fatigue resistance and non-volatility of the rBN samples. **h**, Comparison between rBN and other reported ferroelectric dielectrics, demonstrating the overall advantage of rBN sliding ferroelectric materials^{35–39}.

Furthermore, the interlayer stacking order of the grown rBN wafers was explicitly deciphered through an atomic-scale characterization scheme. The absence of first-order diffraction spots in the planar selected-area electron diffraction (SAED) pattern (Fig. 2a), combined with the inclined cross-sectional SAED pattern (Fig. 2c), illustrated a strong signature of the rhombohedral stacking structure (schematics of atomic arrangements and simulated SAED results of hBN and rBN shown as Extended Data Fig. 6a–d)³³. Besides, typical planar high-resolution transmission electron microscopy (HRTEM) images revealed an equal-contrast triangular lattice (Fig. 2b and Extended Data Fig. 6e), whereas cross-sectional scanning transmission electron microscopy (STEM) captured the unidirectional sliding of adjacent layers (Fig. 2d and Extended Data Fig. 6f). These results definitively corroborated the parallel stacking configuration and a congruently aligned B–N atom arrangement of the rhombohedral phase.

Owing to the difference in electronegativity between B and N atoms, the B–N pairs would give rise to electric dipole moments. Macroscopically, all bonded B–N pairs along the armchair direction create an in-plane polarization ($\mathbf{P}_{\parallel}$), whereas the aligned B–N pairs along the z direction generate an out-of-plane polarization ($\mathbf{P}_{\perp}$) in the rBN film (Extended Data Fig. 6b). In contrast, both in-plane and out-of-plane dipole moments will be cancelled at each interface in the hBN case, meaning that neither $\mathbf{P}_{\parallel}$ nor $\mathbf{P}_{\perp}$ are observed in hBN crystals (Extended Data Fig. 6a). To evaluate the polarization distribution of rBN, we conducted tilted-incidence SHG measurements (Fig. 2e)³⁴, where the $\mathbf{P}_{\parallel}$ and $\mathbf{P}_{\perp}$ induce s -polarized (I_s) and p -polarized (I_p) signals, respectively. In our experiment, I_p and I_s were synchronously collected under various tilted angles (θ) (Fig. 2f and Extended Data Fig. 6g, h) and the I_p/I_s ratio was extracted to exclude the collection efficiency errors. When $\theta = 0^\circ$, $\mathbf{P}_{\perp}$ was perpendicular to the incident field; thus, the I_p was nearly zero. As θ increased, due to the potentiated vertical component of the incident field, I_p/I_s increased, affirming the present $\mathbf{P}_{\perp}$ of rBN layers (Fig. 2g). And the uniform I_p/I_s mapping obtained at the fixed θ of 11.3° further revealed that the as-grown rBN wafer possessed the consistent $\mathbf{P}_{\perp}$ at a large scale (Fig. 2h and Extended Data Fig. 6i, j).

Ferroelectric properties of rBN crystals

In principle, the $\mathbf{P}_{\perp}$ of rBN can be switched through the external-field-induced interlayer sliding (Fig. 3a). We constructed the metal-ferroelectric-metal (MFM) capacitance devices, demonstrating the sliding ferroelectricity of rBN layers via polarization-electric field (P - E) loops with the observable hysteresis behaviour and corresponding current density-electric field (J - E) curves with distinct switching current peaks (Extended Data Fig. 7). The piezo-response force microscopy (PFM) measurements on the rBN films also exhibited the ferroelectric characteristics, featuring the consonant butterfly loops of the amplitude and hysteresis loops of the phase (Fig. 3b and Extended Data Fig. 8a). Furthermore, cross-sectional STEM images collected at the reversely poled regions indicated that the polarization switching behaviour corresponded to the $ABC \leftrightarrow CBA$ stacking transformation in the rBN multilayers (Extended Data Fig. 8b–e).

Hereafter, we fabricated a FeFET device using rBN flakes as the dielectric layer and graphene monolayers as the channel layer to investigate the device-level ferroelectric properties (Fig. 3c and Extended Data Fig. 9a). The forward and backward sweeps of the resistance (R) of the graphene sensor versus the gate voltage (V_G) displayed noticeable hysteresis at room temperature (Fig. 3d), confirming the robust polarization switching of rBN layers (Extended Data Fig. 9b). Then the switching speed was evaluated by implementing the pulse voltage (V_{pulse}) with different pulse widths (t_{pulse}) and monitoring the transition behaviour of the resistance state under t_{pulse} ranging from 1 ns to 10 μ s (Fig. 3e and Extended Data Fig. 9c). Our results revealed that the t_{pulse} required for switching could be shortened with increasing V_{pulse} , and the minimum pulse width (t_{pulse}^*) was estimated to be at the nanosecond timescale. This switching speed reaches the detection limit of our setup (Extended Data Fig. 9d), and is almost the fastest result achievable in sliding ferroelectrics.

After the switching speed measurement, the endurance and retention characteristics were sequentially tested on the same rBN/graphene FeFET device to evaluate its fatigue resistance and non-volatility. Periodic bipolar pulses of ± 4 V were applied to

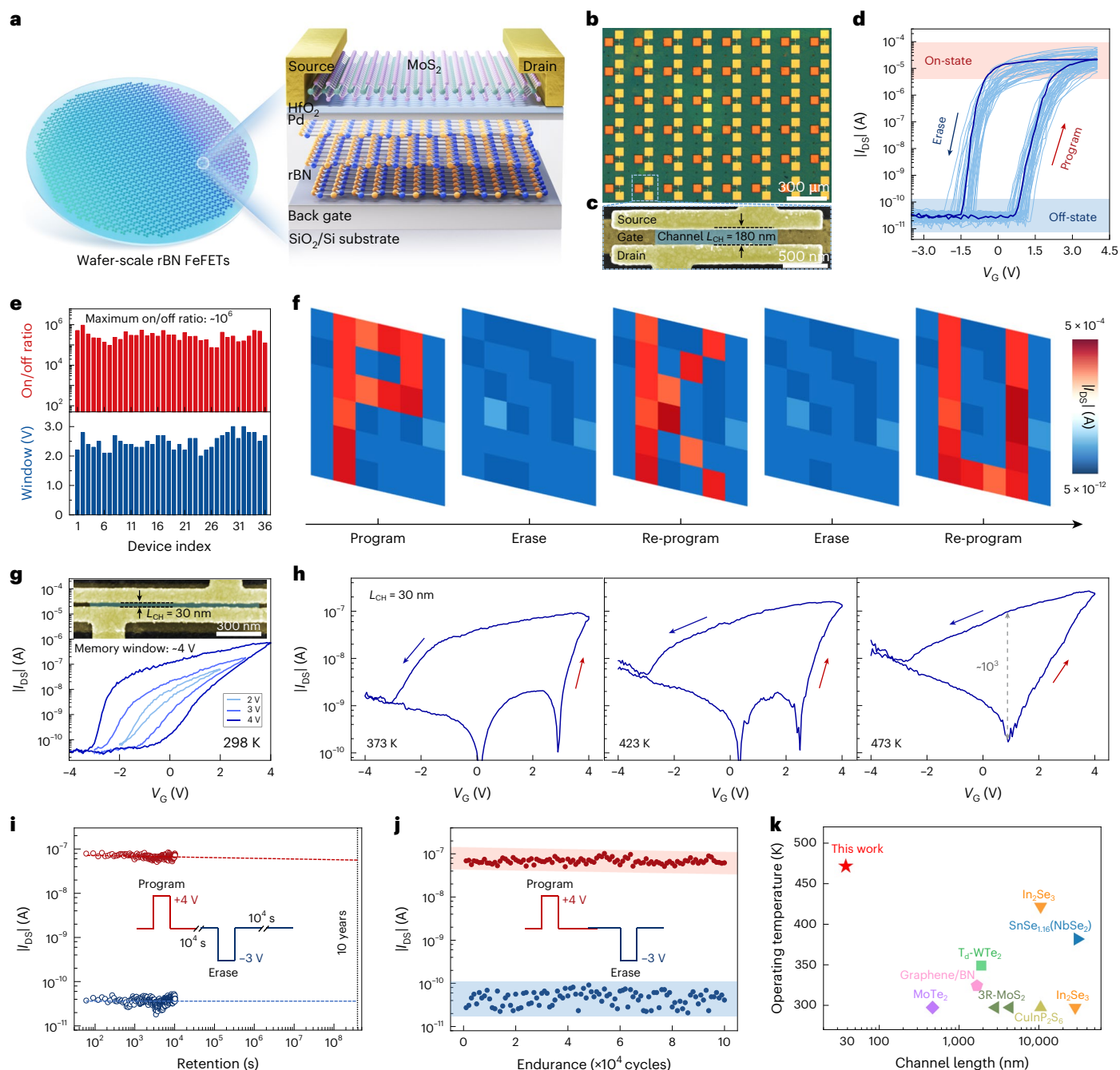


Fig. 4 | Wafer-scale ferroelectric performance of rBN/MoS₂ FeFET arrays.

a, b, Schematic of the device structure (**a**) and optical image (**b**) of large-scale rBN/MoS₂ FeFET arrays. **c**, Typical false-colour SEM image of a rBN/MoS₂ FeFET with the channel length (L_{CH}) of 180 nm. **d, e**, Ferroelectric hysteresis loops (**d**) and corresponding on/off ratio and memory window statistics (**e**) of rBN/MoS₂ FeFETs. **f**, Series of non-volatile memory maps with the characters of 'P', 'K' and 'U', produced during multiple programming-erasing cycles. **g**, Typical

transfer characteristic curve of a rBN/MoS₂ FeFET with the channel length of 30 nm. Inset: false-colour SEM image of the device. **h**, Evolution of ferroelectric hysteresis loops under operating temperatures of 373 K, 423 K and 473 K. **i, j**, Retention (**i**) and endurance (**j**) characteristics of the 30-nm-channel rBN devices. The programmed and erased pulse widths were set as 20 ns. **k**, Benchmarks of the channel lengths and operating temperatures of 2D-ferroelectric devices^{1,6,8,9,40,43–46}.

repeatedly reverse the polarization of the rBN FeFET, and the switching property remained almost unchanged exceeding 2×10^9 cycles (Fig. 3f and Extended Data Fig. 9e). In the subsequent retention test, the high and low resistance states were pre-programmed by V_G values of -4 and $+4$ V, respectively, and continuous monitoring at a probing V_G of 0.5 V revealed no appreciable degradation over 10^5 s (Fig. 3g). The projected retention could be extrapolated to be more than 10 years, demonstrating the long-term, valid data storage capability of this system. Such fatigue-resistant and non-volatile performance

of rBN crystals is on par with that of the best artificially AB-stacked bilayer flakes¹⁶.

Briefly, we summarized the properties of rBN and typical ferroelectric dielectrics from multiple perspectives, including the speed, retention, endurance, breakdown field and thermal conductivity (Fig. 3h and Supplementary Table 1, top)^{35–39}. Overall, the rBN sliding ferroelectric exhibited distinct advantages over its traditional counterparts in the construction of ultrafast, high-reliability and non-volatile memories.

Wafer-scale fabrication and scaling of ferroelectric devices

To demonstrate the industrial compatibility potential of the sliding ferroelectric memory technology, we fabricated a large-scale rBN FeFET array (Fig. 4a). The devices are designed in a typical metal–ferroelectric–metal–insulator–semiconductor (MFMIS) structure^{40,41}, utilizing rBN thin films as the dielectric layer and monolayer MoS₂ as the channel layer with the channel length (L_{CH}) of 180 nm (Fig. 4b,c). In addition, the palladium (Pd) intermediate metal layer was used as a screening electrode to reduce the depolarization field and an equipotential conductor to facilitate the more complete polarization switching of rBN layers. The HfO₂ barrier layer was used to suppress charge injection from the channel into the Pd layer. The output characteristic curves under gradually increased V_{G} values confirmed the good contact of devices (Extended Data Fig. 9f).

The ferroelectric nature of the rBN FeFETs was evinced by the counterclockwise hysteresis of the transfer characteristics, possessing a significant on/off ratio of 10^6 (Fig. 4d and Extended Data Fig. 9g,i). In contrast, no obvious counterclockwise memory window was observed in the identical hBN device (Extended Data Fig. 9h), indicating the hysteresis behaviour originating from the intrinsic ferroelectricity of rBN layers. The statistical rBN FeFETs exhibited superb reproducibility with an average on/off ratio of 3×10^5 and an average memory window of 2.5 V (Fig. 4e). A FeFET array was subsequently programmed by applying V_{G} values of +4 V and −3 V for the on and off states, respectively. By mapping the source–drain current (I_{DS}) values, our non-volatile memory array showed the characters of ‘P’, ‘K’ and ‘U’, highlighting the uniformity of rBN sliding ferroelectric arrays (Fig. 4f).

Another key criterion for the industrial viability of rBN FeFETs is the scalability to advanced nodes, as the channel length of existing sliding ferroelectric devices still remain on the micrometre level. Here we constructed short-channel rBN FeFETs with a reduced L_{CH} of 30 nm. The typical transfer curves with the counterclockwise hysteresis exhibited an ultra-large memory window of up to 4 V and a notable on/off ratio of 10^4 at room temperature (Fig. 4g). The ferroelectric behaviour observed in ultra-scale devices is consistent with the theoretical calculation, in which the width of ferroelectric domain walls is ~ 10 nm in grown rBN crystals¹⁸.

In situ heating measurements were also carried out to assess the thermal stability of rBN short-channel devices. Our results showed that the ferroelectric characteristics remained robust even up to 473 K, featuring a large memory window of 4 V and an on/off ratio of 10^3 (Fig. 4h), which far surpasses the chip thermal dissipation limit of 358 K (ref. 42). Moreover, the future introduction of a higher-barrier interlayer (for example, Al₂O₃ to replace HfO₂) could mitigate the slight off-state current degradation caused by thermal-assisted tunnelling at elevated temperatures.

We further investigated the retention characteristics of short-channel devices. The on/off ratio showed a minimal decay after 10^4 s and remained over 10^3 to 10 years by extrapolation (Fig. 4i). In addition, the performance of the rBN FeFETs did not markedly decrease after undergoing 10^5 polarization switching cycles (Fig. 4j), verifying the robust endurance feature of short-channel devices. Overall, the rBN/MoS₂ FeFETs exhibited the exceptional thermal stability and high-density integration potential, outperforming all the reported 2D-ferroelectric transistors (Fig. 4k and Supplementary Table 1, bottom)^{1,6,8,9,40,43–46}. For future integration of 2D sliding ferroelectric memories, we would like to note that the operating voltage can be further decreased through reducing the thickness of the rBN ferroelectric layer, introducing an interfacial barrier layer with a larger dielectric constant, or optimizing the interface quality.

Conclusions

We develop a reliable methodology for the batch production of 4-inch rBN wafers with consistently high crystal quality and phase purity.

Superior sliding ferroelectric properties with an ultrafast switching speed and high fatigue resistance, as well as integrated device performance with an ultrashort channel length and robust thermostability, are exhaustively demonstrated. Wafer-scale FeFET arrays indicate the great potential of 2D sliding ferroelectric materials for utilization in high-density data storage and in-memory computing technologies.

Online content

Any methods, additional references, Nature Portfolio 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/s41565-026-02280-4>.

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Methods

Preparation of Ni–B films

A 4-inch *m*-plane sapphire template was loaded into a corundum furnace (Tianjin Kaiheng Co. Ltd, custom designed) and annealed at 1,350 °C in air for 10 h to obtain the parallel high surface steps. Subsequently, a Ni–B film with a thickness of 500 nm and a boron atomic content of 15 at.% was deposited on the stepped sapphire template by the direct-current magnetron sputtering of the Ni–B targets (Zhongnuoxincai Co., Ltd) in a high-vacuum deposition system (X-TRP450F, Shenke Instrument Co., Ltd). The sputtering process was carried out at a substrate temperature of 400–600 °C with an applied power of 100 W and a deposition pressure of 1 Pa. For the high crystallinity of Ni–B alloy film, it should be slightly annealed at 500 °C for 1 h under a reducing atmosphere (300 standard cubic centimetres per minute (sccm) Ar and 30 sccm H₂) before growing the rBN layers.

Growth of single-crystal rBN wafers

The growth experiments of rBN films were carried out in a tube furnace with a chamber diameter of 12 cm (Tianjin Kaiheng Co. Ltd). The stepped Ni–B substrate was firstly annealed at 1,000 °C for 5 min under a reducing atmosphere (300 sccm Ar and 30 sccm H₂) to remove the surface impurities and oxides. Subsequently, the gas flux was modified to be of N₂ (300 sccm) and H₂ (100 sccm) at atmospheric pressure to grow the rBN wafers with different thicknesses. Then, the system was kept for 5 min with 200 sccm Ar and 100 sccm H₂ to etch the excess rBN domains and obtain the uniform rBN films. After growth, the furnace was naturally cooled to room temperature with Ar (300 sccm) and H₂ (10 sccm). To observe the aligned few-layer rBN domains at the initial stage, the growth process was left for about 5 min, followed by the etching process for 5 min. For the separated multilayer rBN domains, the Ni–B substrates with high surface steps were used at a relatively low temperature (950 °C) in a hydrogen-rich environment (500 sccm H₂ and 100 sccm N₂) for 30 min. In our control experiments, an identical procedure was carried out for synthesizing rBN thin layers using the Ni₆B₄ alloy substrate with a boron atomic content of 4 at.% and hBN samples based on the *c*-plane and *a*-plane sapphire templates, while the growth temperature was raised to 1,050 °C to obtain the hBN films on a flat Ni–B surface.

Transferring rBN wafers and exfoliating rBN flakes

The as-grown rBN wafers were wet transferred onto the target substrates using the well-established polypropylene carbonate (PPC) assisted technique⁴⁷. A PPC film was first spin-coated with a speed of 1,500 rpm for 1 min and baked at 90 °C for 3 min. Then, the wafers were immersed into the 0.1 mol l⁻¹ Na₂S₂O₈ solution at 60 °C for enough time to remove the metals. The floated PPC/rBN films and the sapphire substrates were soaked in deionized water several times to clean the surfaces. Finally, the PPC/rBN films were attached to the target substrates and annealed at 400 °C for 8 h under a low-pressure condition with 500 sccm Ar and 50 sccm H₂ to remove the PPC residue. To obtain the rBN flakes with the cleaner surface, we picked up the transferred rBN films with a thickness of ~45 nm using the Scotch tape method and exfoliated the flakes onto the SiO₂/Si substrates.

Ferroelectric measurements in rBN/graphene FeFET devices

The devices were fabricated using a tap-based transfer method with a polycarbonate/polydimethylsiloxane stamp. The thicknesses of selected rBN flakes were 10–20 nm. The Hall bar geometry for the transport measurements was patterned using electron beam lithography (EBL) and reactive ion etching. Subsequently, Cr (5 nm)/Au (60 nm) electrodes were deposited via standard EBL and thermal evaporation. The lithography resist was e-beam resist poly(methyl methacrylate) AR-P 679.04 with a thickness of about 400 nm, and the lithography process was carried out in the TESCAN MIRA SEM-EBL System. Direct-current gate biases were supplied using Keithley 2400 source

meters, where the four-terminal resistance of monolayer graphene was recorded using Stanford Research Systems SR860 lock-in amplifiers. For the ultrafast switching experiments, voltage pulses shorter than 10 μs were generated by a DTG5274 data timing generator connected to the direct-current port of the bias tee. Endurance characterization was performed across a range of frequencies; specifically, for cycling tests exceeding 10⁸ cycles, a maximum switching frequency of 10⁷ Hz was maintained.

Ferroelectric measurements in rBN/MoS₂ FeFET devices

The high-quality monolayer MoS₂ wafer was synthesized by the local element supply method⁴⁸. The devices were fabricated using the optimized PPC-assisted wet transfer method. The bottom electrodes were first fabricated using EBL (Raith Voyager), ultrahigh vacuum electron beam evaporation and lift-off techniques, with a Ti/Pd (5/20 nm) metal stack for the bottom electrodes. To improve the device stability, a 5-nm Pd intermediate layer was prepared onto the transferred rBN film, followed by the deposition of 5-nm HfO₂ as a barrier layer using atomic layer deposition (BeneQ TFS 200). Then, the grown MoS₂ monolayer was wet transferred. The rBN and MoS₂ regions were defined separately using reactive ion etching. Finally, source–drain contacts were fabricated using a Cr/Au (5/70 nm) metal stack. Electrical transport measurements were performed using a Keithley HSQB4200A semiconductor parameter analyzer. High-temperature experiments were conducted using a MPI TS300 12-inch manual probe station. Pulsed measurements were performed using a Keysight B1500A and 1530 semiconductor device analyzer. All of the source–drain currents were read out at a source–drain bias of 0.5 V and gate voltage of 0 V.

SHG measurements

The normal-incidence SHG was conducted by WiTec alpha300R with the incident light wavelength of 1,064 nm and the fixed excitation power of 20 mW. The laser-induced damage threshold was measured with the 546-nm femtosecond pulses (~120 fs, 250 kHz), which were generated by an ultrafast amplifier (Spectra-Physics, Spirit-HE) equipped with an optical parametric amplifier (Spectra-Physics, Spirit-NOPA-VISIR) and focused on the rBN samples by an Olympus objective (×20, numerical aperture of 0.4). The typical single spectra were excited by the 546-nm femtosecond pulses with an excitation power of 1.25 mW. The tilted-incidence SHG was excited by 800-nm femtosecond pulses from a Coherent laser (~150 fs, 250 kHz) generated by a Ti:sapphire oscillator. A collimated *p*-polarized laser beam (with a 2-mm spot size) was guided into the back aperture of a Nikon objective (×50, numerical aperture of 0.8). The incident angle was tuned by moving the beam position at the objective's back aperture using a retroreflector on a one-dimensional motorized stage. The reflected SHG signal was collected using the same objective after filtering the excitation laser and analysed by a polarizer along *p* polarization to exclude *s*-polarized SHG induced by the in-plane dipoles. To further exclude the in-plane dipole-induced SHG along *p* polarization, the sample was rotated to a specific crystal angle.

Other characterizations

P-*E* loops were characterized using an aixACCT ferroelectric test system (TF-Analyser 3000) via the dynamic hysteresis measurement mode at frequencies ranging from 0.1 kHz to 2 kHz. Electric breakdown measurements were performed using a Keysight B1500A semiconductor device analyzer. Thermal conductivity was measured by the frequency domain thermo-reflectance technique²³. SEM and energy dispersive spectroscopy images were collected by an FEI Nova NanoSEM 430 scanning electron microscope equipped with Ultim max100 nano-probe. Electron backscatter diffraction mappings were obtained using a PHI 710 scanning Auger nanoprobe. LEED patterns were obtained by a LEED-Auger spectrometer BDL800IR system in ultrahigh vacuum

with the pressure below 3×10^{-7} Pa. AFM topographic mappings were obtained by a tapping mode on Asylum Research MFP-3D and PFM measurements were performed on Asylum Research Cypher ES. SAED, HRTEM and STEM experiments were obtained in a JEOL 2010F system and a FEI Titan Themis G2 300 system, operated at 300 kV. XRD, UV-vis, XPS and Raman spectra were obtained by X'Pert³ MRD system, Varian Cary 5000 UV-vis-NIR spectrophotometer, Axis Ultra Imaging X-ray photoelectron spectrometer and WiTec alpha300R with the excited wavelength of 514 nm and laser power of 20 mW, respectively.

Data availability

The data supporting the findings of this study are presented within the paper. Additional data are available from the corresponding authors upon reasonable request. Source data are provided with this paper.

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Author contributions

K.L. supervised the project. K.L., Z.Z., C.Q. and L.W. conceived the project. K.L., Z.Z., L.W., E.W. and J.Q. developed the growth mechanism. J.Q., T.G., L.C., C.Z. and E.T. prepared the sputtered Ni-B films and rBN wafers. J.T., Qiu Yang, L.G., T.G., J.Q., C.Q. and X.L. fabricated FeFET devices and measured the ferroelectric performances. J.Q., T.G., M.Z., Qianqian Yang, K.X., F.Y., B.S., L.Z. and X.Z. performed SEM, AFM, LEED, electron backscatter diffraction, XRD, UV-vis, XPS, Raman, SHG, *P*–*E* loop, frequency domain thermo-reflectance and PFM measurements. Q.G. and P.W. performed SAED, HRTEM and STEM experiments. All authors discussed the results and wrote the paper.

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Competing interests

The authors declare no competing interests.

Additional information

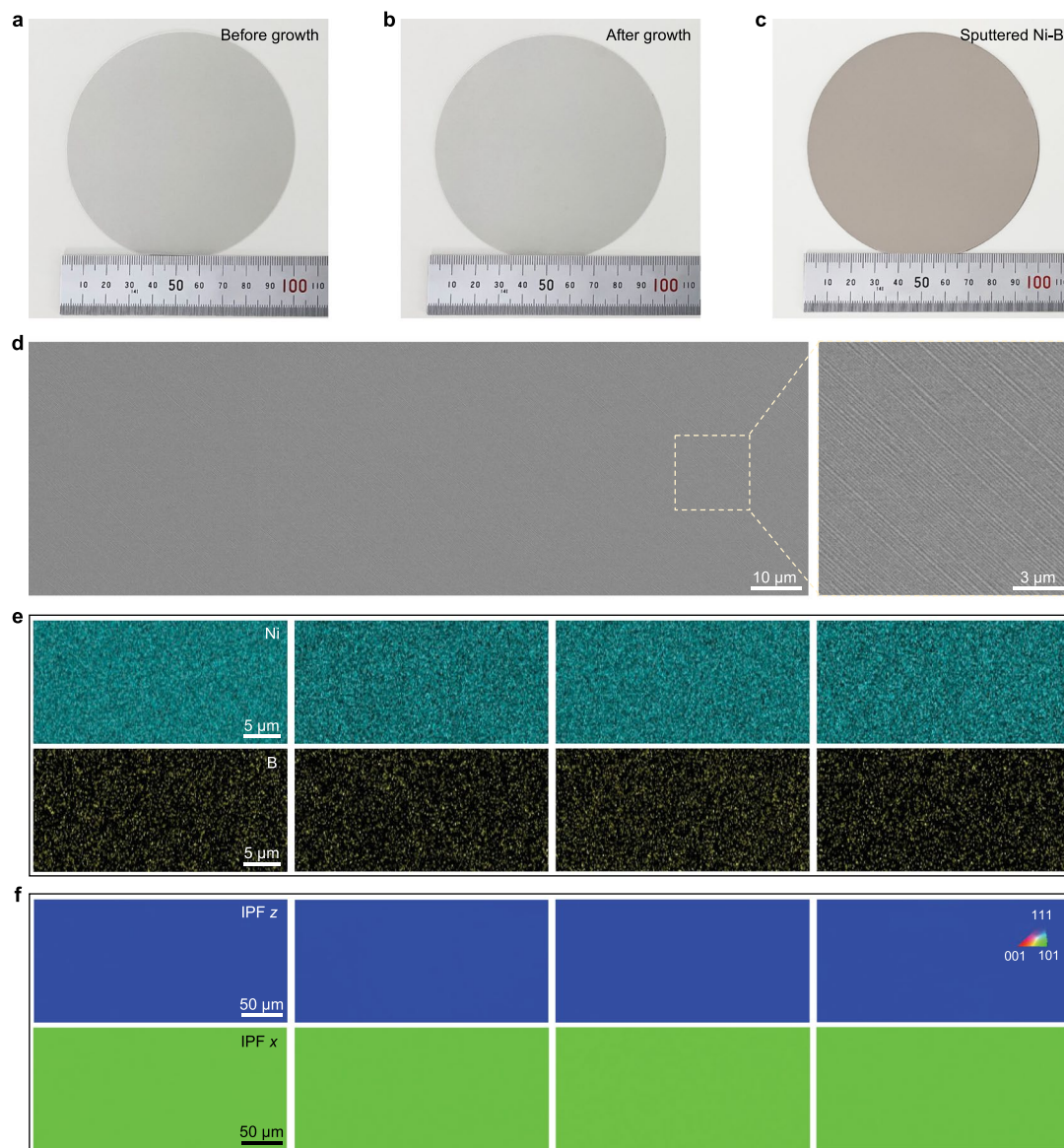
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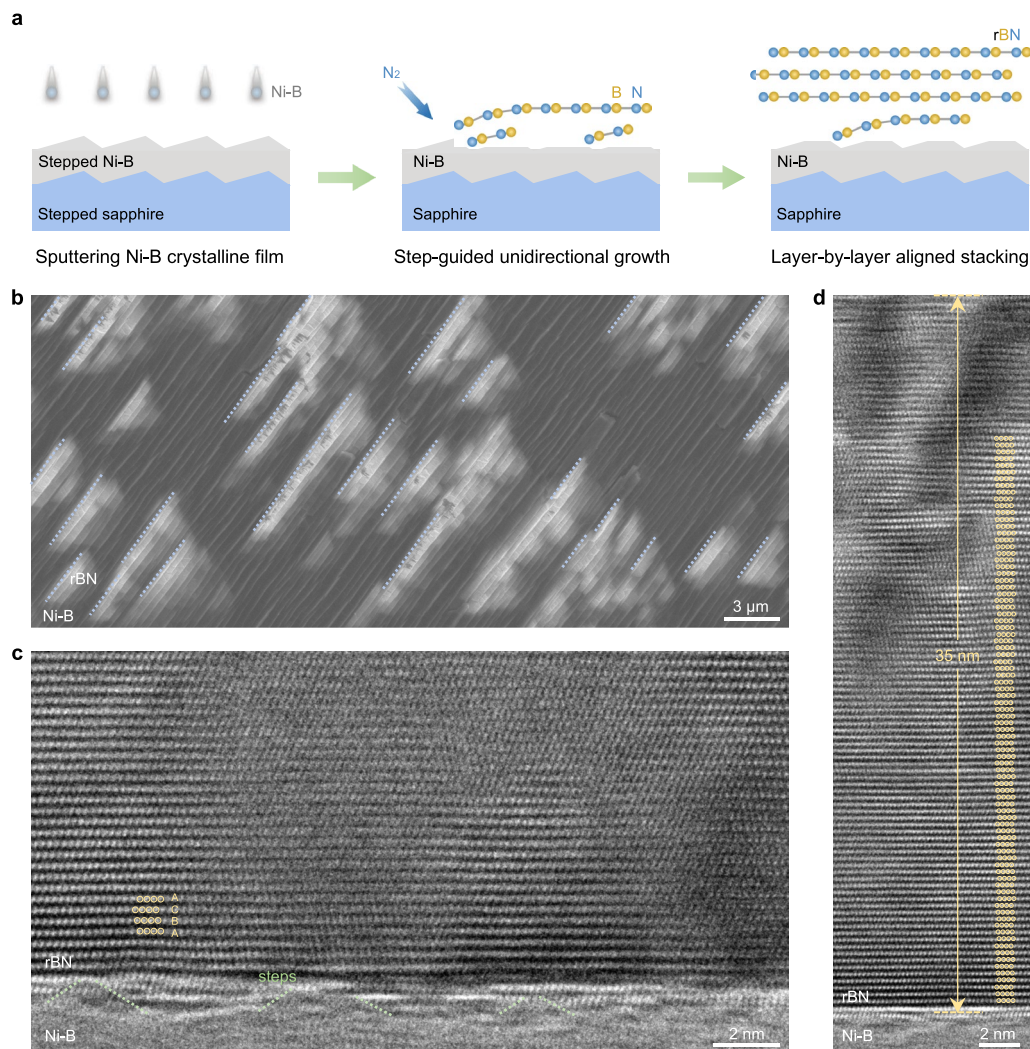
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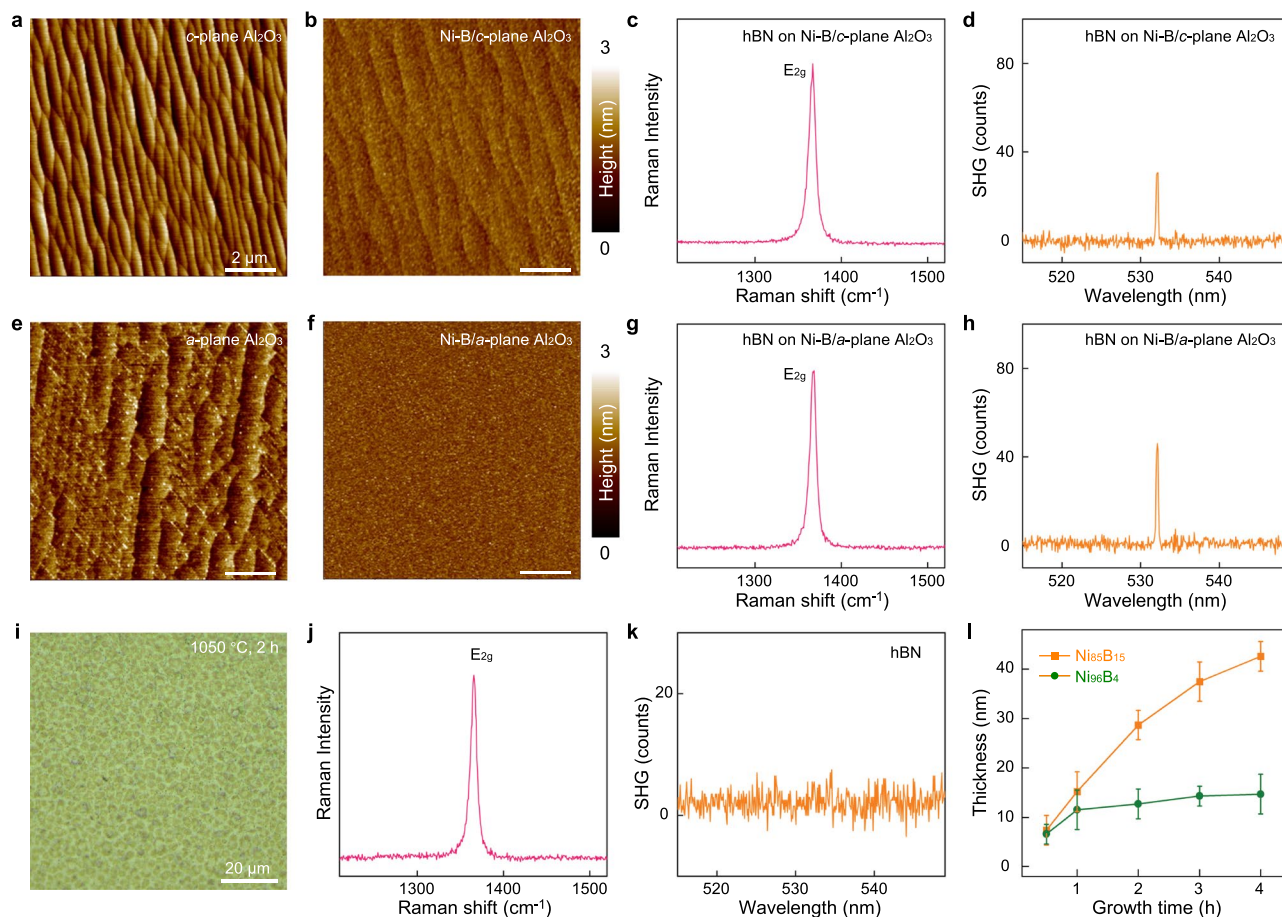
Extended Data Fig. 1 | Characterizations of Ni-B film deposited by magnetron sputtering. **a-c**, Photographs of the stepped $m\text{-Al}_2\text{O}_3$ templates before **(a)** and after **(b)** growth, and the $m\text{-Al}_2\text{O}_3$ template with sputtered Ni-B film **(c)**. **d**, Typical SEM image of the sputtered Ni-B film. **e**, Corresponding EDS mappings of B

and Ni, showing the uniform element distribution. **f**, EBSD inverse pole figure (IPF) mappings along the z and x directions, verifying the single crystallinity of sputtered Ni-B films at a large scale. All images are of the same scale.



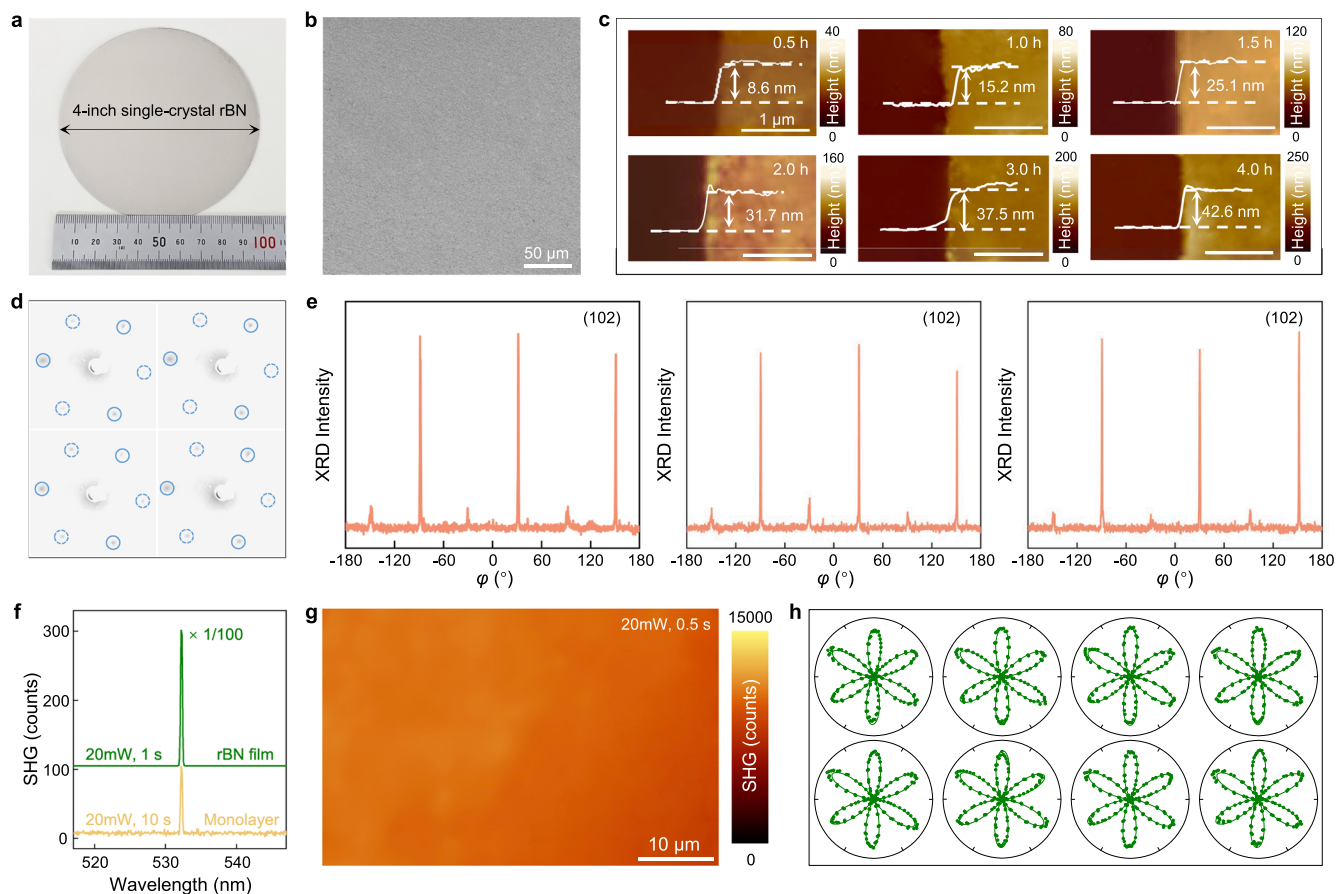
Extended Data Fig. 2 | Microscopic mechanism of the stabilization of the rhombohedral phase. **a**, Schematic of step-templated interfacial epitaxy of rBN layers. **b**, Typical SEM image of aligned rBN multilayer domains guided by the surface steps of Ni-B alloy film. The blue dashed lines highlight the unidirectional orientation of rBN domains. **c,d**, Cross-sectional HRTEM images of as-grown

rBN layers on the stepped Ni-B surface, demonstrating the rhombohedral phase stabilized by surface steps (**c**) and the perfect ABC stacking order of rBN layers over the whole film region (**d**). The green dashed lines in (**c**) mark the steps of Ni-B film, and the yellow cycles in (**c**) and (**d**) mark the B-N pairs.



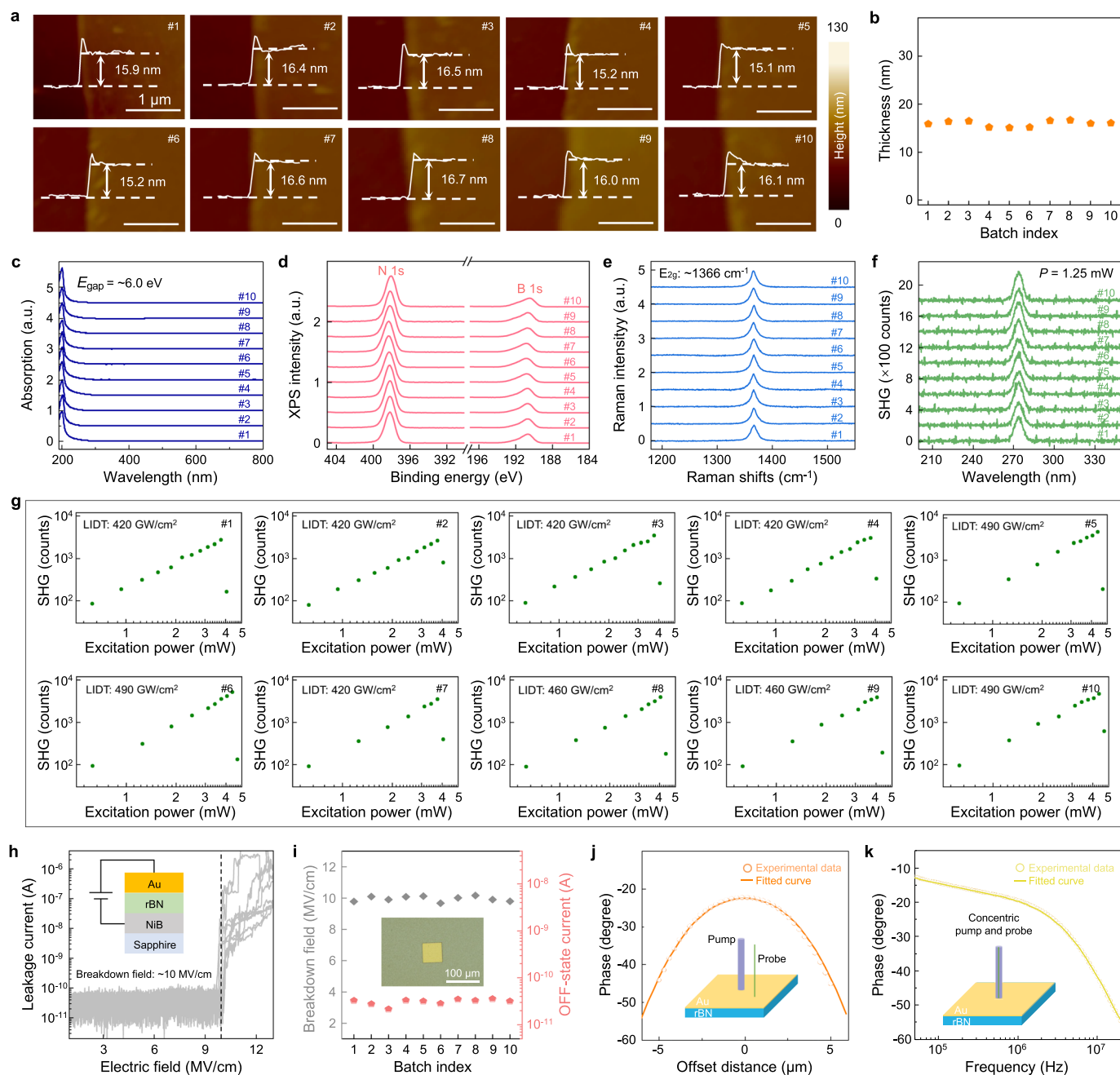
Extended Data Fig. 3 | Control experiments of different growth parameters. **a,b**, AFM images of *c*-plane Al₂O₃ after annealing (**a**) and subsequent Ni-B deposition (**b**). **c,d**, Raman (**c**) and SHG (**d**) measurements of the grown films (primarily hBN) based on *c*-plane Al₂O₃. **e,f**, AFM images of *a*-plane Al₂O₃ after annealing (**e**) and subsequent Ni-B deposition (**f**). **g,h**, Raman (**g**) and SHG (**h**) measurements of the grown films (primarily hBN) based on *a*-plane Al₂O₃. Collectively, these results verify the critical role of surface steps on *m*-plane Al₂O₃

in modulating the ABC stacking order of rBN layers. **i-k**, Optical microscopic image (**i**), Raman (**j**) and SHG (**k**) of BN samples grown at an elevated temperature of 1,050 °C for 2 h, demonstrating the formation of the hexagonal phase. **l**, Thickness evolution of rBN layers over growth time using Ni₈₅B₁₅ and Ni₉₆B₄ films. The variations in thickness measurements arise from ten different positions on the each transferred rBN film.



Extended Data Fig. 4 | Characterizations of 4-inch rBN wafers. **a**, Photograph of a 4-inch rBN wafer grown on the $m\text{-Al}_2\text{O}_3$ template. **b**, SEM image of the rBN film. **c**, Typical AFM mappings collected at the edge of transferred rBN films with the different growth time. All images are of the same scale. **d,e**, LEED patterns (**d**) and XRD ϕ -scan data (**e**) obtained at the randomly-selected areas in one wafer, showing the large-scale single crystallinity of rBN films. **f**, SHG spectra of

the monolayer and rBN film with the thickness of ~ 15 nm under the excitation wavelength of 1,064 nm. The SHG intensity of rBN is enhanced by around 2,000 times compared with that of the monolayer. **g,h**, Homogenous SHG intensity mapping (**g**) and identical polarized SHG patterns (**h**) of rBN wafers, respectively, verifying a uniform configuration of rBN single crystals.



Extended Data Fig. 5 | Quality characterizations of rBN wafer batches.

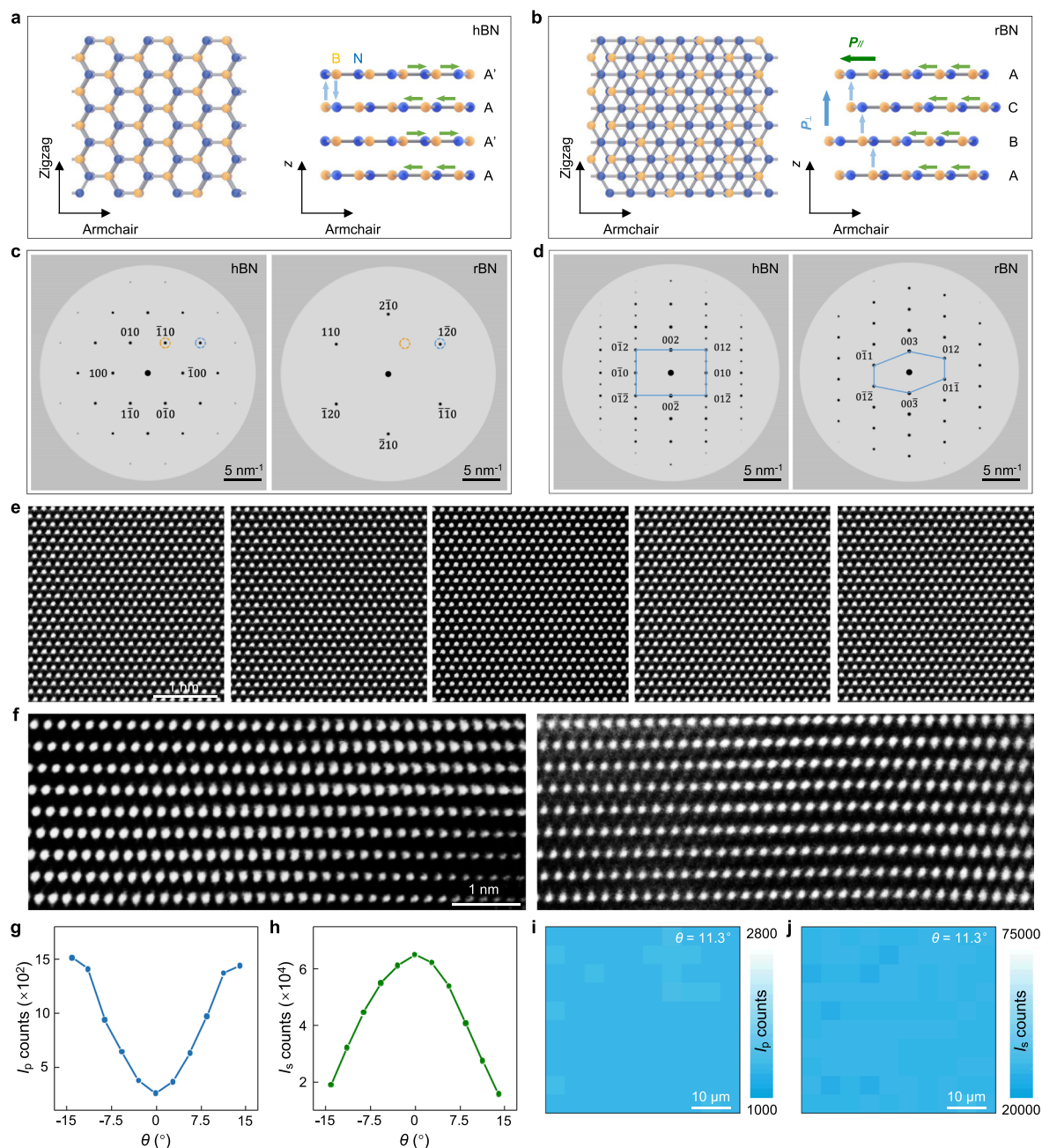
a,b, AFM height mappings (**a**) and corresponding data statistics (**b**) of different batches of rBN wafers grown for 1 h. All images are of the same scale.

c-f, UV-vis (**c**), XPS (**d**), Raman (**e**), and SHG (**f**) spectra of ten batches of rBN wafers, respectively, showing the congruent quality of different rBN batches.

g, Power-dependent SHG signals under the excitation wavelength of 546 nm.

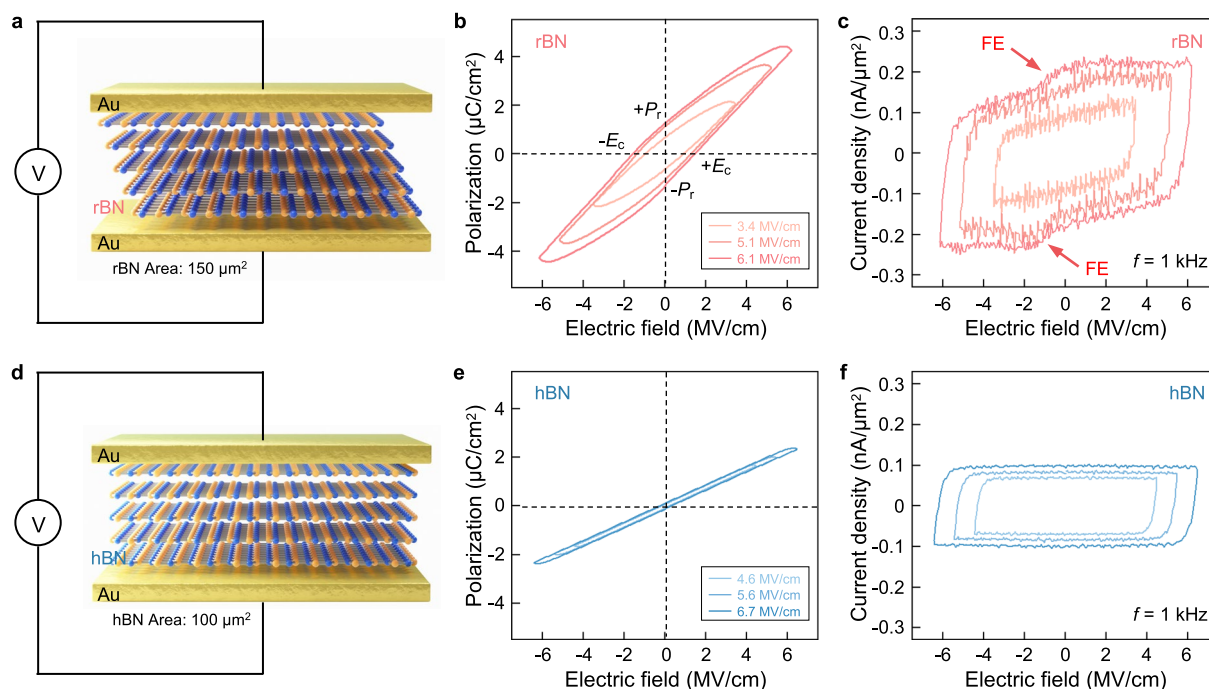
h,i, Typical current-electric field characteristics (**h**) and the corresponding statistics (**i**) of rBN films, demonstrating the exceptional insulating property with a high breakdown field of ~ 10 MV/cm and a small OFF-state current of $\sim 8 \times 10^{-10}$ A. Insets, schematic of the device structure (**h**) and optical image of the device (**i**).

j,k, FDTR phases from beam-offset (**j**) and f -sweep (**k**) measurements together with the fitted curves. Insets, schematics of the two experimental schemes.



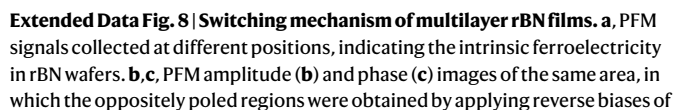
Extended Data Fig. 6 | Characterizations of atomic arrangement of rBN crystals. **a,b**, Schematics of the stacking configuration of hBN (**a**) and rBN (**b**) from the top and side view. The large green and blue arrows represent the in-plane ($P_{\parallel}$) and out-of-plane ($P_{\perp}$) polarizations, and the small green and blue arrows mean the dipole moments of in-plane and out-of-plane B-N pairs, respectively. **c,d**, Stimulated planar (**c**) and cross-sectional (**d**) SAED patterns of

hBN and rBN. The orange and blue dashed circles in (**c**) mark the first-order and second-order diffraction spots of planar SAED patterns, respectively. **e,f**, Planar HRTEM (**e**) and cross-sectional STEM (**f**) images collected at different positions of rBN. All images are of the same scale. **g,h**, Tilted angle (θ) dependent I_p (**g**) and I_s (**h**) signals. **i,j**, Large-scale I_p (**i**) and I_s (**j**) intensity mappings obtained at the fixed θ of 11.3° , verifying the polarization uniformity of the rBN crystals.

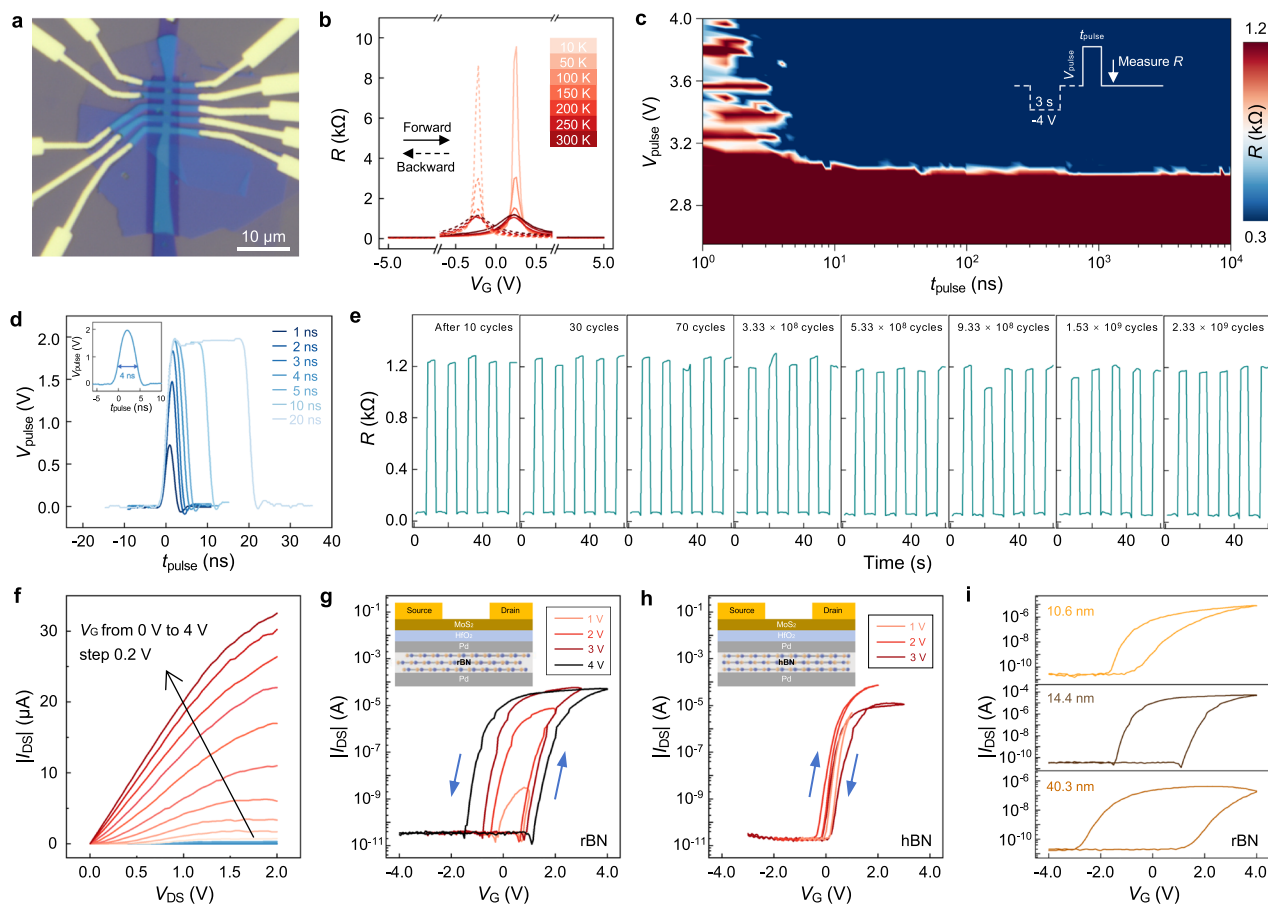


Extended Data Fig. 7 | P - E loop measurements. **a**, Schematic of the MFM capacitance device using the transferred rBN film as the ferroelectric layer and the deposited Au as electrodes. **b,c**, Typical P - E loops (**b**) and corresponding J - E curves (**c**) obtained on the rBN device. P_r and E_c in (**b**) denote the remnant polarization and coercive field, respectively. The P_r is estimated to be $-1.3\ \mu\text{C}/\text{cm}^2$. The switching current peaks are highlighted as the ferroelectricity (FE) signal by the red arrows in (**c**). **d**, Schematic of the capacitance device using hBN as

the dielectric layer. **e,f**, Typical P - E loops (**e**) and corresponding J - E curves (**f**) obtained on the hBN device. No significant hysteresis was observed in the P - E loops and the current remained smooth with varying electric fields in the J - E curves, verifying the absence of ferroelectric switching arising from the hBN multilayers or metal-BN-metal device architecture. The areas of rBN and hBN samples have been listed in the schematic diagrams.



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Extended Data Fig. 9 | Ferroelectric performance of rBN/FeFET devices.

a, Optical microscopic image of a rBN/graphene FeFET device. **b**, Forward and backward sweeps of the graphene sensor's resistance versus the gate voltage at different temperatures ranging from 10 K to 300 K. **c**, Complete switching speed measurement of rBN/graphene FeFET devices using the V_{pulse} with t_{pulse} of 1 nanosecond to 10 microseconds. **d**, Oscilloscope traces of applied pulses, showing the amplitude attenuation and waveform distortion for $t_{\text{pulse}} < 3$ ns due to a finite instrument bandwidth. **e**, Representative resistance-switching cycles

during the endurance test. **f**, Output characteristic curves of a rBN/MoS₂ device. **g, h**, Transfer characteristics of rBN (**g**) and hBN (**h**) devices. Insets, the schematics of the BN/MoS₂ device architectures. The blue arrows mark the counterclockwise and clockwise sweeps of rBN and hBN devices, respectively. **i**, Typical transfer curves of rBN devices using the samples with different thicknesses. The thicker rBN layers lead to larger memory windows, with values of 1.7 V for the 10.6-nm device and 4.4 V for the 40.3-nm device.