

NANOMATERIALS

Step geometry-guided growth of rhombohedral graphene

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Rhombohedral (ABC)–stacked graphene has emerged as a platform for exploring correlated and topological physics. However, its large-scale, pure-phase synthesis has been hindered by intrinsic thermodynamic metastability and kinetic instability. In this work, we introduce a step geometry-guided epitaxial strategy to deterministically control interlayer slip, enabling the synthesis of pure-phase rhombohedral graphene. Using this approach, we obtained pure (phase purity > 99%) rhombohedral graphene with an area reaching 160 micrometers by 80 micrometers and thickness ranging from ~15 layers to ~120 nanometers. The resulting samples establish the first comprehensive reference dataset of Raman fingerprints and intrinsic band structures from few-layer films to ~200-layer bulk. Electronic transport measurements reveal a layer-antiferromagnetic state and the quantum anomalous Hall effect, enabling scalable exploration of next-generation quantum science and technology applications.

Interlayer stacking is a key degree of freedom that dictates the properties of multilayer two-dimensional (2D) materials. For graphene, two prominent stacking orders are broadly observed: the common Bernal (AB) stacking (hexagonal, D_{6h} symmetry) and the rare metastable rhombohedral (ABC) stacking (trigonal, D_{3d} symmetry). With a simple interlayer slip, Bernal-stacked graphene can be transformed into the rhombohedral form, and such a transition would reshape its electronic landscape by generating flat bands and nonzero Berry curvature that are absent in the Bernal phase (1–7). These features are indicative of strong electronic correlations and nontrivial topology and would make rhombohedral graphene an ideal platform for exploring exotic quantum phenomena, including superconductivity (8–12), Chern insulating states (13–20), and the quantum anomalous Hall effect (21–25).

The exploration of these phenomena requires the stable production of pure-phase rhombohedral graphene. However, most previous studies have relied on mechanical exfoliation to obtain rhombohedral graphene, which is inherently limited by small sample sizes, appreciable phase impurities, and laborious phase identification. For example, five-layer pure-phase rhombohedral graphene is buried in eight possible mixed-phase variants, with a theoretical fabrication probability of <1% (26, 27). Such intrinsic limitations hinder systematic investigations and scalable applications.

Substantial efforts have been devoted to the synthesis of rhombohedral graphene, including methods based on silicon carbide (SiC) surface decomposition (28) and strain-assisted stabilization of local ABC domains on curved substrates (29, 30). These studies provided an important foundation for understanding how the substrate can

influence stacking order; however, the growth of rhombohedral graphene with both large in-plane size and high out-of-plane phase purity remained elusive. This difficulty arises from two fundamental barriers. First, thermodynamically, the formation energy of rhombohedral graphene is ~0.2 meV per atom greater than that of Bernal stacking (31). Second, kinetically, postgrowth perturbations, such as thermal fluctuations and cooling-induced local strains, would readily trigger interlayer sliding that irreversibly converts the rhombohedral phase into the Bernal phase.

Recent advances in 2D material growth highlight both challenges and opportunities (32–34). In C_3 -symmetry binary systems, such as boron nitride (BN) and transition-metal dichalcogenides (TMDs), the rhombohedral phase has already been realized. In these systems, the ABC (3R) phase is the local lowest-energy configuration when layers share the same in-plane orientation, and their strong coupling with the substrate can be used to lock the orientation of each layer and enable stacking control (35, 36). Moreover, once the 3R phase is formed, its conversion to the globally stable AA' (2H) phase requires a 60° rotation of the entire layer, imposing a kinetic barrier that protects the rhombohedral order. However, graphene has a single-element composition and a higher C_6 lattice symmetry, so the layers intrinsically align in the same orientation, making Bernal stacking the most stable configuration. This higher symmetry also lacks orientational protection that is present in BN and TMDs. A graphene lattice rotated by 60° is identical to its original lattice, so the rhombohedral stacking order can be switched to the Bernal stacking easily by a simple slip of one C–C bond length. Therefore, achieving stable, pure-phase rhombohedral graphene requires a fundamental reconceptualization of the growth strategies that can thermodynamically guide the formation of ABC stacking and kinetically suppress its relaxation to Bernal stacking.

Growth mechanism of rhombohedral graphene

We present a step geometry-guided mechanism for rhombohedral graphene growth by controlling the interlayer displacement. A graphene sheet on a substrate has an edge plane serving as a terminating boundary (Fig. 1A). This edge plane enforces a precise slip magnitude (governed by the slip angle φ , defined between the edge plane and the substrate plane) and slip direction (governed by the orientation angle θ , defined between the confinement direction $\mathbf{n}$ and the graphene armchair axis $\mathbf{a}$) during the growth process. The slip angle φ determines the degree of geometric constraint on interlayer displacement. When $\varphi = 90^\circ$ ($2\varphi = 180^\circ$, corresponding to an unconfined edge on a flat surface), Bernal stacking is energetically favorable. Therefore, achieving rhombohedral stacking requires $\varphi < 90^\circ$ to impose appropriate confinement (Fig. 1, B and C).

The orientation angle θ determines the alignment between the confinement direction and the interlayer displacement. When $\theta = 0^\circ$, the confinement aligns precisely with the displacement required for rhombohedral stacking, enabling ideal phase control (Fig. 1, C and D, and fig. S1). When $\theta = 30^\circ$, however, the confinement direction becomes parallel to the graphene zigzag axis, eliminating confinement and resulting in Bernal stacking.

By controlling φ and θ , the overall displacement toward rhombohedral stacking can be achieved. However, stable phase formation also requires that every graphene layer responds coherently to this confinement. During growth, φ enforces discrete bending of each layer at distinct atomic sites (Fig. 1E). If these bending sites are random across layers, the displacement would lose coherence, and rhombohedral stacking could not form. Through our density functional theory calculations,

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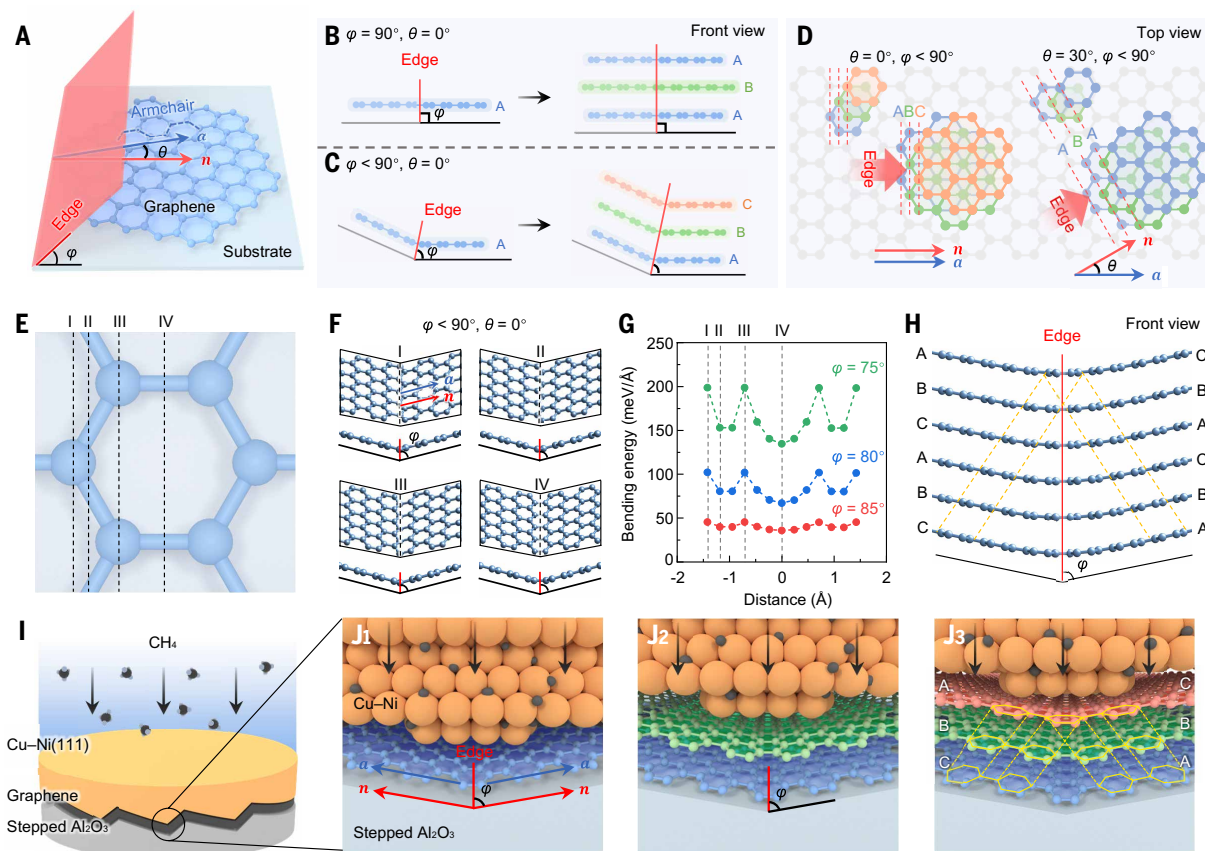


Fig. 1. Step geometry-guided growth mechanism for rhombohedral graphene. (A) Schematic of the geometric alignment between the graphene lattice and the edge plane on a substrate. (B and C) Role of the slip angle φ . (B) At $\varphi = 90^\circ$, the geometric constraint is absent, leading to thermodynamically favorable Bernal stacking. (C) At $\varphi < 90^\circ$, the geometric constraint induces interlayer slip and leads to rhombohedral stacking. (D) Role of orientation angle θ : alignment at $\theta = 0^\circ$ favors rhombohedral stacking, whereas misalignment at $\theta = 30^\circ$ yields Bernal stacking. (E) Atomic structure of graphene with possible bending sites I to IV. (F) Relaxed atomic structures after bending at the four sites. (G) Bending energy as a function of φ for different bending sites. (H) MD-simulated rhombohedral structures at $\varphi = 78^\circ$ and $\theta = 0^\circ$. (I) Schematic of the growth design. (J1 to J3) Schematic of the growth process, with the formation of mirror-symmetric ABC and CBA domains.

we reveal that bending preferentially occurs at the highly symmetric midpoints of C–C bonds (Fig. 1, F and G, site IV), and such a preference becomes more dominant as φ decreases (that is, as the confinement increases) (Fig. 1G). Our calculations also show that nucleation is energetically preferred at the step edge, making graphene formation overwhelmingly more likely to initiate at the step-confined region (fig. S2). As a result, the graphene layers nucleate and bend at the same site IV, collectively forming an edge plane that anchors the graphene edge and systematically guides the stacking sequence toward the rhombohedral order (Fig. 1H).

To achieve such coordinated control over both angles, we developed a geometrically confined architecture using stepped sapphire (Al_2O_3) and single-crystal copper-nickel alloy (Cu–Ni) foils for rhombohedral graphene growth (Fig. 1I and supplementary materials, materials and methods). In this work, stepped Al_2O_3 serves as a geometrical confinement to enforce the interlayer displacement of the graphene, and Cu–Ni foil serves as the growth substrate that enables interfacial epitaxy of the graphene. The introduction of Ni increases the carbon solubility in the substrate, facilitating carbon transport. Consequently, carbon atoms from methane (CH_4) decomposition continuously diffuse through the Cu–Ni foil to the buried growth interface, ensuring a sufficient and sustained carbon supply throughout the growth process. For the geometrical confinement, the angle φ is determined by the Al_2O_3 step morphology, and the angle θ is controlled by the alignment of the Al_2O_3 steps with the lattice orientation of the Cu–Ni

foil (fig. S3). Because graphene nucleates at the Cu–Ni/ Al_2O_3 interface and conforms to both surfaces, each newly grown layer must fit the step geometry (Fig. 1, J1 to J3). This step confinement guides each new layer to nucleate and grow with a specific, atomically precise displacement relative to the layer below it, ensuring that the entire stack grows as a single, coherent rhombohedral crystal (fig. S4).

With this strategy, we synthesized rhombohedral graphene at a growth temperature of $\sim 1150^\circ\text{C}$. However, during subsequent cooling to room temperature, the relaxation of interfacial stress and local temperature fluctuations can kinetically introduce interlayer sliding, converting the metastable rhombohedral stacking into the Bernal phase. Fortunately, our growth strategy incorporated a kinetic stabilization mechanism. Because of the structural equivalence across the step, graphene grew symmetrically and formed mirror-symmetric ABC and CBA domains (Fig. 1J3). In this regime, any spontaneous transition to Bernal stacking on one side would correspondingly induce AA stacking on the other side (fig. S5), whose energy is ~ 10 meV per atom higher than that of AB stacking (37). Such an energetically unfavorable pathway effectively locks the rhombohedral stacking order, suppressing phase conversion throughout the entire growth process.

Phase control of rhombohedral graphene

On the basis of the designed mechanism, we first manipulated the slip angle φ . At $\varphi \sim 75^\circ$ (with $\theta = 0^\circ$ if not specified) (Fig. 2A1), the increased geometric confinement enabled the conformal growth of ultrathin

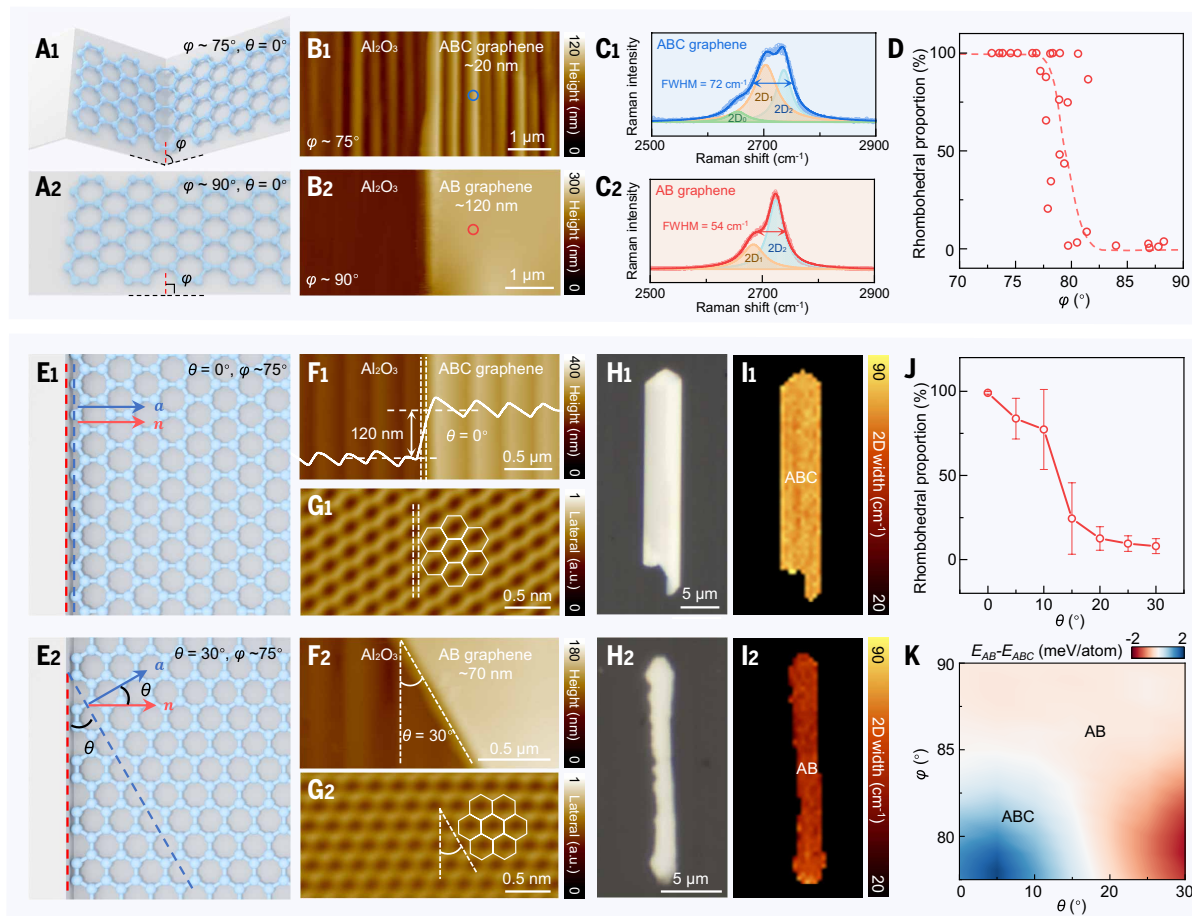


Fig. 2. ϕ - and θ -dependent phase control of rhombohedral graphene. (**A₁** and **A₂**) Schematics of graphene growth (**A₁**) with and (**A₂**) without geometric confinement. (**B₁** and **B₂**) AFM images near graphene edges showing (**B₁**) a conformal morphology with confinement and (**B₂**) a flat morphology without confinement. (**C₁** and **C₂**) Raman spectra (circles) with Lorentzian multipole fits (lines) displaying (**C₁**) ABC stacking at $\phi \sim 75^\circ$ and (**C₂**) AB stacking at $\phi \sim 90^\circ$, corresponding to the marked circle regions in (**B₁**) and (**B₂**). (**D**) Rhombohedral proportion versus ϕ across samples. (**E₁** and **E₂**) Schematics of the graphene lattice alignment at (**E₁**) $\theta = 0^\circ$ and (**E₂**) $\theta = 30^\circ$. (**F₁** and **F₂**) AFM images near graphene edges at (**F₁**) $\theta = 0^\circ$ and (**F₂**) $\theta = 30^\circ$. (**G₁** and **G₂**) High-resolution AFM images identifying lattice directions at (**G₁**) $\theta = 0^\circ$ and (**G₂**) $\theta = 30^\circ$. (**H₁** and **H₂**) Optical images of the graphene domains at (**H₁**) $\theta = 0^\circ$ and (**H₂**) $\theta = 30^\circ$. (**I₁** and **I₂**) Raman 2D peak width maps revealing (**I₁**) the pure ABC stacking at $\theta = 0^\circ$ and (**I₂**) the complete AB stacking at $\theta = 30^\circ$. (**J**) Experimental dependence of the rhombohedral proportion on θ . (**K**) MD-simulated phase diagram showing the relative energy of AB and ABC stacking as a function of θ and ϕ .

graphite on stepped Al_2O_3 , as revealed with atomic force microscopy (AFM) (Fig. 2B₁). Under these growth conditions, rhombohedral stacking was readily obtained and confirmed by characteristic Raman signatures (38, 39): a broad 2D peak with a full-width at half-maximum (FWHM) of $\sim 72 \text{ cm}^{-1}$ and three well-resolved subpeaks (2D_0 , 2D_1 , and 2D_2) (Fig. 2C₁).

By contrast, when ϕ approaches 90° , geometric confinement was insufficient to drive interlayer slip (Fig. 2A₂). In this regime, the grown ultrathin graphite exhibited a flat morphology (Fig. 2B₂), and its Raman spectrum featured a narrow 2D peak ($\sim 54 \text{ cm}^{-1}$) without the presence of a 2D_0 subpeak (Fig. 2C₂), revealing a Bernal structure (38, 39). These Raman signatures were also validated by means of infrared scanning near-field optical microscopy (IR-SNOM) (29, 40, 41), confirming the stacking identification (fig. S6). Through our systematic investigations, we established $\phi \sim 80^\circ$ as the critical threshold, only below which the geometric confinement could stabilize the rhombohedral order (Fig. 2D and fig. S7).

Next, we examined the role of the orientation angle θ . At $\theta = 0^\circ$ (with $\phi \sim 75^\circ$ if not specified) (Fig. 2E₁), the confinement direction aligned perfectly with the rhombohedral slip direction, yielding ultrathin pure-phase rhombohedral graphite (Fig. 2, F₁ to I₁). Atomic-resolution AFM

confirmed directional alignment and revealed that the steps could confine graphite growth to thicknesses reaching 120 nm (Fig. 2, F₁ and G₁). This thickness scalability arises from the interfacial-growth characteristics of our process, in which each new layer forms under identical geometrical constraints, enforcing a deterministic interlayer displacement and stacking sequence (fig. S4). Further statistical analysis across multiple samples consistently exhibited a high phase purity of $>99\%$ under the optimized growth conditions (Fig. 2J), demonstrating the robustness of this strategy against thermal and mechanical perturbations during growth.

We then explored how variations in θ influence stacking stability (fig. S8). At $\theta = 5^\circ$, although mixed Bernal domains began to appear, rhombohedral stacking remained dominant (fig. S9, A₁ to E₁). At $\theta = 10^\circ$, however, coexisting rhombohedral and Bernal regions were observed (fig. S9, A₂ to E₂), and the average rhombohedral portion substantially decreased (Fig. 2J). When $\theta \geq 20^\circ$, the geometric manipulation completely lost phase control, and the ultrathin graphite converted entirely to Bernal stacking (Fig. 2, E₂ to I₂, and fig. S9, A₃ to E₃). These results demonstrate that the stacking order of the graphite layers could be precisely regulated by deliberately adjusting the orientation angle θ , enabling reliable production of graphene with the desired stacking configuration for practical applications (fig. S9F).

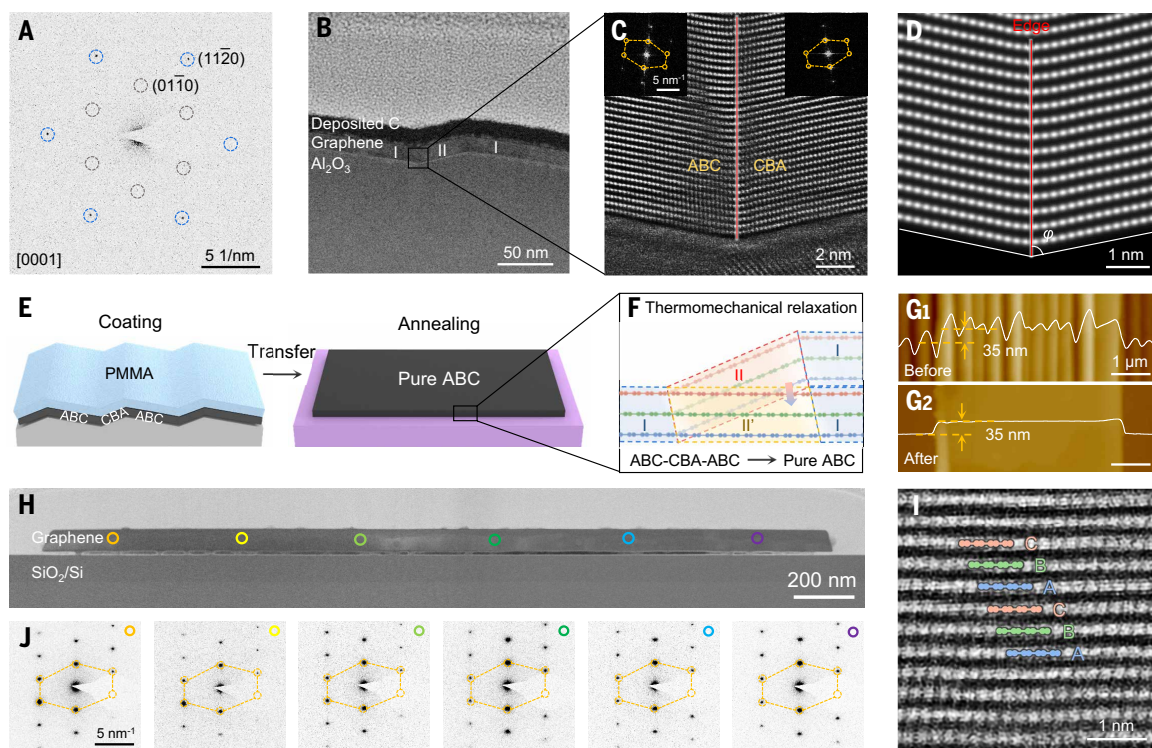


Fig. 3. Structural characterization and thermomechanical relaxation of rhombohedral graphene. (A) SAED pattern of rhombohedral graphene along the [0001] axis, showing pure ABC stacking signatures with the absence of Bernal-specific primary diffraction spots (dashed gray circles). (B) Low-magnification cross-sectional STEM image revealing conformal graphene growth along the Al_2O_3 step. (C) Atomic-resolution STEM image from the boxed region in (B). (Insets) Fast Fourier transform (FFT) patterns from each side. (D) Simulated cross-sectional STEM image. (E) Schematic of the thermomechanical relaxation process. (F) Schematic of interlayer slip (region II $\rightarrow$ region II') during relaxation. (G₁ and G₂) AFM morphology of rhombohedral graphene (G₁) before and (G₂) after relaxation. (H) Cross-sectional STEM image of atomically flat ABC graphene after relaxation. (I) Atomic-resolution STEM image of rhombohedral graphene. (J) SAED patterns from multiple positions in (H), indicated with circles with corresponding colors.

These findings, together with molecular dynamics (MD) simulations, outline a phase diagram that is coregulated by the orientation angle θ and slip angle ϕ (Fig. 2K). The diagram defines the stability windows of different stacking orders and provides guidance for their controlled growth. The optimal rhombohedral growth conditions were identified at $\theta < 10^\circ$ and $\phi < 80^\circ$, at which coupled angle control reshapes the energy landscape and ensures the precise growth of rhombohedral graphene. In this optimal window, we ultimately achieved the growth of large-area ($\sim 160 \mu\text{m}$ by $80 \mu\text{m}$), thin (~ 24 layers) pure-phase rhombohedral graphene (fig. S10).

Structural characterization of rhombohedral graphene

To validate the phase purity and material quality of the grown rhombohedral graphene, we conducted comprehensive structural analyses. Selected-area electron diffraction (SAED) revealed characteristic out-of-plane rhombohedral signatures, with Bernal-specific primary diffraction spots entirely absent (Fig. 3A). Cross-sectional scanning transmission electron microscopy (STEM) characterization demonstrated conformal growth along the substrate steps, with the graphene layers maintaining intimate contact and structural continuity across the terraces (Fig. 3B). Atomic-resolution STEM imaging exhibited mirror-symmetric ABC and CBA configurations (Fig. 3C), which was consistent with the simulated STEM images that reproduce rhombohedral stacking (Fig. 3D). In addition, rhombohedral graphene samples with thicknesses ranging from ~ 15 layers to $\sim 120 \text{ nm}$ were also characterized, all of which exhibited rhombohedral stacking features and consistent step-geometry-guided growth behavior (figs. S11 and S12). Together, these results provide direct evidence of long-range ordered rhombohedral stacking and validate the step-geometry-guided growth mechanism.

Although the mirror-symmetric ABC-CBA structure kinetically stabilized the rhombohedral phase during the cooling process, it also introduced linear domain boundaries in the samples. To remove these boundaries and achieve a fully uniform structure, we developed a thermomechanical relaxation process (Fig. 3E). After etching away the metal substrate, the rhombohedral graphene was coated with a soft polymethyl methacrylate (PMMA) layer and transferred onto a flat substrate. Afterward, we used a mild annealing procedure ($\leq 400^\circ\text{C}$) to release the residual strain. Because graphene grown on Al_2O_3 steps consisted of long (region I) and short (region II) planes (Fig. 3B), strain redistribution during annealing was dominated by the long planes and drove the graphene on the short planes to adopt the major stacking order (region II'), ultimately forming a uniform crystal order (Fig. 3F and fig. S13). This behavior is supported by MD simulations, which show that once the step-geometry constraint is removed during transfer, the CBA-to-ABC rearrangement proceeds spontaneously with no obvious barrier, enabling the formation of a uniform ABC sequence (fig. S14).

As a result, our method used thermally and mechanically driven structural reorganization to transform the initial ABC-CBA structure into a pure ABC rhombohedral phase (Fig. 3, G₁ and G₂, and fig. S15), yielding a structurally uniform material essential for reliable characterization and device fabrication. We also found that the annealing temperature was a key parameter: Mild annealing ($\leq 400^\circ\text{C}$) fully preserved the rhombohedral phase during relaxation, whereas annealing above $\sim 800^\circ\text{C}$ could activate a transition toward Bernal stacking (fig. S16). We further characterized the structure of the transferred rhombohedral samples. Cross-sectional STEM imaging confirmed that the film thoroughly relaxed into an atomically flat morphology with no residual

step structures (Fig. 3H). Atomic-resolution imaging revealed a strict three-layer periodicity with no detectable Bernal configuration (Fig. 3I). Consistent SAED patterns across the sample with identical orientations confirmed the homogeneous rhombohedral phase, demonstrating the successful elimination of the ABC-CBA boundaries (Fig. 3J). This thermomechanical relaxation process was also effective for thinner rhombohedral graphene films, yielding uniform rhombohedral graphene after transfer and relaxation (fig. S17).

Spectroscopic and electronic properties of rhombohedral graphene

The successful large-scale growth of pure-phase rhombohedral samples enabled the exfoliation of high-quality graphene for few-layer characterizations. The as-grown samples were transferred onto SiO₂/Si substrates and then thinned by means of a standard mechanical exfoliation method. These samples established a comprehensive layer-resolved reference dataset of Raman fingerprints and intrinsic band structures, spanning from a few layers to ~200 layers. Stacking-sensitive Raman spectroscopy revealed distinct 2D-band features across this thickness

range (Fig. 4, A and B). Compared with Bernal stacking, rhombohedral graphene consistently exhibited higher 2D₁ peak area to total 2D peak area ratios (I_{2D1}/I_{total}) at all thicknesses (Fig. 4C). In addition, the 2D peak in rhombohedral graphene broadened progressively with increasing thickness, in contrast to the narrowing tendency in Bernal stacking (Fig. 4D). Across the measured range, the rhombohedral 2D peaks were 4 to 18 cm⁻¹ wider than their Bernal counterparts, reflecting their distinctive band structures with broader resonances in the phonon momenta (42). These systematic spectroscopic fingerprints established a reliable reference for the identification of rhombohedral stacking.

The exceptional phase purity of these samples also enabled direct visualization of the stacking-dependent electronic structures with the use of nanoscale angle-resolved photoemission spectroscopy (nano-ARPES) (Fig. 4, E₁ to E₄). The measured band dispersions exhibited well-defined flat surface bands near the Fermi level ($E_F \pm 50$ meV). The flat bands were most profound in three- to five-layer samples and persisted up to the ~200-layer bulk (43, 44), directly revealing the origin of the reduced quasiparticle velocity and increased electronic correlations of rhombohedral graphene (23). In addition, the number of subbands away from

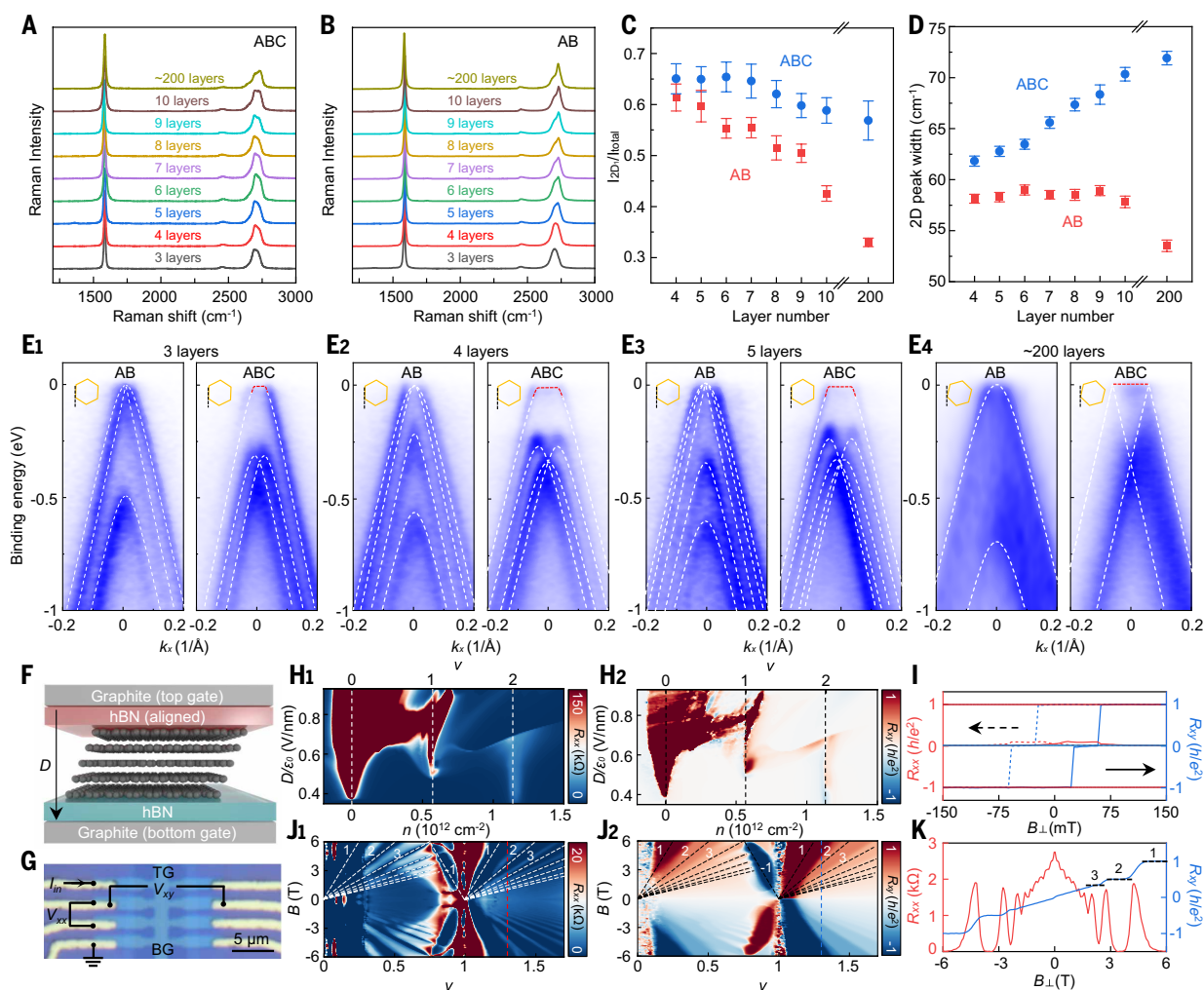


Fig. 4. Spectroscopic and electrical characterization of rhombohedral graphene. (A and B) Raman spectra of three- to ten-layer and bulk graphene with (A) rhombohedral and (B) Bernal stacking. (C) Ratios of the 2D₁ peak area to the total 2D peak area for rhombohedral and Bernal stackings as a function of layer number. (D) 2D peak widths versus layer number for both stackings. (E₁ to E₄) Nano-ARPES spectra with corresponding simulations (dashed curves) for three- to five-layer and bulk (left) Bernal and (right) rhombohedral graphene. Red dashed lines indicate the flat bands. (Insets) Cut directions (black lines) and the graphene Brillouin zone (yellow hexagons). (F) Schematic of the five-layer rhombohedral graphene/hBN moiré device. (G) Optical image of the device. BG, bottom gate; TG, top gate. (H₁ and H₂) Symmetrized longitudinal resistance (R_{xx} , H₁) and antisymmetrized Hall resistance (R_{xy} , H₂) versus carrier density n and displacement field D under $B = 0.1$ T. (I) R_{xx} and R_{xy} as functions of B at $\nu = 1.03$ and $D/\epsilon_0 = 0.54$ V/nm. (J₁ and J₂) Landau fan diagrams of (J₁) R_{xx} and (J₂) R_{xy} at $D/\epsilon_0 = 0.54$ V/nm. (K) Linecut taken along the dashed lines in (J₁) and (J₂) at $\nu = 1.3$.

E_F increased with increasing layer thickness, and the subbands eventually merged into a bulk Dirac cone in the bulk limit. The measured band structures differ drastically from those of Bernal-stacked graphene with the same layer numbers and show excellent agreement with tight-binding and first-principles calculations (45–47). The sharp spectra and well-resolved thickness-dependent band structure evolution confirm the quality and uniformity of our samples.

Then, we performed high-resolution electronic transport investigations on devices fabricated from graphene exfoliated from the as-grown samples. In a dual-gated seven-layer device at 1.5 K (fig. S18, A and B), we observed two well-defined insulating states near charge neutrality—that is, a correlation-driven layer-antiferromagnetic insulating (LAF) state at zero displacement field (D) and a band insulating (BI) state at $|D| > 0.3$ V/nm (fig. S18, C and D) (14, 15). Quantum oscillations were also revealed at low magnetic fields (fig. S18E). These results confirmed that such quantum phenomena can be accessed in our grown rhombohedral graphene samples.

We next fabricated a five-layer rhombohedral graphene device aligned with hexagonal boron nitride (hBN) at a twist angle of 0.27° (Fig. 4, F and G). At an ultralow temperature (10 mK), we identified a series of vertical resistance peaks corresponding to correlated states at integer fillings of 0, 1, and 2 electrons per moiré unit cell in the phase diagram (Fig. 4, H_1 and H_2) (19–22). At a moiré filling of $\nu = 1$, the system exhibited the quantum anomalous Hall effect, in which the Hall resistance (R_{xy}) reached a quantized resistance of h/e^2 (where h is Planck's constant and e is the electron charge), whereas the longitudinal resistance (R_{xx}) nearly disappeared at zero magnetic field (Fig. 4I). Landau fan diagram measurements further resolved the gap at $\nu = 1$, with a Chern number $C = 1$ that could survive at zero magnetic field (Fig. 4, J_1 and J_2). In addition to the $C = 1$ gap, other quantum Hall states were also well developed under finite magnetic fields, exhibiting quantized R_{xy} and vanishing R_{xx} (Fig. 4K). Collectively, the observation of these well-resolved correlated states, pronounced phase transitions, and distinct topological quantum phenomena reflects the exceptional homogeneity and low intrinsic disorder of our grown samples.

Discussion and outlook

We introduce a step geometry-guided epitaxial strategy that overcomes the intrinsic thermodynamic and kinetic barriers of rhombohedral graphene growth. This method yields high-purity (>99%) rhombohedral graphene with both a large in-plane area (160 μm by 80 μm) and high out-of-plane coherence (from ~15 layers to ~120 nm), providing an unprecedented platform for exploring flat-band physics, strong correlations, and topological order. In addition, our strategy opens the possibility for the controlled synthesis of graphene beyond Bernal and rhombohedral stacking orders, such as phases with larger unit cells along the out-of-plane direction (for example, graphene with ABAC and ABCBA stacking orders). Our approach establishes a paradigm for stacking-sequence engineering in emergent quantum materials and paves the way for their scalable applications in future quantum science and technologies.

REFERENCES AND NOTES

1. K. F. Mak, J. Shan, T. F. Heinz, *Phys. Rev. Lett.* **104**, 176404 (2010).
2. W. Bao et al., *Nat. Phys.* **7**, 948–952 (2011).
3. C. H. Lui, Z. Q. Li, K. F. Mak, E. Cappelluti, T. F. Heinz, *Nat. Phys.* **7**, 944–947 (2011).
4. G. R. Chen et al., *Nat. Phys.* **15**, 237–241 (2019).
5. Y. Shi et al., *Nature* **584**, 210–214 (2020).
6. H. Zhou et al., *Nature* **598**, 429–433 (2021).
7. T. Han et al., *Nature* **623**, 41–47 (2023).
8. G. Chen et al., *Nature* **572**, 215–219 (2019).
9. H. Zhou, T. Xie, T. Taniguchi, K. Watanabe, A. F. Young, *Nature* **598**, 434–438 (2021).

10. Y. Choi et al., *Nature* **639**, 342–347 (2025).
11. T. Han et al., *Nature* **643**, 654–661 (2025).
12. J. Yang et al., *Nat. Mater.* **24**, 1058–1065 (2025).
13. G. Chen et al., *Nature* **579**, 56–61 (2020).
14. T. Han et al., *Nat. Nanotechnol.* **19**, 181–187 (2024).
15. K. Liu et al., *Nat. Nanotechnol.* **19**, 188–195 (2024).
16. Y. Sha et al., *Science* **384**, 414–419 (2024).
17. J. Ding et al., *Phys. Rev. X* **15**, 011052 (2025).
18. J. Xie et al., *Nat. Mater.* **24**, 1042–1048 (2025).
19. S. H. Aronson et al., *Phys. Rev. X* **15**, 031026 (2025).
20. D. Waters et al., *Phys. Rev. X* **15**, 011045 (2025).
21. Z. Lu et al., *Nature* **626**, 759–764 (2024).
22. Z. Lu et al., *Nature* **637**, 1090–1095 (2025).
23. T. Han et al., *Science* **384**, 647–651 (2024).
24. B. Zhou, H. Yang, Y. H. Zhang, *Phys. Rev. Lett.* **133**, 206504 (2024).
25. F. Winterer et al., *Nat. Phys.* **20**, 422–427 (2024).
26. Y. Yang et al., *Nano Lett.* **19**, 8526–8532 (2019).
27. C. H. Lui et al., *Nano Lett.* **11**, 164–169 (2011).
28. W. Norimatsu, M. Kusunoki, *Phys. Rev. B Condens. Matter Mater. Phys.* **81**, 161410 (2010).
29. Z. Gao et al., *Nat. Commun.* **11**, 546 (2020).
30. C. Bouhafs et al., *Carbon* **177**, 282–290 (2021).
31. J. P. Nery, M. Calandra, F. Mauri, *2D Mater.* **8**, 035006 (2021).
32. B. Qin et al., *Science* **385**, 99–104 (2024).
33. L. Wang et al., *Nature* **629**, 74–79 (2024).
34. L. Liu et al., *Nat. Mater.* **24**, 1195–1202 (2025).
35. C. Cazorla, T. Gould, *Sci. Adv.* **5**, eaau5832 (2019).
36. A. Fan et al., *Sci. Adv.* **11**, eadx8192 (2025).
37. G. Savini et al., *Carbon* **49**, 62–69 (2011).
38. C. Cong et al., *ACS Nano* **5**, 8760–8768 (2011).
39. S. L. L. M. Ramos, M. A. Pimenta, A. Champi, *Carbon* **179**, 683–691 (2021).
40. K. G. Wirth et al., *ACS Nano* **16**, 16617–16623 (2022).
41. D. Beitner et al., *Nano Lett.* **23**, 10758–10764 (2023).
42. A. Torche, F. Mauri, J. C. Charlier, M. Calandra, *Phys. Rev. Mater.* **1**, 041001 (2017).
43. H. Xiao et al., *Adv. Sci. (Weinh.)* **12**, e2412609 (2025).
44. H. Zhang et al., *Proc. Natl. Acad. Sci. U.S.A.* **121**, e2410714121 (2024).
45. M. Koshino, *Phys. Rev. B Condens. Matter Mater. Phys.* **81**, 125304 (2010).
46. H. Henck et al., *Phys. Rev. B* **97**, 245421 (2018).
47. Y. Zhang et al., *Nat. Nanotechnol.* **20**, 222–228 (2025).
48. M. Zhao et al., Dataset for “Step geometry-guided growth of rhombohedral graphene”. Zenodo (2026); <https://doi.org/10.5281/zenodo.20374368>.

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Author contributions: K.L., X.L., Z.L., F.D., and Zh.Z. supervised the project. K.L., Zh.Z., and M.Z. conceived the experiments. M.Z., Zh.Z., and X.S. conducted the growth and transfer experiments. X.S. fabricated the devices. Q.G. performed the TEM experiments. C.Q. and F.D. conducted the MD and DFT simulations. Za.Z., Z.F., and X.L. performed the electronic transport experiments. H.X. and Z.L. performed the ARPES experiments. J.Z. and S.W. performed the IR-SNOM experiments. Y.Y. and T.D. conducted the tight-binding calculations. M.Z., Zh.Z., X.S., and K.L. wrote the article. X.-Z.L., L.W., and E.W. revised the manuscript. All the authors discussed the results and commented on the manuscript. **Competing interests:** A Chinese patent application (no. 2026102199718) related to this article was submitted by Peking University (K.L., M.Z., Zh.Z., X.S., and Q.G.). All other authors declare that they have no competing interests. **Data, code, and materials availability:** All data and details of materials synthesis are available in the main text or the supplementary materials. Additional datasets are available in Zenodo (48). **License information:** Copyright © 2026 the authors, some rights reserved; exclusive licensee American Association for the Advancement of Science. No claim to original US government works. <https://www.science.org/about/science-licenses-journal-article-reuse>

SUPPLEMENTARY MATERIALS

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Step geometry–guided growth of rhombohedral graphene

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Editor's summary

Phase-pure rhombohedral graphene, which has an ABC layer stacking, has been grown through step geometry–guided epitaxy. Zhao *et al.* used a copper-nickel–coated sapphire surface with a stepped geometry chosen so that interlayer slip and orientation angles of the graphene layers favored rhombohedral stacking. The layer-antiferromagnetic state and the quantum anomalous Hall effect for this phase were determined from electronic transport measurements. —Phil Szuromi

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