

Ultraclean CrOCl/bilayer graphene heterointerfaces enable enhanced electrical modulation

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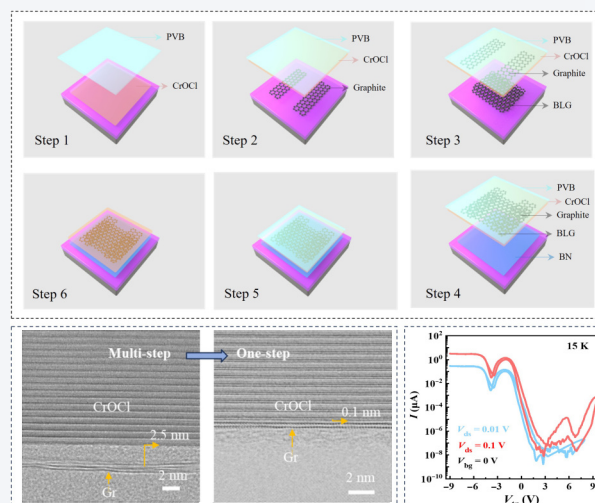
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ABSTRACT: Layer-by-layer assembly is widely used for constructing two-dimensional van der Waals heterostructures, yet polymer-induced interfacial contamination in conventional transfer processes often limits device performance. Here, we show that ultraclean chromium oxychloride (CrOCl)/bilayer graphene (BLG) heterointerfaces, realized via a one-step transfer process, enable significantly enhanced electrical modulation. Precise control of the adhesion and release behavior of a polyvinyl butyral support effectively suppresses crack formation and minimizes interfacial residues, yielding uniform and contamination-free heterointerfaces. As a result, directly laminated CrOCl/BLG/boron nitride (BN) devices exhibit an exceptional switching ratio exceeding 10^8 without post-annealing, outperforming layer-by-layer assembled counterparts by nearly seven orders of magnitude. These results highlight the critical role of interface cleanliness in governing the electrical performance of bilayer graphene heterostructures and provide a robust strategy for high-performance two-dimensional hetero-integration.

KEYWORDS: polyvinyl butyral, ultraclean CrOCl/BLG heterointerfaces, one-step transfer, switching ratio



1 Introduction

van der Waals (vdW) heterostructures provide a powerful platform for modulating charge transport, band alignment, and many-body interactions in two-dimensional (2D) materials, enabling a wide range of electronic, optoelectronic, and quantum phenomena [1–5]. Among these systems, graphene-based heterostructures are

particularly sensitive to interfacial conditions, as charge transfer, electrostatic screening, and interlayer coupling predominantly occur within an atomic-scale region at the interface [6, 7]. Consequently, even trace interfacial contamination can strongly perturb carrier mobility, bandgap opening, and electrostatic gating efficiency in graphene devices.

Despite extensive progress in vdW heterostructure assembly, most state-of-the-art fabrication strategies rely on polymer-assisted and layer-by-layer transfer processes [8–14]. While polymer supports facilitate deterministic stacking, residual polymers and interfacial adsorbates are difficult to eliminate completely and inevitably accumulate at buried interfaces [15]. These residues introduce interfacial trap states and charge scattering centers, degrading key device metrics such as subthreshold swing, on/off current ratio, and transport uniformity in graphene field-effect

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transistors [16, 17]. For bilayer graphene (BLG), where electrostatic modulation and bandgap opening critically depend on a clean and well-defined interfacial electrostatic environment, interface cleanliness becomes a decisive factor governing electrical modulation efficiency [18, 19].

Recent studies have highlighted that ultraclean interfaces are essential for realizing emergent electronic states in graphene heterostructures, including quantum Hall states, correlated insulating phases, and electrically tunable transport regimes [7, 20–27]. In this context, chromium oxychloride (CrOCl), a layered antiferromagnetic insulator with a van der Waals crystal structure, has emerged as an attractive interfacial material for graphene-based heterostructures [28–32]. Owing to its strong interfacial charge coupling and electrically tunable electronic states, CrOCl has been shown to induce unconventional transport phenomena in graphene, such as low-field quantum Hall effects and gate-controllable correlated insulating behavior [20, 21, 33–35]. These findings underscore that the electronic response of graphene is not solely determined by material composition, but is critically governed by the cleanliness and quality of the heterointerface. However, the realization of such interface-driven electronic modulation is fundamentally constrained by contamination introduced during conventional multi-step transfer processes. Achieving ultraclean CrOCl/graphene interfaces therefore remains a key material challenge.

In this work, we address this limitation by employing a polyvinyl butyral (PVB)-assisted one-step transfer strategy that enables the direct formation of CrOCl/BLG/boron nitride (BN) heterostructures with minimal interfacial residues. By precisely controlling the adhesion and release behavior of the PVB support,

the process suppresses crack formation during pickup and significantly improves interfacial cleanliness. As a result, the ultraclean CrOCl/BLG heterointerfaces achieved in this work enable markedly enhanced electrical modulation, yielding BLG devices with an on/off ratio exceeding 10^8 without post-annealing, which is nearly seven orders of magnitude higher than that of devices fabricated via conventional layer-by-layer transfer. These results demonstrate that interface cleanliness plays a decisive role in governing the electrical performance of graphene heterostructures and establish a robust route toward high-performance and interface-engineered 2D electronic devices.

2 Result and discussion

Figure 1(a) depicts two heterojunction transfer strategies (multi-step and the one-step transfer techniques). In the multi-step transfer technique, similar to conventional transfer processes, PVB is contacted with each-layer to fabricate the heterojunction. This necessitates multiple isopropyl alcohol (IPA) solution removals of PVB during the transfer process. Frequent PVB interface contact and cleaning inevitably introduce defects such as bubbles and residues, leading to reduced carrier transport efficiency and structural stability of the heterojunction. In contrast, the one-step technique employs a simplified process: PVB is used solely to peel off the topmost CrOCl layer (step 1 in Fig. 1(a)), which then serves as a transfer carrier to lift the underlying graphite electrode and graphene (steps 2 and 3 in Fig. 1(a)). Subsequently, the pre-assembled stacked structure is transferred onto a BN substrate (steps 4 and 5 in Fig. 1(a)), and finally, the topmost PVB layer is removed via IPA solution cleaning to yield the final heterojunction

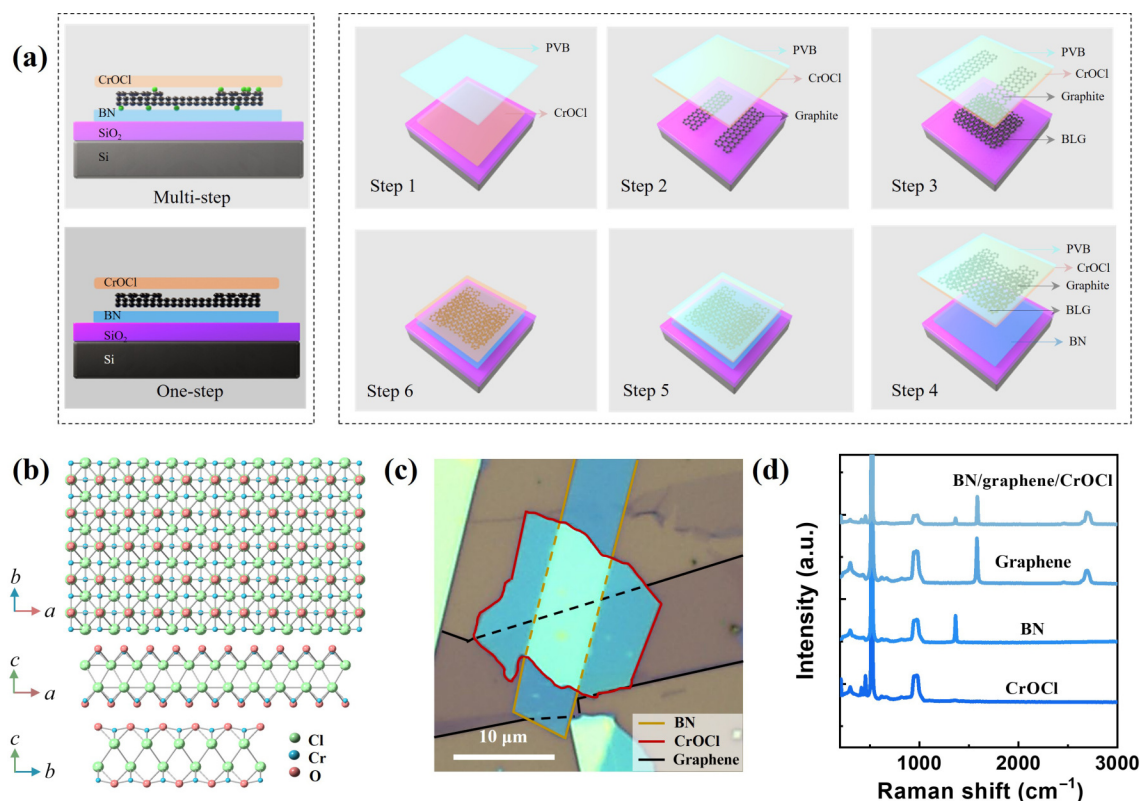


Figure 1 PVB-assisted one-step transfer technology for constructing CrOCl/graphene/BN heterojunctions. (a) Detailed process of the transfer technique. (b) Schematic diagram of typical CrOCl crystal structure. (c) Optical image of the CrOCl/graphene/BN heterojunction. (d) Raman characterization of the CrOCl/graphene/BN heterojunction.

(step 6 in Fig. 1(a)). Direct PVB–two-dimensional material interface contact is confined to the top layer, while internal interfaces remain pristine. This intrinsic cleanliness significantly enhances interfacial compatibility and optimizes heterojunction interface contact. As shown in Fig. 1(b), the bulk phase CrOCl belongs to the orthorhombic crystal system within the $Pmmn$ space group. The fundamental structural unit is the CrO_4Cl_2 octahedron, wherein each Cr^{3+} ion is surrounded by four O^{2-} ions and two Cl^- ions, forming a distorted octahedron [30]. Cr–O atomic layers are interleaved between Cl–Cl layers, stacked along the a -axis through weak van der Waals forces [31]. The crumpled chain structure of the Cr–O layers endows CrOCl with pronounced anisotropic physical properties. Therefore, the system exhibits low symmetry characterized by either a triple rotation (C3) or no rotational symmetry (C1), providing crucial conditions for emerging novel physical properties such as strongly correlated effects, anisotropic transport, and topologically non-trivial energy states.

The BN/graphene/CrOCl heterostructure was assembled through dry transfer method, and the optical image is shown in Fig. 1(c). Owing to its sensitivity to electronic structure, Raman spectroscopy was employed to probe interfacial interactions, particularly the mechanism of the insulating state in heterostructure. Figure 1(d) and Fig. S1 in the Electronic Supplementary Material (ESM) display the Raman spectra of graphene, CrOCl, BN, and CrOCl/BLG/BN heterostructure. Apparently, a typical Raman mode of BN is located at 1365 cm^{-1} , corresponding to the E_{2g} mode. For CrOCl, there exists three characteristic modes of A_g^1 , A_g^2 , and A_g^3 , locating at 208.4, 414.8, and 455.9 cm^{-1} , respectively. In the high-frequency region, two prominent Raman peaks are observed at 1580 and 2700 cm^{-1} , corresponding to the G mode and 2D mode, respectively. Characteristic Raman peaks associated with BN and CrOCl were simultaneously observed, indicating the successful formation of the CrOCl/BLG/BN heterostructure. As shown in Figs. S1(b)–S1(d) in the ESM, the G band of the bilayer graphene/CrOCl heterostructure exhibits a distinct blue shift (1.87 cm^{-1}) compared to graphene's Raman spectrum, attributed to the doping effects induced by BN and CrOCl. Additionally, the G peak positions across different samples exhibit a linear trend, indicating a pronounced hole doping effect at the heterointerface (Fig. S1(d) in the ESM). Notably, CrOCl encapsulation causes a significant attenuation in the G band intensity of bilayer graphene, with the I_{2D}/I_G ratio decreasing markedly from 4.8 to 3.1, confirming efficient charge transfer between graphene and CrOCl. The I_{2D}/I_G ratio is a signature metric for graphene p-type doping. Undoped graphene exhibits a high I_{2D}/I_G due to the strong double-resonant scattering of the 2D band. Upon hole doping (from electron transfer to CrOCl), the Fermi level shifts downward, filling valence band states, enhancing G-band intensity (Pauli exclusion blocks phonon decay) and suppresses 2D-band scattering (reduced double-resonant pathways), leading to a dramatic I_{2D}/I_G reduction. As shown in Fig. S2 in the ESM, the Raman spectra of different samples all exhibit peak position stability, indicating that the heterojunctions obtained via the PVB-assisted one-step transfer technique possess interface-free interfaces.

To demonstrate the effective control of heterojunction interface cleanliness achieved by the PVB-assisted one-step transfer technology, the material surfaces of heterojunctions fabricated by both techniques were characterized in detail. As shown in the optical images and atomic force microscopy (AFM) maps in Figs. 2(a) and 2(c), and Fig. S3 in the ESM, among randomly

constructed CrOCl/BLG/BN heterojunctions, the one-step transfer technology yielded heterojunctions with uniform surface roughness (0.778 nm for one-step transfer and 1.83 nm for multi-step transfer), whereas the multi-step technology produced heterojunctions with significantly higher surface roughness and numerous high points caused by colloidal residue. To further visually compare local potential changes at the CrOCl/bilayer graphene interface, CrOCl/BLG/BN heterojunctions were constructed using mechanically exfoliated CrOCl with a thickness of approximately 23 nm . As is well known, interface trap states are the root cause of charge scattering and device instability, severely affecting the carrier mobility and electrical stability of devices. As shown in Figs. 2(b) and 2(d), the surface potential difference between CrOCl and BLG in the one-step transfer heterojunction is 145 mV , which is higher than the surface potential difference (40 mV) observed in the multi-step transfer heterojunction. This indicates that the CrOCl/graphene interface in the one-step transfer heterojunction exhibits stronger electronic coupling, potentially enabling CrOCl to more effectively modulate the carrier concentration and band structure of graphene via electric fields. Moreover, potential changes reveal a systematic upward shift in contact potential difference (CPD) for both one-step and multi-step transfer samples relative to pristine graphene, confirming a downward shift of Fermi level toward the valence band maximum. This can be identified as p-type hole doping driven by interfacial electron transfer from graphene to CrOCl. This result is consistent with Raman's results (G-band blue shift and reduced I_{2D}/I_G ratio). As seen in Fig. 2(b), the surface potential distribution in the one-step transfer heterojunction is highly uniform, reducing carrier scattering and enhancing interfacial electrical stability. Furthermore, cross-sectional scanning transmission electron microscopy (STEM) analysis was employed to investigate the atomic-level interfaces between CrOCl and bilayer graphene in the heterojunctions prepared by both techniques. As shown in Fig. 2(e), CrOCl atoms in the CrOCl/BLG/BN heterojunction prepared by the one-step technology exhibit perfect alignment with bilayer graphene atoms, indicating virtually no impurities. In contrast, the CrOCl atoms in the CrOCl/BLG/BN heterojunction prepared via the multi-step technology (Fig. 2(i)) exhibit a significant spacing (exceeding 2 nm) from the bilayer graphene atoms. Figures 2(f)–2(h) and 2(j)–2(l) show the energy dispersive X-ray spectroscopy (EDS) distribution maps of heterojunctions prepared by the two techniques, respectively. The carbon distribution range of the one-step-prepared heterojunction is significantly narrower than that of the multi-step-prepared heterojunction, confirming that the one-step transfer technology suffers less impurity contamination than the multi-step transfer technology. Microstructural analysis indicates that the PVB-assisted one-step transfer technology effectively avoids colloidal residue and impurity introduction during heterojunction assembly, enabling seamless van der Waals bonding. This provides crucial support for studying charge transfer between two-dimensional heterojunction materials.

To compare the impact of CrOCl interface cleanliness on the performance of bilayer graphene devices, BN/BLG/CrOCl heterojunctions were fabricated using both PVB-assisted one-step transfer and multi-step transfer techniques to investigate the advantages of the one-step transfer technique in heterojunction device fabrication. Figure 3(a) shows a schematic diagram of the BN/BLG/CrOCl heterojunction device. In this device structure, the use of blue tape exfoliation to prepare two-dimensional single

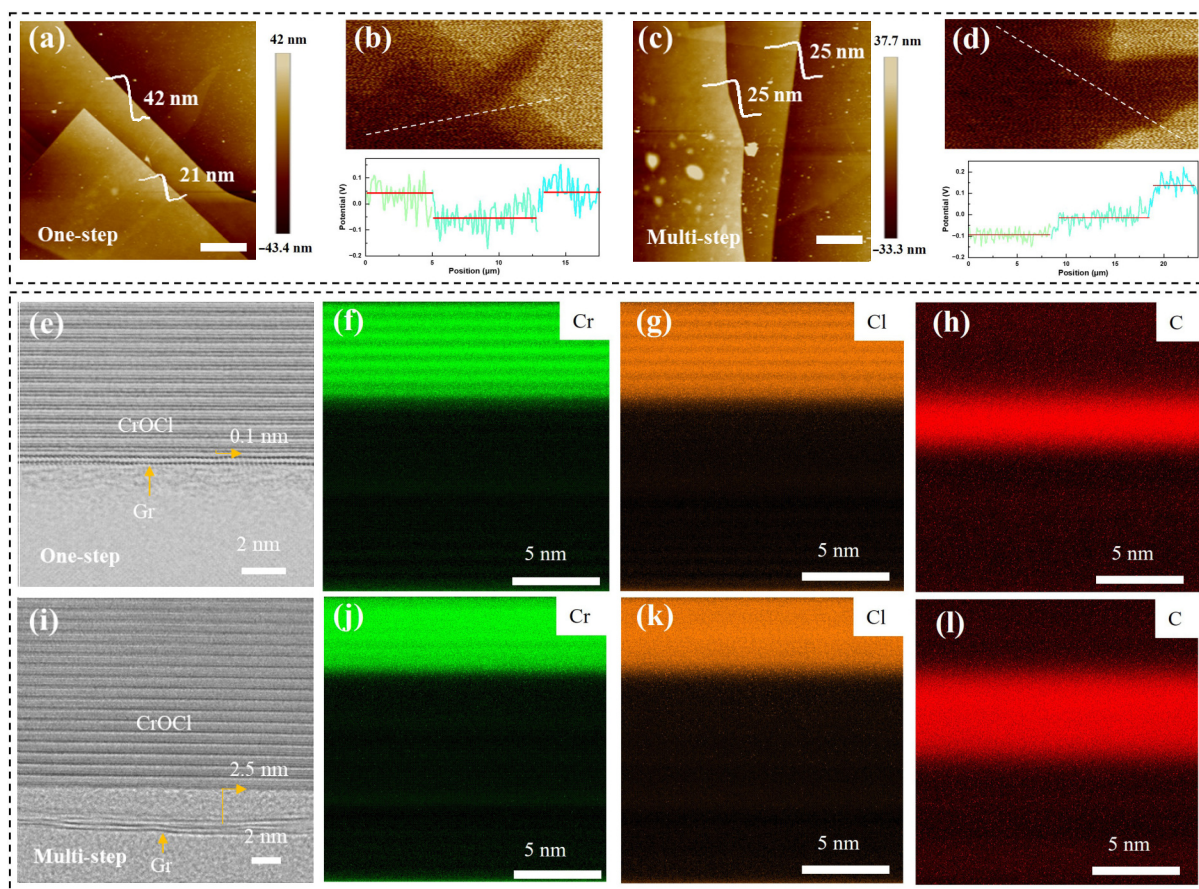


Figure 2 Microscopic morphological characterization of the interface quality of heterojunctions fabricated by different transfer techniques. (a) AFM image of the heterojunction prepared by the one-step transfer technology. (b) Surface potential distribution of the heterojunction prepared by the one-step transfer technology. (c) AFM image of the heterojunction prepared by the multi-step transfer technology. (d) Surface potential distribution of the heterojunction prepared by the multi-step transfer technology. ((e)–(h)) Atomic-level interface characterization of the heterojunction prepared by the one-step transfer technology: (e) STEM image, (f) EDS spectrum of Cr, (g) EDS spectrum of Cl, and (h) EDS spectrum of C. ((i)–(l)) Atomic-level interface characterization of the heterojunction prepared by the multi-step transfer technology: (i) STEM image, (j) EDS spectrum of Cr, (k) EDS spectrum of Cl, and (l) EDS spectrum of C.

crystals results in polymer residue on the upper surface of CrOCl. Within the BN/BLG/CrOCl heterojunction device, the one-step technology yields a cleaner lower surface of the bilayer graphene compared to the multi-step technology. As shown in Fig. 3(b), devices fabricated using the multi-step transfer technology exhibit poorer performance. The back-gate (CrOCl) exerts minimal influence on the top gate (BN) regulation of the bilayer graphene, resulting in a maximum on/off ratio of only 10^2 under dual-gate cooperative control (Fig. 3(c) and Fig. S4(a) in the ESM). In contrast, devices fabricated using the one-step transfer technology exhibit significant CrOCl-mediated control over vertical electrical transport, as shown in Fig. 3(d). This initially moderate on/off ratio may correspond to a regime with a relatively small vertical displacement field (D) applied by the dual gates, leading to a narrowing of the electronic bandgap in the bilayer graphene. As depicted in Fig. 3(e) and Fig. S4(b) in the ESM, the on/off ratio of the devices progressively increases with rising back-gate voltage, reaching up to 10^4 . The extracted on/off ratio in Fig. 3(f) clearly distinguishes the two approaches and highlights the superiority of the one-step transfer strategy. Figures S5 and S6 in the ESM compare the hysteresis and leakage current for heterostructures (BN as bottom gate) fabricated by one-step and multi-step transfer. As demonstrated by the electrical data of graphene devices in the multi-step technology, the presence of impurities, contaminants, or

residual support material on the bottom surface of graphene during transfer introduces additional scattering centers. These scattering centers impede carrier transport within graphene, reducing the electrical conductivity of device. On the other hand, an unclear transfer interface compromises its flatness and continuity, weakening the van der Waals coupling between CrOCl and the bilayer graphene. This may further diminish CrOCl's electronic modulation effect on graphene, making it difficult to achieve a significant on-off ratio. To achieve a higher on-off ratio, it is essential to minimize charge inhomogeneities and scattering phenomena at the interface-polymer contaminants reduce the minimum conductivity in the off state. These results demonstrate that the PVB-assisted one-step transfer technology facilitates bandgap opening in bilayer graphene, thereby endowing BN/BLG/CrOCl heterojunction devices with superior valve control capability.

To evaluate the impact of the PVB-assisted one-step transfer technology on the heterojunction electrical transport properties while maintaining high surface cleanliness on CrOCl, we fabricated double-gate bilayer graphene field-effect transistors (FETs) using graphite electrodes. Figure 4(a) shows the optical image and schematic diagram of the CrOCl/BLG/BN heterojunction device, respectively. As shown in Fig. 4(b), the output curve at 80 K indicates that the open-state current of the bilayer graphene reaches

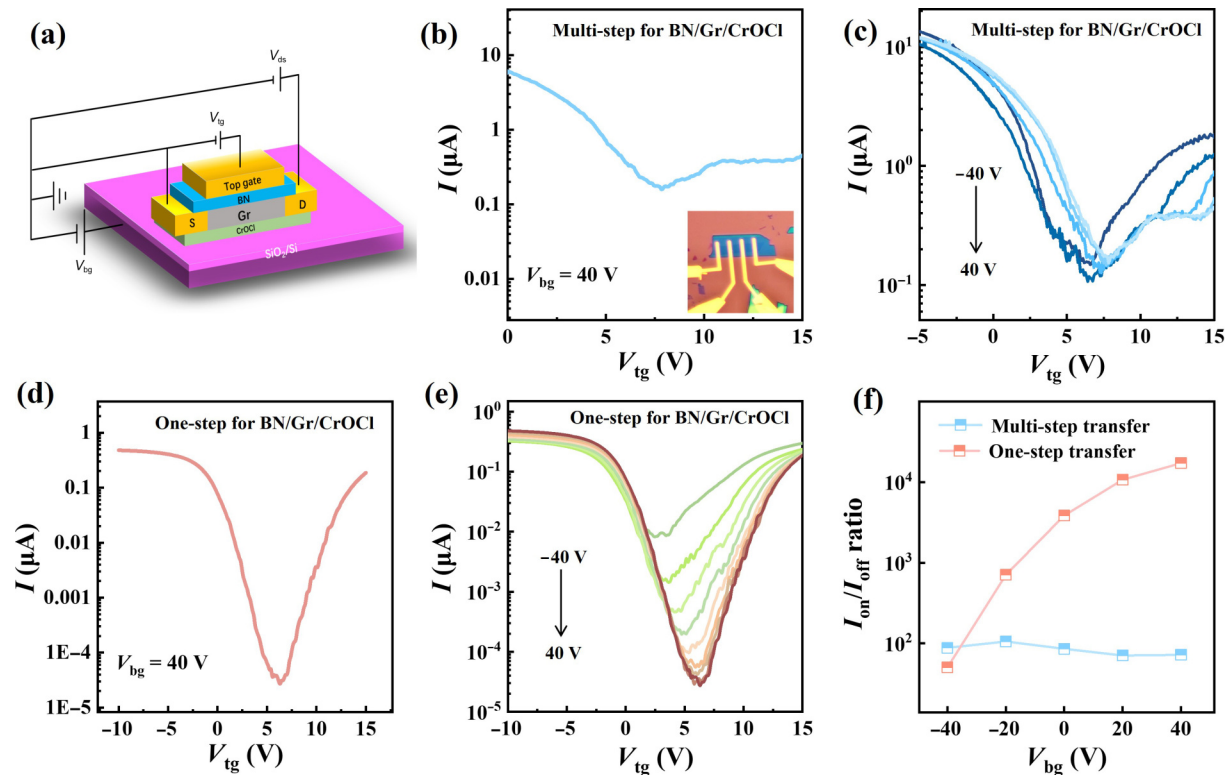


Figure 3 Electrical characterization of BN/graphene/CrOCl heterojunction. (a) Schematic diagram of the device structure. (b) Transfer characteristic curve of the device fabricated by the multi-step transfer technology, with an optical image inset. (c) V_{bg} -dependent top gate transfer characteristic curves of the device fabricated by the multi-step transfer technology: $V_{bg} = -40, -20, 0, 20$, and 40 V. (d) Transfer characteristics curve of the device fabricated by the one-step transfer technology. (e) V_{bg} -dependent top gate transfer characteristics curve of the device fabricated by the one-step transfer technology: $V_{bg} = -40, -20, 0, 20$, and 40 V. (f) On/off ratio variation with V_{bg} for different devices fabricated by the two techniques at different V_{bg} values.

up to $1 \mu\text{A}$ under no gate voltage conditions. Transfer curves (Fig. 4(c)) reveal that at $V_{ds} = 0.01$ V and without a back gate, the device exhibits distinct p-type dominant bipolar transport characteristics under a small electric field applied to the top gate (CrOCl). Figures S7(a) and S7(b) in the ESM display the output transfer characteristics of device at lower temperatures. It is evident that CrOCl exhibits stronger bipolar electrical control over the bilayer graphene, significantly enhancing electron transport capability ($T = 15$ K). In contrast, BN primarily modulates hole transport in bilayer graphene, requiring substantial gate voltage to induce polarity reversal (Figs. S7(c) and S7(d) in the ESM). This difference likely stems from CrOCl's higher dielectric constant compared to BN, coupled with its stronger interfacial coupling with bilayer graphene. To achieve substantial gate control over bilayer graphene with smaller gate voltages, we employed dual-gate simultaneous control in the device. As shown in Fig. 4(d), under conditions of $V_{bg} = 90$ V and $V_{ds} = 0.01$ V, CrOCl exhibits stronger bipolar modulation capability and a high on/off ratio. Moreover, the excellent leakage current performance of CrOCl in Fig. S8 in the ESM demonstrates its superior gate control capability as a gate dielectric. Further investigations reveal that both lower temperatures and higher gate voltages favor the enhancement of the on/off ratio, with the device achieving a ratio as high as 10^8 (Fig. 4(e)). Additionally, as shown in Fig. 4(f), when the gate voltage V_{bg} increases from -40 to 60 V, the on/off current ratio of the device fabricated by the one-step transfer technology varies between 1.16 and 10^8 , demonstrating the unique transport capabilities of this bilayer graphene heterojunction device. To investigate the dual-gate

controllability of heterostructure device, we characterized its dual-gated transfer out curve using color mapping, as shown in Figs. 4(g) and 4(h). Under dual-gate modulation with BN top gate and CrOCl/SiO₂ back gate, increasing the back-gate voltage elevates the on/off ratio from 1.12 to 10^8 at a bias voltage of 0.01 V. When the bias voltage increases to 0.1 V, no significant modulation of the on/off ratio is observed in heterostructure prepared by the one-step transfer technology, indicating the bias-voltage-independent transport characteristics. The exceptional on/off ratio (up to 10^8) demonstrates the comparable performance to transition metal dichalcogenides (TMD) such as MoS₂, highlighting the potential for both electrical and optoelectrical applications. In contrast, the dual-gated bilayer graphene with a graphite electrode, fabricated via the multi-step dry transfer (Figs. S9 and S10 in the ESM), exhibited unexpected transfer characteristic, featuring a relatively high off-state current of $0.5 \mu\text{A}$ and a consequently degraded on-off ratio. Notably, the transfer output characteristics remained essentially unchanged with varying back-gate voltage. Additionally, Fig. 4(i) demonstrates that this double-gate and bilayer graphene device exhibits stable and high on/off ratios under various bias conditions. The exceptionally high on/off ratio in dual-gated bilayer graphene originates from symmetry breaking induced by charge transfer at the heterointerface. This strong interlayer charge transfer dramatically modulates the electronic band structure, allowing for precise control of the transport properties. Therefore, the PVB-assisted one-step transfer technology can effectively control interface cleanliness to optimize heterojunction quality, thereby achieving superior electrical transport properties.

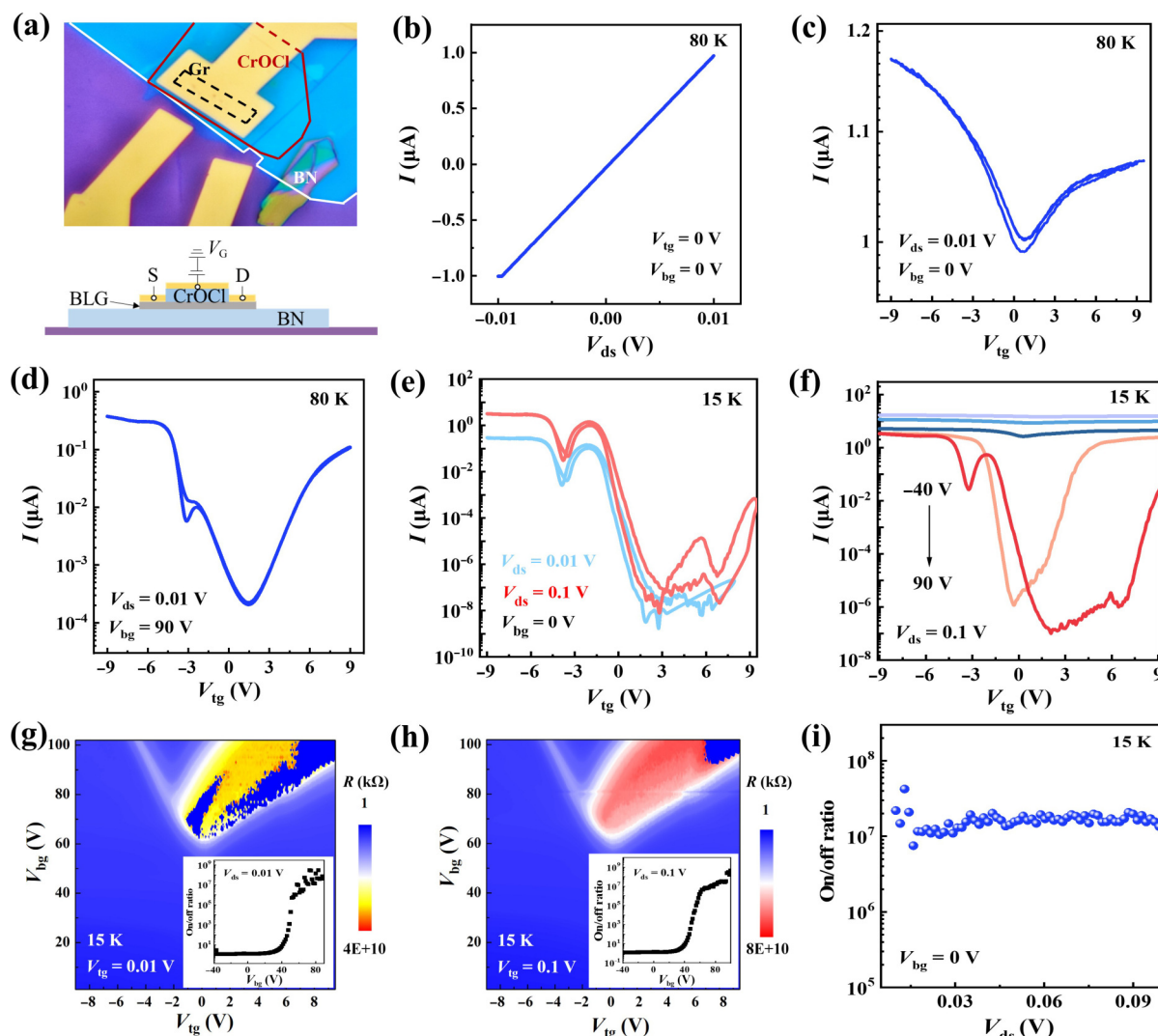


Figure 4 Electrical characterizations of BN/graphene/CrOCl heterostructure by one-step transfer technology. (a) Optical and device schematic image of heterostructure. (b) The drain source current–drain source voltage (I_{ds} – V_{ds}) output curve at 80 K. (c) The drain source current–drain source voltage (I_{ds} – V_g) transfer curve at 80 K ($V_{ds} = 0.01$ V). (d) The drain source current–gate source voltage (I_{ds} – V_g) transfer curve at 80 K with a back-gate voltage of 90 V ($V_{ds} = 0.01$ V). (e) Top gate transfer curves at 15 K with back-gate voltage of 90 V, for bias voltages of 0.1 and 0.01 V. (f) The drain source current–gate source voltage (I_{ds} – V_g) transfer characteristic curve at 15 K with back-gate voltage varying from –40 to 90 V ($V_{ds} = 0.01$ V). (g) The dual-gate modulation dispersion diagram at 15 K ($V_{ds} = 0.01$ V), and the inset shows the corresponding on/off ratio. (h) The dual-gate modulation dispersion diagram at 15 K ($V_{ds} = 0.01$ V), and the inset shows the corresponding on/off ratio. (i) Maximum on/off ratio of the device under dual-gate modulation at different bias voltages.

3 Conclusions

In conclusion, we demonstrate that PVB-assisted one-step transfer enables the fabrication of ultraclean BN/BLG/CrOCl heterostructures with exceptional interface quality. The resulting devices exhibit a remarkable on/off ratio exceeding 10^8 , reflecting highly efficient interfacial charge transfer and precise control over the electronic states in bilayer graphene. Compared with conventional multi-step transfers, this approach minimizes polymer residues and suppresses interfacial defects, directly enhancing device performance and reproducibility. Our results highlight the decisive role of interface cleanliness in governing the electronic properties of graphene-based heterostructures and establish the PVB-assisted one-step transfer as a robust strategy for assembling high-performance van der Waals heterojunctions. These insights provide a valuable foundation for the design and integration of scalable and vertically stacked 2D electronic devices.

4 Methods

4.1 Materials preparation

2D material crystals (including hexagonal BN, graphene, and CrOCl) prepared by chemical vapor transport (CVT) method were purchased from Shanghai Onway Technology Co., Ltd., China. Heterostructure was fabricated via PVB-assisted transfer technique. Firstly, 2D materials were mechanically exfoliated from the CVT crystal and transferred onto P-doped Si substrate with 300 nm SiO_2 . The thicknesses of graphene, BN, and CrOCl were first preliminarily determined by an optical microscope (Nikon ECLIPSE LV 150NL, Japan) and subsequently verified through AFM. Using three-dimensional (3D) micromanipulation equipment for precise alignment, the exfoliated graphene and BN flakes were transferred onto the CrOCl flake surface with the assistance of a PVB film. The PVB film was removed by IPA solution at 50 °C.

First, 3 g of PVB purchased from Aladdin was dissolved in 100 mL of ethanol or isopropanol. The mixture was heated on a hot plate at 50 °C to achieve complete dissolution. Subsequently, the as-prepared PVB solution was cast onto a silicon wafer, followed by being kept 12 h at room temperature. In this way, the desired PVB film was successfully obtained. The above detailed preparation method of PVB film was added in the revised manuscript for clarity and completeness.

4.2 Fabrication of heterostructure device

The heterostructure device was fabricated by standard micro-nano processing technology. After spin-coated PMMA onto the target sample at 4000 rpm for 60 s, device patterning was achieved by electron-beam exposure at 10 kV. Ohmic and gate electrodes were deposited via e-beam evaporation, with Cr (5 nm) serving as an adhesion layer and Au (50 nm) as the conductive layer. Finally, liftoff was executed in heated acetone at 60 °C for 10 min, stripping residual PMMA and excess metal to yield patterned electrodes.

4.3 Characterizations and measurements

Raman spectra were collected using a confocal micro-Raman spectrometer system (Witec Alpha 300R) that was configured with an objective (100X, NA: 0.9). The excitation wavelength and excitation power were 532 nm and 1 mW, respectively. The electrical transport measurements were performed by a cryogenic probe station integrated with a precision source-meter in a high vacuum chamber (10^{-8} Torr). AFM was conducted to characterize heterostructure's morphology and thickness. Transmission electron microscopy samples for cross-sectional imaging were fabricated with a dual-beam focused ion beam (FIB) system (JEOL JEM-F200). High-angle annular dark-field STEM (HAADF-STEM, Titan Cubed Themis G2300) was used to capture the atomic structure of heterostructure interface.

Electronic Supplementary Material: Supplementary material (Raman spectra analysis of various graphene heterojunction samples (Figs. S1–S2), optical and AFM morphological characterization of heterojunctions prepared by one-step and multi-step transfer techniques (Fig. S3), I_{ds} – V_{g} curves, hysteresis characteristics, leakage current, output, and transfer curves under varied temperatures and gate configurations (Figs. S4–S10)) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94908726>.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

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Declaration of competing interests

All the contributing authors report no conflict of interests in this work.

Author contribution statement

S. Q. Q., B. Z., and G. P. conceived the research and designed the experiments. Y. H. W. performed the experiments. J. Z. wrote the paper and performed the experimental analysis. Y. H. W. and X. Y. L. carried out the device fabrication and electrical measurements. F. H., D. K. D., J. X. Z., W. F. T., Y. W. Z., and W. L. contributed to discussions and data analysis. All the authors have approved the final manuscript.

Use of AI statement

None.

References

- [1] Zhao, B.; Wan, Z.; Liu, Y.; Xu, J. Q.; Yang, X. D.; Shen, D. Y.; Zhang, Z. C.; Guo, C. H.; Qian, Q.; Li, J. et al. High-order superlattices by rolling up van der Waals heterostructures. *Nature* **2021**, *591*, 385–390.
- [2] Pham, P. V.; Bodepudi, S. C.; Shehzad, K.; Liu, Y.; Xu, Y.; Yu, B.; Duan, X. F. 2D heterostructures for ubiquitous electronics and optoelectronics: Principles, opportunities, and challenges. *Chem. Rev.* **2022**, *122*, 6514–6613.
- [3] Guo, D. L.; Fu, Q.; Zhang, G. T.; Cui, Y. Y.; Liu, K. Y.; Zhang, X. L.; Yu, Y. L.; Zhao, W. W.; Zheng, T.; Long, H. R. et al. Composition modulation-mediated band alignment engineering from type I to type III in 2D vdW heterostructures. *Adv. Mater.* **2024**, *36*, 2400060.
- [4] Castellanos-Gomez, A.; Duan, X. F.; Fei, Z.; Gutierrez, H. R.; Huang, Y.; Huang, X. Y.; Quereda, J.; Qian, Q.; Sutter, E.; Sutter, P. van der Waals heterostructures. *Nat. Rev.* **2022**, *2*, 58.
- [5] Geim, A. K.; Grigorieva, I. V. van der Waals heterostructures. *Nature* **2013**, *499*, 419–425.
- [6] Li, C. S.; Xu, F.; Li, B. H.; Li, J. Y.; Li, G. A.; Watanabe, K.; Taniguchi, T.; Tong, B. B.; Shen, J.; Lu, L. et al. Tunable superconductivity in electron- and hole-doped Bernal bilayer graphene. *Nature* **2024**, *631*, 300–306.
- [7] Lu, Z. G.; Han, T. H.; Yao, Y. X.; Reddy, A. P.; Yang, J. X.; Seo, J.; Watanabe, K.; Taniguchi, T.; Fu, L.; Ju, L. Fractional quantum anomalous hall effect in multilayer graphene. *Nature* **2024**, *626*, 759–764.
- [8] Wu, W. J.; Luo, S. S.; Li, T.; Zhai, E. Z.; Wang, Z. Y.; Li, X. Y.; Han, Y. M.; Lin, Y. C.; Zhao, Y. J.; Kono, J. et al. Manufacturing chip-scale 2D monolayer single crystals through wafer-bonder-assisted transfer. *Nano Lett.* **2025**, *25*, 14395–14403.
- [9] He, Y. Y.; Lv, Z. X.; Liu, Z. C.; Yang, M. J.; Ai, W.; Chen, J. B.; Chen, W. Y.; Wang, B.; Fu, X. W.; Luo, F. et al. Sacrifice-layer-free transfer of wafer-scale atomic-layer-deposited dielectrics and full-device stacks for two-dimensional electronics. *Nat. Commun.* **2025**, *16*, 5904.
- [10] Zheng, X. X.; Han, L.; Fatima, S.; Khan, S.; Sun, Y.; Li, Z. H.; Ning,

- Y. F.; Leifer, K.; Zhu, G. C.; Li, H. et al. Large-scale and ultraclean dry transfer of two-dimensional materials via liquid nitrogen-assisted cryogenic exfoliation. *Nano Lett.* **2025**, *25*, 9967–9975.
- [11] Zhang, X. C.; Zhou, L. Y.; Wang, S. P.; Li, T.; Du, H. Y.; Zhou, Y. C.; Liu, J. Y.; Zhao, J. J.; Huang, L. F.; Yu, H. et al. Se-mediated dry transfer of wafer-scale 2D semiconductors for advanced electronics. *Nat. Commun.* **2025**, *16*, 4468.
- [12] Liu, J. Y.; Zhao, J. J.; Li, T.; Ji, D. P.; Dai, L. Y.; Li, L.; Wei, Z.; Li, J. W.; Wang, Q.; Yu, H. et al. Direct bonding and debonding of two-dimensional semiconductors. *Nat. Electron.* **2025**, *8*, 1038–1045.
- [13] Zhao, Y. X.; Song, Y. Q.; Hu, Z. N.; Wang, W. D.; Chang, Z. H.; Zhang, Y.; Lu, Q.; Wu, H. Y.; Liao, J. H.; Zou, W. T. et al. Large-area transfer of two-dimensional materials free of cracks, contamination and wrinkles via controllable conformal contact. *Nat. Commun.* **2022**, *13*, 4409.
- [14] Lu, Z. Y.; Chen, Y.; Dang, W. Q.; Kong, L. G.; Tao, Q. Y.; Ma, L. K.; Lu, D. L.; Liu, L. T.; Li, W. Y.; Li, Z. W. et al. Wafer-scale high- κ dielectrics for two-dimensional circuits via van der Waals integration. *Nat. Commun.* **2023**, *14*, 2340.
- [15] Zheng, X. D.; Wang, J. T.; Jiang, J. F.; Zhang, T. Y.; Zhu, J. D.; Dang, T.; Wu, P.; Lu, A. Y.; Chen, D. R.; Yang, T. H. et al. Electrostatic-repulsion-based transfer of van der Waals materials. *Nature* **2025**, *645*, 906–914.
- [16] Quellmalz, A.; Wang, X. J.; Sawallich, S.; Uzlu, B.; Otto, M.; Wagner, S.; Wang, Z. X.; Precht, M.; Hartwig, O.; Luo, S. W. et al. Large-area integration of two-dimensional materials and their heterostructures by wafer bonding. *Nat. Commun.* **2021**, *12*, 917.
- [17] Nakatani, M.; Fukamachi, S.; Solís-Fernández, P.; Honda, S.; Kawahara, K.; Tsuji, Y.; Sumiya, Y.; Kuroki, M.; Li, K.; Liu, Q. et al. Ready-to-transfer two-dimensional materials using tunable adhesive force tapes. *Nat. Electron.* **2024**, *7*, 119–130.
- [18] Yuan, G. W.; Liu, W. L.; Huang, X. L.; Wan, Z. H.; Wang, C.; Yao, B.; Sun, W. J.; Zheng, H.; Yang, K. H.; Zhou, Z. J. et al. Stacking transfer of wafer-scale graphene-based van der Waals superlattices. *Nat. Commun.* **2023**, *14*, 5457.
- [19] Xia, F. N.; Farmer, D. B.; Lin, Y. M.; Avouris, P. Graphene field-effect transistors with high on/off current ratio and large transport band gap at room temperature. *Nano Lett.* **2010**, *10*, 715–718.
- [20] Wang, Y. N.; Gao, X.; Yang, K. N.; Gu, P. F.; Lu, X.; Zhang, S. H.; Gao, Y. C.; Ren, N. J.; Dong, B. J.; Jiang, Y. H. et al. Quantum Hall phase in graphene engineered by interfacial charge coupling. *Nat. Nanotechnol.* **2022**, *17*, 1272–1279.
- [21] Yang, K. N.; Gao, X.; Wang, Y. N.; Zhang, T. Y.; Gao, Y. C.; Lu, X.; Zhang, S. H.; Liu, J. P.; Gu, P. F.; Luo, Z. P. et al. Unconventional correlated insulator in CrOCl-interfaced Bernal bilayer graphene. *Nat. Commun.* **2023**, *14*, 2136.
- [22] Geisenhof, F. R.; Winterer, F.; Seiler, A. M.; Lenz, J.; Xu, T. Y.; Zhang, F.; Weitz, R. T. Quantum anomalous Hall octet driven by orbital magnetism in bilayer graphene. *Nature* **2021**, *598*, 53–58.
- [23] Choi, Y.; Kim, H.; Peng, Y.; Thomson, A.; Lewandowski, C.; Polski, R.; Zhang, Y. R.; Arora, H. S.; Watanabe, K.; Taniguchi, T. et al. Correlation-driven topological phases in magic-angle twisted bilayer graphene. *Nature* **2021**, *589*, 536–541.
- [24] Yu, Y.; Zhang, K. D.; Parks, H.; Babar, M.; Carr, S.; Craig, I. M.; Van Winkle, M.; Lyssenko, A.; Taniguchi, T.; Watanabe, K. et al. Tunable angle-dependent electrochemistry at twisted bilayer graphene with moiré flat bands. *Nat. Chem.* **2022**, *14*, 267–273.
- [25] Seiler, A. M.; Geisenhof, F. R.; Winterer, F.; Watanabe, K.; Taniguchi, T.; Xu, T. Y.; Zhang, F.; Weitz, R. T. Quantum cascade of correlated phases in trigonally warped bilayer graphene. *Nature* **2022**, *608*, 298–302.
- [26] Dey, A.; Cottam, N.; Makarovskiy, O.; Yan, W. J.; Mišekis, V.; Coletti, C.; Kerfoot, J.; Korolkov, V.; Eaves, L.; Linnartz, J. F. et al. Thermally stable quantum Hall effect in a gated ferroelectric-graphene heterostructure. *Commun. Phys.* **2023**, *6*, 216.
- [27] Liu, C.; Wang, X. Z.; Shen, C.; Ma, L. P.; Yang, X. Q.; Kong, Y.; Ma, W.; Liang, Y.; Feng, S.; Wang, X. Y. et al. A hot-emitter transistor based on stimulated emission of heated carriers. *Nature* **2024**, *632*, 782–787.
- [28] Hu, B. Q.; Xie, X.; Chen, J. Y.; Li, S. F.; He, J.; Liu, Z. W.; Wang, J. T.; Liu, Y. P. Symmetry breaking and magnetically tuned anisotropic behavior in WSe₂/CrOCl/ReSe₂ heterostructures. *Nano Res.* **2024**, *18*, 94907478.
- [29] Zhu, H. R.; Shao, B.; Zuo, X. Enhancing ferromagnetic coupling in CrXY (X = O, S, Se; Y = Cl, Br, I) monolayers by turning the covalent character of Cr-X bonds. *npj Comput. Mater.* **2023**, *9*, 56.
- [30] Tang, Y.; Ping, Y.; Yang, X. X.; Xing, J. B.; Chen, J. B.; Wang, X.; Lu, J. B.; Jing, H. M.; Liu, K. Q.; Wu, J. X. et al. Synthesis of highly anisotropic 2D insulator CrOCl nanosheets for interfacial symmetry breaking in isotropic 2D semiconductors. *Adv. Mater.* **2025**, *37*, 2405358.
- [31] Yang, Y.; Zhou, Y.; Tang, Z. M.; Liu, Y. L.; Quan, W. M.; Zhou, J.; Li, X. K.; Xi, X. X.; Gong, Q. H.; Zhang, L. F. et al. Unusual thermal transport in few-layer van der Waals antiferromagnet CrOCl. *Adv. Sci.* **2025**, *12*, 2502440.
- [32] Ding, Y. F.; He, J. D.; Zhang, S. H.; Zuo, H. K.; Gu, P. F.; Cai, J. J.; Zeng, X. H.; Yan, P.; Cai, J.; Cao, K. C. et al. Constructing the Fulde–Ferrell–Larkin–Ovchinnikov state in a CrOCl/NbSe₂ van der Waals heterostructure. *Nano Lett.* **2024**, *24*, 12814–12822.
- [33] Guo, Y. M.; Li, J. X.; Zhan, X. P.; Wang, C. W.; Li, M.; Zhang, B.; Wang, Z. R.; Liu, Y. Y.; Yang, K. N.; Wang, H. et al. van der Waals polarity-engineered 3D integration of 2D complementary logic. *Nature* **2024**, *630*, 346–352.
- [34] Guo, D.; Li, W. J.; Gu, P. F.; Dong, W. K.; Rui, X. Y.; Watanabe, K.; Taniguchi, T.; Ye, Y.; Zheng, F. W.; Zhou, J. D. et al. A van der Waals broadband infrared optical synapse enabling orientation detection. *Adv. Sci.* **2025**, *12*, e07530.
- [35] Ying, B. Y.; Xin, B. J.; Li, M. M.; Zhou, S. Y.; Liu, Q.; Zhu, Z. H.; Qin, S. Q.; Wang, W. H.; Zhu, M. J. Efficient charge transfer in graphene/CrOCl heterostructures by van der Waals interfacial coupling. *ACS Appl. Mater. Interfaces* **2024**, *16*, 43806–43815.



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