

REPORT

2D MAGNETISM

Direct observation of van der Waals stacking-dependent interlayer magnetism

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Controlling the crystal structure is a powerful approach for manipulating the fundamental properties of solids. In van der Waals materials, this control can be achieved by modifying the stacking order through rotation and translation between the layers. Here, we observed stacking-dependent interlayer magnetism in the two-dimensional (2D) magnetic semiconductor chromium tribromide (CrBr₃), which was enabled by the successful growth of its monolayer and bilayer through molecular beam epitaxy. Using in situ spin-polarized scanning tunneling microscopy and spectroscopy, we directly correlate the atomic lattice structure with the observed magnetic order. Although the individual monolayer CrBr₃ is ferromagnetic, the interlayer coupling in bilayer depends on the stacking order and can be either ferromagnetic or antiferromagnetic. Our observations pave the way for manipulating 2D magnetism with layer twist angle control.

The type of van der Waals (vdW) stacking has been extensively recognized as a critical component in determining the properties of layered vdW materials. In particular, weak interlayer vdW interactions allow for the control over the rotational and translational degrees of freedom between layers, creating a host of new materials with distinct stacking symmetries and functionalities (1–3). Whereas most previous work has focused on the electronic and optical properties associated with the vdW stacking (4–7), the recent discovery of magnetism in two-dimensional (2D) materials, achieved through both mechanical exfoliation (8–13) and molecular beam epitaxy (MBE) (14, 15), provides an exciting opportunity to explore the effects of stacking order on a material's magnetic properties.

Among the 2D magnetic materials discovered so far, the family of chromium trihalides, CrX₃ (where X = Cl, Br, or I), has garnered special interest (8, 16). For instance, ferromagnetism perpendicular to the 2D plane persists in monolayer CrI₃, whereas its bilayer exhibits interlayer antiferromagnetism. These magnetic structures lead to a number of emerging phenomena, such as giant tunneling magnetoresistance in spin-filter magnetic

tunnel junctions (17–19), the electrical control of 2D magnetism (20–22), and giant non-reciprocal optical second-harmonic generation (23). In contrast to CrI₃, recent tunneling measurements suggest that the interlayer coupling in atomically thin CrBr₃ is ferromagnetic (24). This was confirmed through polar reflectance magnetic circular dichroism (RMCD) measurements of mechanically exfoliated bilayers of CrBr₃, which reveal magnetic hysteresis behavior centered around zero applied magnetic field, distinct from the three-step staircase pattern observed in bilayer CrI₃ (fig. S1). Given that CrI₃ and CrBr₃ are isostructural (25), such a substantial difference in the magnetic properties between their bilayers prompts a thorough investigation of the mechanisms governing the interlayer magnetic coupling (26).

Here, we used in situ spin-polarized scanning tunneling microscopy and spectroscopy (27, 28) to establish a direct correlation between the interlayer magnetic coupling and the stacking structures in bilayer CrBr₃. The CrBr₃ films were grown on freshly cleaved, highly oriented pyrolytic graphite (HOPG) substrates by MBE. During the growth, the sample surface was monitored in situ by reflection high-energy electron diffraction (RHEED). Figure 1, A and B, respectively, show the RHEED patterns before and after growing CrBr₃ with the diffraction orders indicated. The stripe-like RHEED pattern from the CrBr₃ indicates the formation of a 2D crystalline thin film, confirmed by scanning tunneling microscopy (STM) to be a monolayer CrBr₃ (Fig. 1C). Further deposition of CrBr₃ led to the appearance of bilayer CrBr₃ islands (Fig. 1D) [for details about the MBE growth, see (29)].

Figure 1E shows the atomically resolved STM image of the monolayer CrBr₃, revealing periodically spaced triangular cluster protrusions. To understand this image, we consider the monolayer crystal structure of CrBr₃. As shown in Fig. 1F, Cr atoms are arranged in a honeycomb lattice structure, and each atom is surrounded by an octahedron of six Br atoms (16). Within a single honeycomb formed by six Cr atoms, there are three Br atoms at the top and bottom surfaces, marked by solid and dotted green triangles with opposite orientations, respectively. Thus, the monolayer CrBr₃ has three-fold rotational symmetry, with the rotational axis across either the Cr atoms or the center of the honeycomb. By overlaying the atomic structure on top of the STM image in Fig. 1E, we determined that the observed triangular cluster is formed by the three top Br atoms (solid green triangle in Fig. 1E). The measured in-plane lattice constant is 6.3 Å, consistent with the bulk lattice constant (6.26 Å) (25). The monolayer thickness is determined to be 6.5 Å by atomic force microscopy (AFM), as shown in Fig. 1G. Structural domains and their boundaries are also clearly seen in the image. Both the large-scale topography and the atomically resolved STM images demonstrate the high-quality growth of the monolayer CrBr₃ films.

To measure the magnetic properties, we performed in situ spin-polarized STM measurements with an applied out-of-plane magnetic field at 5 K [for spin-polarized STM measurements, see (29)]. We used antiferromagnetic Cr-coated tungsten tips in which magnetization at the tip apex remained stable while the external magnetic field was swept (30). Figure 2A shows the tunneling spectra (dI/dV) from a monolayer CrBr₃ with oppositely applied fields ($B = \pm 0.3$ T), measured on the center of a Br triangular cluster. Although both dI/dV curves show semiconducting behavior near zero bias voltage, they clearly differ at some bias voltage V_b , which results from the magnetization flip of the monolayer CrBr₃ under the reversal of the external magnetic field. To further confirm the existence of ferromagnetism, we measured a series of dI/dV spectra while sweeping the magnetic field back and forth. Figure 2B plots the hysteresis loop at $V_b = 1.4$ V. Additional data at different bias voltages are shown in fig. S2. A rectangular hysteresis loop was observed with a coercive field of ~30 mT, with the two dI/dV plateaus corresponding to different magnetization states (spin up or spin down). This observation suggests that epitaxial CrBr₃ monolayers grown on HOPG are semiconducting ferromagnets with a well-defined, out-of-plane easy axis. The dI/dV plateaus have sensitive dependence on bias voltages, atomic sites, and Cr tips (see figs. S2 and S3) because the dI/dV signal is determined by the spin- and energy-dependent local density of states of the tip and the sample (29).

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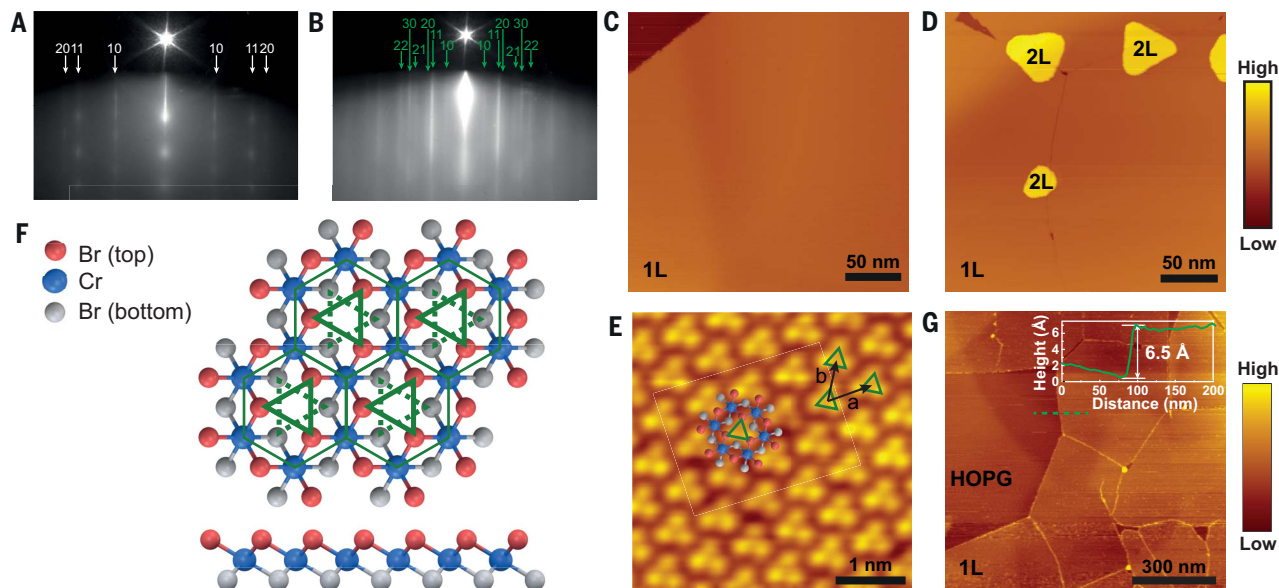
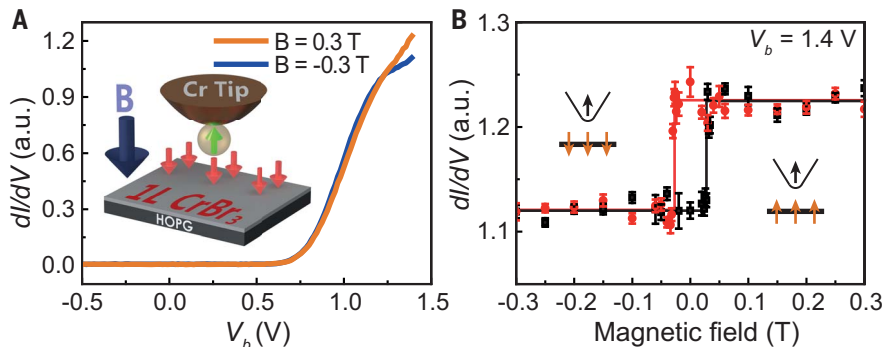


Fig. 1. MBE Growth of CrBr₃ monolayer and bilayer on HOPG. (A and B) RHEED patterns with indicated diffraction orders of (A) the bare HOPG substrate and (B) the MBE-grown CrBr₃ film. (C and D) STM images of (C) the CrBr₃ monolayer with (D) bilayer islands. The scan parameters were as follows: $V_b = 1.1$ V, $I = 100$ pA, $T = 5$ K for (C) and $V_b = 1.5$ V, $I = 100$ pA, $T = 5$ K for (D). (E) Atomically resolved image of a monolayer CrBr₃ with an overlaid atomic structure. The scan parameters were as follows: $V_b = 1.5$ V, $I = 500$ pA, $T = 5$ K. The lattice constants were determined to be 6.3 Å for

the primitive vectors **a** and **b**, consistent with the bulk values. (F) Illustrations of the top and side views of the monolayer CrBr₃ atomic structure. The Cr atoms form a honeycomb lattice sandwiched by Br atoms. Within the Cr honeycomb lattice, the top and bottom surfaces of Br atoms form single triangles but with opposite orientation, indicated by solid and dotted green lines, respectively. (G) AFM image of monolayer CrBr₃ with partial coverage. A line-cut profile across the monolayer and bare substrate is shown with a monolayer height of ~ 6.5 Å.

Fig. 2. Spin-polarized tunneling of monolayer CrBr₃.

(A) Spin-polarized tunneling spectra under positive and negative out-of-plane magnetic fields (± 0.3 T). The inset illustrates the experimental geometry. The magnetization at the tip apex is assumed to be spin up, and the same assumption applies to the following figures [see discussion in (29)]. (B) dI/dV signal as a function of the magnetic field. V_b was fixed at 1.4 V. The out-of-plane magnetic field was swept upward (black data) and downward (red data). The ferromagnetic hysteresis loop is outlined as rectangular solid lines. Insets sketch the two configurations of the magnetization alignment between the Cr tip and the monolayer CrBr₃ film. The in-plane component of magnetization at the Cr tip apex, if any, does not contribute to the magnetic contrast in dI/dV .

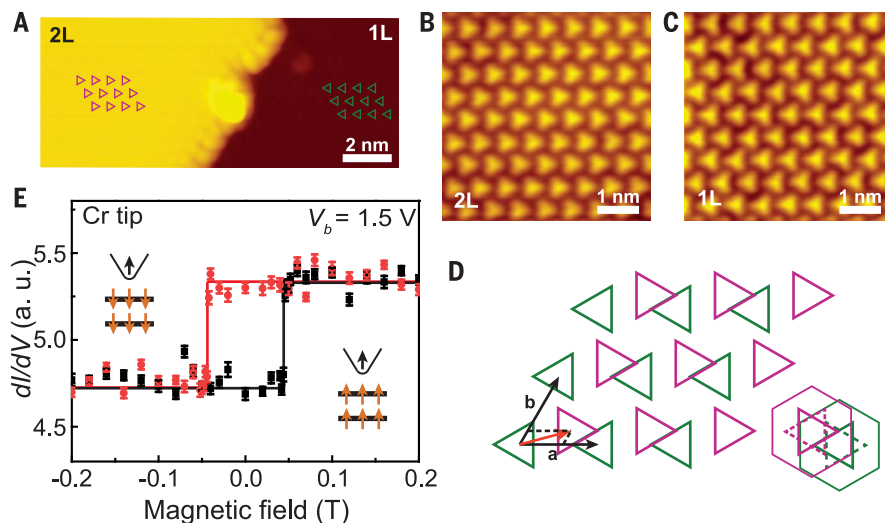


With the atomic structure of monolayer CrBr₃ determined and its ferromagnetism confirmed, we now focus on bilayer CrBr₃. In general, there are two types of stacking structures with the unit cell size the same as that of the monolayer: the R-type, with both layers aligned to the same orientation, and the H-type, with a 180° rotation between the layers. For R-type stacking, two polytypes for CrBr₃ bulk crystals were reported: rhombohedral at low temperature and monoclinic at high temperature (>420 K) (16). These two structures mainly differ in the relative translation between the two monolayers. How-

ever, common to vdW materials (4, 5, 31), the energy differences between different stacking structures can be relatively small, even if the stacking structure is absent in bulk crystal. Therefore, the possible stacking structures in the bilayer case could be far more complex than the two reported in the literature. By only considering the high-symmetry configurations (i.e., hollow, bridge, and top sites) between the halogen atoms at the interface of adjacent layers, there are already six and five polytypes for R-type stacking (table S1) and H-type stacking (table S2), respectively.

We found that the H- and R-type stacking structures can both form in the MBE-grown bilayers and they give rise to distinct interlayer magnetic coupling. The results corresponding to the H-type stacking structure are given in Fig. 3. Figure 3A shows the STM image across the step edge between a bilayer CrBr₃ island and a monolayer CrBr₃ film that is an extension of the bilayer's bottom layer. Their atomically resolved STM images are shown in Fig. 3, B and C, which show individual triangular clusters formed by the top three Br atoms in the unit cell of CrBr₃, similar to those in Fig.

Fig. 3. Interlayer ferromagnetic coupling in an H-type stacked bilayer CrBr₃. (A) STM image of a CrBr₃ film with both a monolayer (1L) region and a bilayer (2L) island. (B and C) Magnified, atomically resolved images of (B) the bilayer region and (C) its extended bottom monolayer at $V_b = 1.9$ V, indicating that the top and bottom layers in the bilayer are anti-aligned, or rotated by 180° (H-type stacking). (D) Atomic structure of the bilayer CrBr₃, as determined from atomically resolved STM images (for further details, see fig. S4). The unit cells of the top and bottom layers are represented by the magenta and green solid triangles, respectively, corresponding to the top surface of Br atoms in each monolayer sheet. These magenta and green solid triangles are also overlaid on the monolayer and bilayer in (A). The unit cell of the top layer (magenta) is a translation by $0.55\mathbf{a} + 0.20\mathbf{b}$ of the bottom layer (green). For comparison with the structures in table S2, the stacking structure is also shown with the bottom surface of Br atoms of each monolayer sheet as dotted triangles and the Cr atoms as solid hexagons. (E) Spin-polarized tunneling on the bilayer CrBr₃ as a function of magnetic field with a Cr tip at $V_b = 1.5$ V. The out-of-plane magnetic field was swept upward (black data) and downward (red data). Like that of the monolayer CrBr₃, a rectangular ferromagnetic hysteresis loop was observed with a coercive field of ~ 45 mT. Insets depict two configurations of the magnetization alignment between the tip and the sample.



IE. The orientation of the triangular clusters from the monolayer region, which has the same crystal structure as the bottom layer of the bilayer, is exactly opposite to the top layer in the bilayer, suggesting H-type stacking.

To determine the translational degree of freedom between the two layers, we overlaid the two CrBr₃ lattices, represented by the magenta and green triangles, on the top and bottom layers of the bilayer island shown in Fig. 3A (for further details, see fig. S4). We found that the top layer is translated to $0.55\mathbf{a} + 0.20\mathbf{b}$ with respect to the bottom layer (Fig. 3D). Given that the experimental uncertainty of translation is $\sim \pm 7\%$ of the lattice constants \mathbf{a} and \mathbf{b} , the closest stacking structure is “bridge I” of H-type stacking listed in table S2. We also observed the equivalent bridge I structure, in which the top layer is translated to $0.76\mathbf{a} + 0.38\mathbf{b}$ of the bottom layer (fig. S5). In terms of both rotational and translational degrees of freedom, this observed stacking structure is distinct from the reported structure models for CrX₃ in the literature (16, 25). Further spin-polarized STM measurement on this H-type stacked bilayer CrBr₃ showed a rectangular magnetic hysteresis loop with a coercive magnetic field of ~ 45 mT (Fig. 3E for $V_b = 1.5$ V and fig. S6 for other bias voltages), similar to the hysteresis observed in CrBr₃ monolayers (Fig. 2B). Thus, in H-type stacked bilayer CrBr₃, the interlayer coupling is ferromagnetic.

The second kind of stacking structure is depicted in Fig. 4, where the triangular clusters from the adjacent monolayer and bilayer regions are oriented in the same direction (Fig. 4, A to C), suggesting R-type stacking. The trans-

lational degree of freedom between the two layers is determined in the same way as in Fig. 3. In R-type stacking, the top layer is translated to $0.48\mathbf{a} + 0.48\mathbf{b}$ with respect to the bottom layer (Fig. 4D; for further analysis details, see fig. S7). In one R-type-stacking sample, we also observed that the top layer was translated to $0.21\mathbf{a} + 0.78\mathbf{b}$ of the bottom layer (fig. S8). Examining their stacking structures, we found that they are close to the structures named “special” and “bridge I” listed in table S1. As illustrated in fig. S9, both structures share the same lattice symmetry (C_{2h}) as the monoclinic structure (16), but with a translation along the mirror plane.

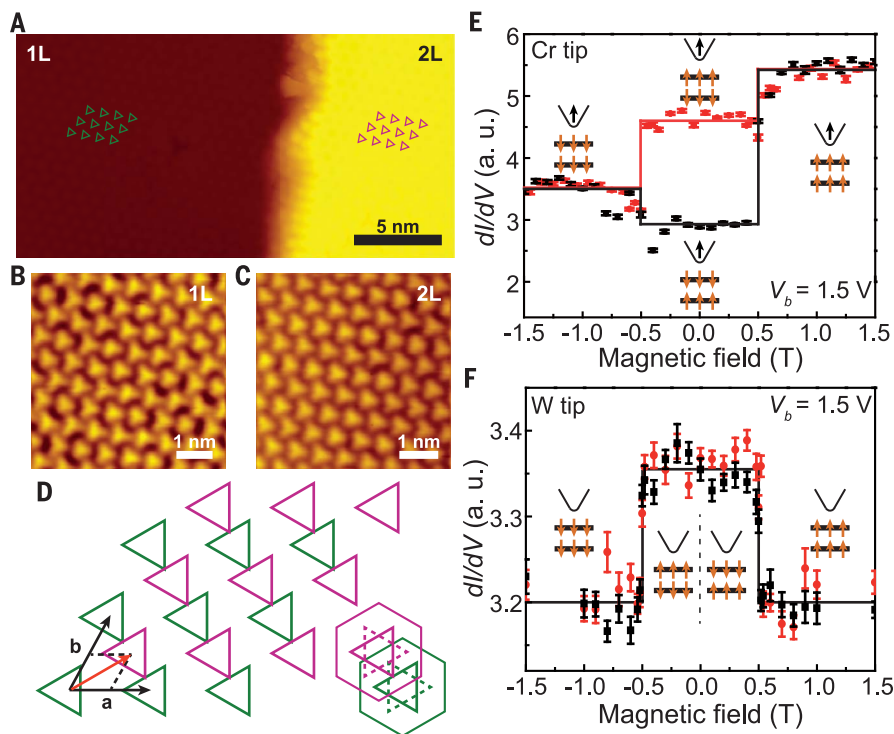
For the R-type stacked bilayer, we observed four plateaus in the hysteresis loop when a Cr tip was used (Fig. 4E and fig. S10), distinct from the two plateaus observed on the CrBr₃ monolayer and the H-type stacked bilayer. This four-plateau behavior suggests that the R-type stacked bilayer is antiferromagnetically coupled in its ground states, giving rise to two additional magnetization configurations. Above $+0.5$ T or below -0.5 T, the magnetization direction of the bilayer CrBr₃ is aligned to the same direction by the external magnetic field. As in the ferromagnetic CrBr₃ monolayer and bilayer, the parallel or antiparallel alignment between the tip and the sample yields two different dI/dV plateaus. Within ± 0.5 T, the individual ferromagnetic layers are antiferromagnetically coupled, which creates an additional magnetic tunnel junction across the vdW gap. Depending on how the spin direction of the top layer aligns with the Cr tip, two new magnetization configurations are formed (inset of Fig. 4E).

The interlayer antiferromagnetism in R-type stacked bilayer CrBr₃ is further corroborated with a nonmagnetic W tip under external magnetic field. Figure 4F shows such a measurement on another R-type stacked bilayer at $V_b = 1.5$ V (see fig. S11 for data with different bias voltages). Starting with zero applied magnetic field, sweeping the external magnetic field in either direction results in a plateau in the dI/dV curve. Beyond magnetic fields of ± 0.5 T, the dI/dV abruptly changes (Fig. 4F). Although the relative magnitude of dI/dV plateaus depends on bias voltage, the two-plateau behavior demonstrates the magnetic field-driven transition from antiferromagnetic to ferromagnetic interlayer coupling. Such tunneling magnetoresistance behavior was recently discovered in spin-filter magnetic tunnel junctions using mechanically exfoliated bilayer CrI₃ (17–19). By contrast, when a nonmagnetic W tip was used for H-type stacked bilayers that were ferromagnetically coupled, no magnetic field-dependent dI/dV features at any bias voltage could be observed (fig. S12). In addition to the dI/dV magnetic contrast, the antiferromagnetic ground state in this R-type stacked bilayer could be captured in the contrast of STM height driven under external magnetic field, as shown in fig. S13.

The distinct interlayer magnetism, from antiferromagnetic coupling in R-type stacking to ferromagnetic coupling in H-type stacking, in our MBE-grown bilayer CrBr₃ clearly demonstrates the substantial tunability of 2D magnetism through the stacking order. The interlayer coupling in bilayer CrBr₃ is mediated through a superexchange interaction, which is controlled by the directional hybridization

Fig. 4. Interlayer antiferromagnetic coupling in an R-type stacked bilayer CrBr₃.

(A) STM image of a CrBr₃ film with both a monolayer (1L) region and a bilayer (2L) island. **(B and C)** Atomically resolved images of (B) the monolayer and (C) the bilayer. $V_b = 1.9$ V. The stacking configuration in the bilayer is identified as R-type, i.e., the top and bottom layers are in the same orientation. **(D)** Atomic structure of the bilayer CrBr₃, as determined from atomically resolved STM images (see fig. S7 for further details). The representation style follows that in Fig. 3. The unit cell of the top layer (magenta) is translated by $0.48\mathbf{a} + 0.48\mathbf{b}$ from the bottom layer (green). **(E)** Spin-polarized tunneling on an R-type stacked bilayer CrBr₃ with a Cr tip at $V_b = 1.5$ V. The insets show four magnetization configurations, including the Cr tip and bilayer CrBr₃, corresponding to different magnetic field-dependent dI/dV plateaus. The out-of-plane magnetic field was swept upward (black data) and downward (red data). **(F)** Spin-dependent tunneling on the bilayer CrBr₃ in (C) with a nonmagnetic W tip at $V_b = 1.5$ V. Abrupt decrease of the dI/dV signal was observed at magnetic fields of $\sim \pm 0.5$ T, suggesting an interlayer antiferromagnetic coupling within ± 0.5 T.



between the Br p-orbitals and Cr d-orbitals (32, 33). Because the bond angles and the bond distances of the Cr–Br–Br–Cr exchange path are strongly dependent on the stacking order, the interlayer magnetism is expected to also depend on the specific stacking structure, including the interlayer distance and atomic site positions. In fact, recent theoretical studies have investigated the interlayer magnetism of the rhombohedral and monoclinic structures in bilayer CrI₃ (34–37).

Although the exact growth mechanism remains to be investigated, our observations illustrate the polytypism in vdW materials and its importance to 2D magnetism. Our work also calls for a close examination for the stacking structures in mechanically exfoliated CrX₃ samples, as the exfoliated bilayer CrI₃ and CrBr₃ exhibit distinct interlayer magnetic coupling. We envision that this working principle could be used to manipulate 2D magnetism, for example, by engineering spatially dependent spin textures uniquely enabled by twisted bilayers and heterostructures (38).

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SUPPLEMENTARY MATERIALS

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Materials and Methods
Figs. S1 to S17
Tables S1 and S2
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Direct observation of van der Waals stacking–dependent interlayer magnetism

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Stacking control

Recent searches for two-dimensional magnets have turned up a number of related van der Waals materials, such as chromium triiodide (CrI_3) and chromium tribromide (CrBr_3). Although their properties are expected to be similar, in bilayer form, the former is antiferromagnetic, whereas the latter appears to be a ferromagnet. Chen *et al.* used spin-polarized scanning tunneling microscopy to determine that the nature of the magnetic state in bilayer CrBr_3 depends on the type of stacking of its monolayers. An antiferromagnetic state formed when the two monolayers were oriented in the same direction, whereas the opposite orientation led to ferromagnetism.

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