Evidence of non-collinear spin texture in magnetic moiré superlattices

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Moiré magnetism is emerging as a platform to design and control exotic magnetic phases in twisted magnetic two-dimensional crystals. Non-collinear spin texture emerging from twisted two-dimensional magnets with collinear spins is one of the most profound consequences of moiré magnetism and forms the basis for realizing non-trivial magnetic orders and excitations. However, no direct experimental observations of non-collinear spins in moiré magnets have been made, despite recent theoretical and experimental efforts. Here, we report evidence of non-collinear spin texture in two-dimensional twisted double bilayer CrI₃. We distinguish the non-collinear spins with a gradual spin flop process from the collinear spins with sudden spin flip transitions and identify a net magnetization emerging from the collinear spins. We also demonstrate that both non-collinear spins and net magnetization are present at twist angles from 0.5° to 5° but are most prominent for 1.1°. We resolve a critical temperature of 25 K for the onset of the net magnetization and the softening of the non-collinear spins in the 1.1° samples. This is substantially lower than the Néel temperature of 45 K for natural few layers. Our results provide a platform to explore non-trivial magnetism with non-collinear spins.

Non-collinear magnetism covers a wealth of exotic magnetic phases in three-dimensional (3D) bulk systems, including chiral antiferromagnetism, spiral magnetism, helical magnetism, skyrmions, and magnetic multipoles. Many of these hold promise for spintronic applications. It has been demonstrated that magnetic competition, where competing forces for opposite spin alignments cannot be satisfied simultaneously, is one route to introduce non-collinear spin textures. Down to the two-dimensional (2D) limit, all 2D magnets discovered to date, both ferromagnetic and antiferromagnetic, primarily host collinear spins. Guided by moiré electronics where new electronic phases emerge owing to the presence of moiré superlattices, the magnetic analogue, moiré magnets made by twisting two magnetic atomic layers, is expected to introduce emergent magnetic phases distinct from those in natural 2D magnets. An immediate question in this context is whether 2D non-collinear spin texture can be introduced in moiré superlattices made of twisted 2D magnets with collinear spins.

Very recently, this scenario was theoretically predicted in moiré magnets where stacking dependent interlayer exchange couplings with opposite signs coexist and compete within individual moiré supercells. However, experimental studies of moiré-induced 2D non-collinear spin texture remain absent despite a few very recent efforts to probe coexisting magnetic orders in twisted chromium triiodide (CrI₃) layers, and its long-range magnetic order.

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The moiré superlattice formed at the interface between two 2L CrI₃, namely between the second and third layers. Regions of AA- (green), R- (blue) and M-type (red) stacking geometries are marked in one moiré supercell (black parallelogram). Inset: Schematic of twisting two 2L CrI₃ by an angle α.

The periodically modulating interlayer exchange coupling $J$ at the interface between two 2L CrI₃ survives down to the atomic layer limit. Extensive experiments and calculations have shown that monoclinic interlayer stacking leads to antiferromagnetic (AFM) interlayer exchange coupling whereas rhombohedral interlayer atomic registry results in FM interlayer exchange coupling. In natural few-layer CrI₃, the structure favours the monoclinic stacking, and the magnetism takes the layered AFM order where spins align along the same out-of-plane direction within the layers but in opposite directions between adjacent layers. Importantly, even-layer CrI₃ has a compensated zero magnetization whereas odd-layer CrI₃ ends up with a finite non-zero magnetization. In twisted double odd-layer CrI₃, regions with zero and non-zero magnetization coexist at small twist angles, while in twisted double even-layer CrI₃, an unexpected non-zero magnetization emerges at around a critical twist angle of 1.1° (ref. 19). Yet, the question of spin reorientation, upon moiré modulations, to depart from the collinear arrangement in natural layers remains unexplored in these three pioneering experimental works.

Here, we choose the twisted double bilayer (tDB) CrI₃ system to investigate the non-collinear spins from the moiré engineering. tDB CrI₃ is an artificial four-layer homostructure made by two bilayer (2L) CrI₃ stacked vertically with a controlled twist angle α. Importantly, the interface between the two bilayers, a moiré supercell whose area is three times that of the unit cell seen in this image formed from the second-order moiré superlattice peaks.
the AA sites, approximately 0.04 meV μ_B^2 AFM interlayer coupling at the M sites, and approximately −0.6 meV μ_B^2 FM interlayer coupling at R sites (Fig. 1d). Here μ_B is the Bohr magneton. This interlayer moiré exchange coupling that periodically varies from AFM to FM type prefers to have spatially modulating spin arrangement, whereas the intralayer FM exchange coupling favours uniform out-of-plane spin alignment within a CrI_3 layer. It is the competition between these two exchange couplings that determines the magnetic ground state in tDB CrI_3.

Using a continuous spin model (Methods), we compute the total magnetic energy of the system that includes intralayer FM coupling, uniform interlayer AFM coupling within each bilayer and interlayer moiré exchange coupling between the two bilayers, and identify the ground state spin configuration by minimizing this magnetic energy. We find that the magnetic ground state evolves as the twist angle increases. The computed results at three representative twist angles (0.1°, 1° and 10°) are shown in Fig. 1c–e. At very small twist angles such as 0.1° (Fig. 1c), spatially modulating-out-of-plane magnetization m_z develops in both the first and second layers and is antiparallel between these two layers, whereas homogeneous m_z forms in the third and fourth layers with spin down (↑↑↓↑) in layer 3 and spin up (↑↑↓↑) in layer 4. We further correlate the layered magnetism with the lattice stacking in Fig. 1f. The islands centring at AA stacking and surrounded by M stacking take the ↑↑↓↑ layered magnetic order, and the large remaining region centring at R stacking has the ↑↑↓↑ layered magnetism. The boundaries between the two regions in the top bilayer exhibit the non-collinear spin texture (↑↑↓↑ in layer 1 and ↓↑↑↓ in layer 2). Summing m_z in all four layers for this magnetic ground state of 0.1° tDB CrI_3 leads to zero total magnetization (M_z^F = 0). At intermediate twist angles such as 1°, the computed magnetic ground state (Fig. 1d) shows modulated spins only in the second layer and a net total magnetization (M_z^F ≠ 0), which features islands with non-zero magnetization (↓↑↑↓) centring at AA sites, boundaries with non-collinear spins in layer 2 coinciding with the rings formed by M sites and a background with zero magnetization (↑↑↓↑) centring at R sites. At large twist angles such as 10°, the calculated result shows homogeneous spins within all four layers (Fig. 1e) and takes the ↑↑↓↑ layered magnetic order with total zero magnetization (M_z^F = 0) in Fig. 1f. While the details of the spin configurations may vary depending on the microscopic parameters in the model, the emergence of non-zero magnetization only at intermediate twist angles and the formation of non-collinear spins for small to intermediate twist angles are robust features in our simulated moiré magnetism in tDB CrI_3 (Supplementary Section I).

To connect the theoretical calculations to experimental measurements, we first establish that our tDB CrI_3 hosts a high-quality moiré superlattice, using selected area electron diffraction (SAED) and dark-field transmission electron microscopy (DF-TEM) (Methods). Figure 2a shows the zoom-in SAED pattern of tDB CrI_3 with a twist angle of α = 1.4 ± 0.1° (Supplementary Section 2). In addition to three pairs of Bragg peaks from the CrI_3 honeycomb lattice, we can also observe two additional peaks locating between each pair, which correspond to the second-order moiré superlattice peaks. This clear observation of superlattice peaks in the diffraction pattern is strong and direct evidence of the formation of moiré superlattices. Figure 2b shows three real-space DF-TEM images taken from within the large SAED region using three pairs of Bragg and superlattice peaks, labelled by the corresponding Bragg peak indices. Consistent with the presence of superlattice peaks in the electron diffraction pattern, the DF-TEM image for every pair clearly shows the parallellogram feature, in contrast to parallel lines for the case without superlattice formation. Figure 2c is a composite image made by summing up the three DF-TEM images in Fig. 2b, which clearly shows the superlattice structures uniformly extending over an area of approximately 250 × 125 nm^2 (Supplementary Section 2).

Having shown the high-quality moiré superlattice in tDB CrI_3, we use magnetic circular dichroism (MCD) to directly verify the non-zero magnetization and its magnetic field dependence to reveal the non-collinear spin texture (Methods). Figure 3a shows the MCD data as a function of an out-of-plane magnetic field (B_z) swept from +2 T to −2 T and then back to +2 T for 1.1° tDB, 2L and 4L CrI_3. The spin flip transitions are marked by the black arrows in all three panels. The error bars correspond to one s.d. of 15,000 MCD intensities measured over a time interval of 10 s at every magnetic field. The fitting model is detailed in Methods. a, c. Contributions from collinear (b) and non-collinear (e) spins to the MCD signal extracted from the model fits for 1.1° tDB, 2L and 4L CrI_3. d. Schematics of the M_z distribution evolving upon increasing B_z from 0 T across B_z^top to 2 T.

![Figure 3](https://doi.org/10.1038/s41567-023-02061-z)
We can then model and fit the MCD data of 1.1° tDB CrI₃ with two major contributions: one from the collinear spins that feature the steep spin flip transitions and the other from the non-collinear spins that account for the gradual spin flop process (Methods). The fitted results are shown in Fig. 3a (solid lines), b and c for the sum and each of the contributions in 1.1° tDB CrI₃ (top panels), together with those for 2L and 4L CrI₃ (middle and bottom panels). This fit is independent from but consistent with the simulations in Fig. 1d in terms of the following two key aspects. First, the net magnetization falls into the collinear spin contribution in this fit (Fig. 3b, top panel), matching the magnetized islands with collinear ↑↑↓↑ spin arrangements in the simulations (Fig. 1d). Second, for the non-collinear spin contribution, the two fitted MCD traces (Fig. 3c, top panel) with decreasing and increasing Bᵢ overlap with each other within our fit uncertainties, echoing the computed net zero out-of-plane magnetization at the boundaries between the ↑↑↓↑ islands and the ↑↑↓↑ background in Fig. 1d. As a controlled comparison, the fits for 2L and 4L CrI₃ are dominated by the collinear spin contribution with compensated magnetizations below Bᶜ₁₋₂ (Fig. 3b, middle and bottom panels) and show tiny non-collinear spin contributions (Fig. 3c, middle and bottom panels).

On the basis of the computations and the fit, we further provide a physics picture of the spin texture evolution upon Bᵢ to vividly interpret the MCD data. In Fig. 3d, we start with the computed ground state for 1.1° tDB CrI₃, at 0 T (step I) and depict the evolution of spin states under increasing Bᵢ till 2 T. Initially, with Bᵢ < Bᶜ₁₋₂, the boundaries with non-collinear spins in the second layer gradually move outwards to expand the ↑↑↓↑ islands because the upwards Bᵢ direction aligns along the spin-up orientation for the island (step II). This process accounts for the slow increase of MCD below Bᶜ₁₋₂. Right across Bᶜ₁₋₂, the collinear spins in the background region flip from ↑↑↓↑ to ↑↑↓↑, which concurrently shifts the modulating spin structure from the second to the third layer (step III). This change, responsible for the steep jump in MCD at Bᶜ₁₋₂, happens because it costs the energy penalty of interlayer exchange coupling with one adjacent layer that Bᶜ₁₋₂ can afford, similar to Bᶜ₁₋₄L does in 2L/4L CrI₃. With further increasing Bᵢ, the boundaries with non-collinear spins in the third layer move inwards to shrink the ↑↑↓↑ islands where the upwards Bᵢ points in the opposite direction to the downwards spins in the islands in this third layer (step IV). This evolution explains the further gradual increase of MCD above Bᶜ₁₋₂. Eventually, the islands in the third layer disappear, and spins in all four layers of tDB CrI₃ are polarized to the same direction as Bᵢ (step V). This final state corresponds to the saturation of MCD, which should happen at Bᵢ close to Bᶜ₂₋₃.

Having understood the MCD data for 1.1° tDB CrI₃, we proceed to explore the twist angle dependence of the moiré magnetism. Figure 4a displays the raw MCD data (top panel) normalized to values at 2 T, together with the fits for the sum (top panel) and each (middle and bottom panels) of the contributions from collinear and non-collinear spins, for 4L, α = 0.5°, 1.1°, 2°, 5° and 10° tDB, and 2L CrI₃ (Supplementary Section 3). A general trend can be seen that the MCD data resemble 4L CrI₃, at very small twist angles (for example, 0.5°) in having two spin flip transitions, then distinguish from either 4L or 2L CrI₃ at intermediate twist angles (for example, 1.1°, 2° and 5°) by showing a prominent hysteresis loop as well as an appreciable non-collinear spin contribution, and eventually converge to 2L at large twist angles (for example, 10°).

To better quantify this trend, we plot the twist angle dependence of five important parameters extracted from the fits, namely the spin flip transition field Bᶜᵢ that corresponds to the interlayer coupling within each bilayer (Fig. 4b), the width of the spin flip transitions ΔBᶜᵢ that describes the spatial inhomogeneity from moiré magnetism (Fig. 4c), the slope at 0 T, ΔMCD/ΔBᶜᵢ (0 T), that depicts the susceptibility of the non-collinear spins (Fig. 4d), the ratio of the non-collinear spin contribution to the total MCD Wᵣ₊₋non-collinear, which scales with the weight of the non-collinear spins (Fig. 4e), and the magnitude of net

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**Fig. 4 | Twist angle dependence of MCD data of tDB CrI₃.** a. Normalized MCD data and fits taken at 10 K under Bᵢ, sweeping from +2 T to –2 T and then back to +2 T, for 4L, α = 0.5°, 1.1°, 2°, 5° and 10° tDB, and 2L CrI₃ (top panel), showing hysteresis loops (red shades) and spin flip transitions (black arrows). The error bars correspond to one s.d. of 15,000 MCD intensities measured over a time interval of 10 s at every magnetic field. Contributions from the collinear (middle panel) and the non-collinear (bottom panel) spins are extracted from the model fitting. b–f. Twist angle dependence of the spin flip transition field Bᶜᵢ (b), spin flip transition field width ΔBᶜᵢ (c), the slope at 0 T ΔMCD/ΔBᶜᵢ (d), the weight of the non-collinear spin contribution W₊₋non-collinear (e) and the net magnetization M₀ₓ (f), extracted from the model fitting. The error bars correspond to two s.d. in fitting the MCD data.
For the optimal twist angle of 1.1°, we further carry out the temperature dependence measurement of MCD from 60 to 10 K, across the Néel temperature $T_N = 45$ K for natural few-layer CrI$_3$. Figure 5a shows the raw data and fits of MCD datasets at selected temperatures (Supplementary Section 4). At 60 K, the MCD data show a linear dependence on $B_z$ and confirm the paramagnetic behaviour at this temperature. Below 40 K, the spin flip transitions at $B_{c1}$ become progressively visible, and at 25 K, the hysteresis loop with non-zero MCD at 0 T appears. We first discuss the fitting parameters for the collinear spin contribution (Fig. 5b–d). The fitted values for MCD at the 0 T spectra kick up at around 25 K (Fig. 5b), which is significantly reduced from the onset of around 45 K for the layered AFM in 2L/4L CrI$_3$ (ref. 31). The temperature-dependent $B_{c1}$ follows the same trend as those for 2L/4L CrI$_3$ and shows an onset at around 45 K (Fig. 5c). The temperature dependence of $\Delta B_{c1}$ clearly departs from those for 2L/4L CrI$_3$ at temperatures below about 25 K when the net magnetization builds up. We then move on to the fitting parameters for the non-collinear spin contribution (Fig. 5e–f). The temperature dependence of the slope peaks at around 45 K for both 1.1° tDB and natural 2L/4L CrI$_3$, but it has a much larger magnitude in tDB than 2L/4L CrI$_3$ below 45 K (Fig. 5f). The temperature dependence of $W_{\text{non-collinear}}$ below 45 K shows a decreasing trend for both 1.1° tDB and 2L/4L CrI$_3$ as the thermal excitation-induced spin canting gets suppressed at lower temperatures. But the $W_{\text{non-collinear}}$ of 1.1° tDB CrI$_3$ is much greater than those of 2L and 4L CrI$_3$, and shows a steeper decrease below about 25 K when the net magnetization shows up.

Through temperature-dependent MCD fitting parameters, two characteristic temperatures, $T_{c1} = 45$ K and $T_{c2} = 25$ K, are revealed in 1L° tDB CrI$_3$. To approach the origin of the suppressed net magnetization onset temperature of 25 K, we first examined the temperature-dependent MCD for tDB CrI$_3$ with a larger twist angle (for example, 2°). We confirmed the 45 K magnetization onset temperature in 2° tDB CrI$_3$ (Supplementary Section 5), which convincingly rules out the weakened inter-bilayer exchange coupling as the cause for the suppression in the magnetization onset temperature in 1.1° tDB CrI$_3$. We further performed temperature-dependent polarized Raman spectroscopy measurements in 1.1° tDB CrI$_3$, where the layered-magnetism-assisted Raman scattering probes the layered magnetic orders even with zero magnetization and is complementary to the net magnetization-sensitive MCD. Figure 5g shows Raman spectra in the linearly parallel and crossed channels at selected temperatures. In total, four modes ($U_{\downarrow\downarrow\downarrow\downarrow}$) are observed owing to the Davydov splitting, with three in the crossed channel ($U_{\downarrow\downarrow\downarrow\downarrow}$) from the layered-magnetism-assisted phonon scattering process and the other one in the parallel channel ($U_{\downarrow\downarrow\downarrow\downarrow}$) from the regular phonon scattering $U_4$. The fitted frequencies are very similar to those for 2L/4L CrI$_3$ and shows an onset at around 45 K (Fig. 5c). The temperature dependence of $W_{\text{non-collinear}}$ below 45 K shows a decreasing trend for both 1.1° tDB and 2L/4L CrI$_3$, as the thermal excitation-induced spin canting gets suppressed at lower temperatures. But the $W_{\text{non-collinear}}$ of 1.1° tDB CrI$_3$ is much greater than those of 2L and 4L CrI$_3$, and shows a steeper decrease below about 25 K when the net magnetization shows up.

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In summary, we have used electron microscopy to demonstrate the high-quality moiré superlattice in tDB CrI$_3$, and then magneto-optical measurements to show signatures of an emergent net magnetization and non-collinear spin texture. Demonstrate impacts of moiré-induced magnetic competitions and identify the unique twist angle of around 1.1° for this system. We note that our current study lacks local magnetic imaging to directly visualize the spin texture within moiré supercells. Future magnetic nanoscopy studies will provide details on this emergent magnetism, for example, resolving the real-space distribution between collinear and non-collinear spins within moiré supercells and distinguishing different types of non-collinear spins (for example, Ising, Néel, Bloch or mixed). Their integration with applications of an external magnetic field, if possible, will further enable research on the spin pattern evolution under $B_z$. Moreover, dynamic probes, such as time-resolved magneto-optics, or their near-field versions, or inelastic x-ray scattering, are called for to probe moiré magnons in moiré magnets.

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Methods
Fabrication of 2D natural CrI₃ and tDB CrI₃
High-quality single crystals of CrI₃ were grown by the chemical vapour transport method, as reported in detail in ref. 37. The samples for SAED/DF-TEM and optical measurements were fabricated separately as described below, and different samples were used for SAED/DF-TEM and optical studies. Atomically thin CrI₃ flakes were mechanically exfoliated from bulk crystals onto 285 nm SiO₂/Si substrates inside a nitrogen-gas filled glovebox with O₂ level below 0.1 ppm and H₂O level below 0.5 ppm. The thickness of CrI₃ flakes was initially identified by the optical contrast under an optical microscope inside the glovebox, and then further verified by Raman spectroscopy at 10 K. Using the ‘tear-and-stack’ technique, tDB CrI₃ samples were fabricated by initially picking up a part of 2L CrI₃ and then stacking it on top of the remaining part on the substrate at a targeted twist angle of 6. Both tDB and 4L/2L CrI₃ were encapsulated on both sides with hexagonal boron nitride (hBN) flakes with thickness of ~15 nm. For the optical studies, the final stack of hBN/CrI₃/hBN was released onto the SiO₂/Si substrates and then rinsed in chloroform solvent to clean the polymer residue, before subsequent magneto-optical measurements. For the SAED/DF-TEM studies, thinner hBN (<5 nm thick) was used to encapsulate the tDB CrI₃, thus a 10-nm-thick SiN membrane.

SAED and DF-TEM
SAED and DF-TEM results of tDB CrI₃ were acquired using a Thermo Fisher Scientific Talos, equipped with a Gatan OneView camera. The twist angle was quantified by fitting pairs of (0303), (3030) and (3030) Bragg peaks with two-dimensional Gaussians, with an electron beam field of view of 850 nm. DF-TEM results in Fig. 2b were achieved by acquiring two-beam conditions on (0303), (3030), (3030) Bragg and moiré superlattice satellite reflections. Figure 2c was generated by averaging the three DF-TEM micrographs in Fig. 2b to remove anisotropy. The statistical distribution of twist angles in the real-space DF-TEM images was further analysed by manually segmenting superlattice unit cells (Supplementary Section 2).

MCD
Similar MCD measurements were reported for atomically thin CrI₃ flakes in earlier studies. For the measurements in the current study, normal incident light with a wavelength of 632.8 nm was focused onto the hBN/CrI₃/hBN sample with a full width at half maximum (FWHM) of 2–3 μm using a ×40 transmissive objective, which is substantially smaller than the sample sizes of both tDB and 4L/2L CrI₃. The polarization of the incident light was modulated between left- and right-handed circular polarization (L/RCP) using a photo-elastic modulator (PEM; Hinds Instruments PEM-200) at a modulating frequency of f = 50 kHz. The reflected signal was collected by a biased photodiode detector. Two separate measurements were carried out to determine the difference between and the average of the LCP and RCP light in the reflection. The difference was measured by demodulating the reflected signal against the PEM frequency using a lock-in amplifier, and the average was measured by inserting a mechanical chopper into the incident beam and then demodulating the reflected light at the mechanical chopper frequency with the lock-in amplifier. The MCD value was defined by the ratio of the difference to the average signal.

Polarized Raman spectroscopy
Raman scattering measurements were carried out using a 632.8 nm excitation laser with a FWHM value of 0.85 cm⁻¹, on the resonance with charge transfer and Cr⁴⁺ A₂g to A₁g transitions of CrI₃, to increase the Raman sensitivity. The laser beam on the sample site was focused down to ~2–3 μm FWHM in diameter using a ×40 transmissive objective, and the laser power was kept at about 80 μW to minimize the local heating effect during measurements. A backscattering geometry was used, where the scattered light was dispersed by a Horiba LabHR Evolution Raman microscope (with a grating of 1,800 grooves per millimetre) from HoribaScientific and detected by a thermoelectrically cooled charge-coupled device camera from HoribaScientific, with a spectrometer resolution of 0.3 cm⁻¹. A commercial variable-temperature (~10 to 325 K), closed-cycle microscopy cryostat (Cryo Industries of America) was interfaced with the Raman microscope.

Calculations of magnetic ground state of tDB CrI₃
We include the intralayer exchange coupling, the spin anisotropy energy, the interlayer AFM exchange coupling within each bilayer and the modulated interlayer exchange coupling between the two bilayers in the magnetic Hamiltonian

$$H = \sum_{ij} \frac{\delta}{222} (S_i^x S_j^y + S_i^y S_j^x) + \frac{\Delta}{2} (S_i^x S_j^x + S_i^y S_j^y).$$

For the twist angles that we consider (for example, intermediate twist angles such as ~1°), the moiré wavelength is about ~40 nm, and the magnetization in each layer varies slowly in real space. Therefore, we can take the continuum limit of the following spin Hamiltonian:

$$H = \frac{1}{\delta} \sum_{i,j} \left( S_i^x S_j^y + S_i^y S_j^x \right).$$

With magnetization varies slowly in real space, we can take the continuum limit, replacing \( \mathbf{n} \) with a smooth function \( \mathbf{n}(r) \), which is also a 3D unit vector

$$H = \frac{1}{\delta} \sum_{i,j} \left( n_i \cdot n_j \right).$$

In this continuum limit, we can perform a Taylor expansion, using \( \delta \mathbf{r} - \mathbf{r} \) as a small parameter

$$n_m(r) = n_m(r) + \frac{\mathbf{r}}{2} n_m(r) \left( \delta \mathbf{r} \cdot \nabla n_m(r) \right) + \frac{1}{2} n_m(r) \left( \delta \mathbf{r} \cdot \nabla \right)^2 n_m(r) + O(\delta^2).$$

with \( m = x, y \text{ or } z \), and \( \delta \mathbf{r} \) (or \( \delta \mathbf{r} \) is the displacement vector (distance) from position \( \mathbf{r} \) to position \( \mathbf{r} \)). Because the anisotropy constant \( g = 1.0445 \) is close to 1 in CrI₃, here we take the following approximation. In the leading/order term of this Taylor series, \( n_m(r) \), \( m = x, y \text{ or } z \), we keep \( \mathbf{r} = 1.0445 \), while for higher-order terms, we set \( m = 1 \). This approximation simplifies the Hamiltonian without changing any qualitative features (that is, we use n² terms to describe the easy-axis spin anisotropy of CrI₃, while derivative terms of \( \mathbf{n}(\mathbf{r}) \) preserve the SU(2) spin rotational symmetry). Within this approximation, the above Hamiltonian becomes

$$H = \frac{1}{\delta} \sum_{\text{unit cells}} \left[ \frac{1}{2} m^2 \sum_{\mathbf{n}(\mathbf{r})} \left( \delta \mathbf{r} \cdot \nabla |\mathbf{n}|^2 \right) - \frac{3}{4} \delta^2 |\mathbf{n}|^2 \right].$$

Here, first-order derivative terms cancel out, while the second-order derivative terms can be written as \( \alpha |\mathbf{n}|^2 \sum_{\mathbf{n}(\mathbf{r})} \left( \delta \mathbf{r} \cdot \nabla \right)^2 |\mathbf{n}|^2 \). Higher-order terms beyond \( O(\delta^2) \) are ignored. For the honeycomb lattice of Cr⁺ ions, \( \delta = a/\sqrt{3} \), where \( a \) is the lattice constant, and the sum in the Hamiltonian above (over unit cells) can be replaced

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by the real-space integral \( \frac{1}{l} \int_{d} \int_{dy} dx \), where \( d = \frac{\sqrt{3}}{2} a^{2} \) is the area of one unit cell. Thus, for each layer, the Hamiltonian becomes
\[
H(0) = \int_{dx} \int_{dy} \left( -\frac{a^{2}}{4} \left[ V n_{x}^{2} \right] + 3 \left( n_{x}^{2} + n_{y}^{2} + y n_{y}^{2} \right) \right) \]  
(7)
where \( l = 1, 2, 3, 4 \) is the index of the four layers.

For interlayer couplings, \( H_{M}^{(2,3)} \) and \( H_{M}^{(3,4)} \) describe the interlayer exchange interactions within each bilayer between layers (1, 2) and layers (3, 4)
\[
H_{M}^{(2,3)} = \frac{2J_{m}S^{2}}{a^{2}} \int _{dx} \int _{dy} n_{x}^{(i)} \cdot n_{x}^{(j)}, \]  
(8)
\[
H_{M}^{(3,4)} = \frac{2J_{m}S^{2}}{a^{2}} \int _{dx} \int _{dy} n_{y}^{(i)} \cdot n_{y}^{(j)}. \]  
(9)
where \( J_{m} \approx 0.04 \text{ meV} q_{u}^{2} \) (ref. 27) is the effective AFM-type interlayer exchange coupling constant between pristine monolayerically stacked two layers. \( H_{M}^{(2,3)} \) denotes the periodically modulated magnetic coupling at the twisted interface between layers (2, 3)
\[
H_{M}^{(2,3)}_{\text{moire}} = \int _{dx} \int _{dy} \frac{2J_{m}S^{2}}{a^{2}} n_{x}^{(2)} \cdot n_{x}^{(3)}. \]  
(10)

The moiré modulating interlayer exchange interaction \( J_{m} \) takes the spatial distribution shown in Fig. 1b, which is obtained by adopting the interlayer exchange coupling at a few specific stacking sites\(^{27}\) and then using the harmonic expansion to second order to generate the two-dimensional spatial dependence, in analogy to the Bistritzer-MacDonald model\(^{28}\).

To obtain the lowest energy spin configuration in this continuous model, we discretize a moiré unit cell using a triangular grid of \( L \times L = 60 \times 60 \times 4 \), and define the following effective Hamiltonian on this grid:
\[
H(0) = \frac{J_{m}S^{2}}{3} \sum _{dist} n_{i}^{(i) \cdot n_{j}^{(j)}} + \frac{3J_{m}S^{2}}{a^{2}} \sum _{dist} \left[ n_{ix}^{2} + n_{iy}^{2} + y n_{iy}^{2} \right]. \]  
(11)
\[
H_{M}^{(2,3)} = \frac{2J_{m}S^{2}}{a^{2}} \sum _{dist} n_{x}^{(i)} \cdot n_{x}^{(j)} \]  
(12)
\[
H_{M}^{(3,4)} = \frac{2J_{m}S^{2}}{a^{2}} \sum _{dist} n_{y}^{(i)} \cdot n_{y}^{(j)} \]  
(13)
\[
H_{M}^{(2,3)}_{\text{moire}} = \sum _{dist} \frac{2J_{m}S^{2}}{a^{2}} n_{x}^{(2)} \cdot n_{x}^{(3)} \]  
(14)
where \( a \) is the twist angle and \( L = 60 \) is the linear size of the grid that we use to discretize a moiré unit cell. The dimensionless quantity \( (a/\Delta a) \) where \( a \) and \( \Delta a \) are the areas of a CrI\(_3\) unit cell and a unit cell of this triangular grid, respectively. It is straightforward to verify that this triangular grid model recovers the same continuous model shown above, and thus when \( L \) is sufficiently large, we can use this coarse-grained triangular grid to obtain the spin configuration of a moiré unit cell.

We then minimize this Hamiltonian via the quasi-Newton method. We allowed all spins in the four layers to vary in search of the ground state configuration. In addition, to avoid local energy minima, we examined a variety of initial conditions of spin arrangement, including (1) fully random, (2) largely homogeneous in all four layers and (3) largely homogeneous in some of the layers (random in others).

We then identify the final state with the lowest energy by comparing results obtained from different initial conditions. We note that the lowest energy state is highly robust and always exhibits the same spin configuration (up to the degenerate states related by the spontaneous broken symmetries), and that these lowest energy states can always be accessed from the fully random initial conditions.

The resulting ground state depends on the competition between intralayer and interlayer energy, which depend on the twist angle differently. To see this in our model, the intralayer energy cost occurs at the neighbouring in-plane region between spins of opposite directions, captured by the first term in equation (7), \( -\frac{J_{m}S^{2}}{a^{2}} \int _{dx} \int _{dy} \left[ V n_{x}^{2} \right] \), which scales to the area of the ‘transitioning’ region between two regions with opposite spin alignments, that is, \( 2n_{\alpha_{i}+d} \) with \( \alpha_{i} \) being the moiré wavelength and \( d \) being the width of the ‘transitioning’ region (the non-collinear spin region). According to equation (7), the comparison between the first term and the second term provides the width \( d \), which is twist angle independent. The moiré wavelength \( \alpha_{i} \), on the other hand, is proportional to \( \alpha^{-1} \) with \( \alpha \) being the twist angle. The interlayer energy cost occurs in the interface between layers in equations (8)–(10) whose energy all scales with the area of moiré cells, that is, \( n\alpha_{i}d_{c} \), proportional to \( \alpha^{-2} \). The different scaling leads to changes of the magnetic ground state as the twist angle changes.

**Fitting model for MCD data**

On the basis of observation of the MCD data, we find that, for a typical 11+1 TDB CrI\(_3\) measured below \( T_{B} \), MCD data consist of two components: (1) the sharp spin flip component with critical fields at \( \pm 2B \) from the collinear spins and (2) the slowly varying component between \( -2B \) to \( +2B \) from the non-collinear spins.

For the collinear spin contribution, the out-of-plane spins take two possible directions, spin up and down, that is, \( \mu = \pm 2 \cdot \frac{3}{k_{B}} \). Boltzmann statistics dictates that the mean magnetization under an external field \( B \) follows the tanh function
\[
\mu > = \frac{3\mu_{B}}{k_{B}T} \tanh \frac{3B}{k_{B}T}, \]  
(15)

On the basis of system equivalency under time-reversal operation for the collinear spins as we observed from the natural 2L/4L CrI\(_3\), the spin flip transitions adopt two constraints: (1) ‘flip-up’ and ‘flip-down’ transitions should occur at the same magnetic field strength despite opposite magnetic field directions, and (2) the increasing-field curve and the decreasing-field curve should map to each other when inverting about the origin at \( x = 0 \) and \( y = 0 \) (that is, \( B_{c} = 0 \) and MCD = 0 in the MCD spectra).

For the non-collinear spin contribution, phenomenologically we also take a tanh function with a wide width to describe the slowly varying feature. Yet, no constraints were added, unlike the collinear spin contributions.

As a result, the following model is used to fit the MCD curve:
\[
\begin{align*}
Y_{+} = A_{\text{hgp}}\tanh \frac{3x}{c} + A_{\text{hgp}}\tanh \frac{3-3B}{c} + \alpha_{t}\tanh \frac{x-3B}{c} \\
Y_{-} = A_{\text{hgp}}\tanh \frac{3x}{c} + A_{\text{hgp}}\tanh \frac{3B-c}{c} + \alpha_{t}\tanh \frac{x-3B}{c}
\end{align*}
\]  
(16)
where \( x \) is the external magnetic field \( B_{c} \) and \( y_{+}(-) \) is the MCD signal in an increasing-field (decreasing-field) measurement. For each equation, the first two terms explain the spin-flip transitions of Ising spins and the third term represents the response of the non-collinear spins. \( A_{\text{hgp}} \) describes the magnitude of the spin-flip transition, and \( B \) and \( c \) are the transition field \( B_{c} \) and width \( \Delta B \), respectively, for the collinear part. Those parameters are kept common in fitting \( y_{+} \) and \( y_{-} \) datasets to fulfill the two aforementioned constraints for the collinear
spins. For the non-collinear contribution, $a_\pm$, $b_\pm$ and $c_\pm$ describe the magnitude, centre and width of the slowly varying background in either an increasing (+) or decreasing (−) measurement and are independent between the increasing and decreasing field traces. For this emergent non-collinear contribution, we choose to release the time-reversal operation-related constraints between data of increasing and decreasing $B$ field and check from the fitted results whether the corresponding fitting parameters are indeed the same between up and down sweeps.

**Data availability**

Raw data for SAED and DF-TEM data are images shown in the main text and Supplementary Information. Raw data for MCD and Raman results are provided as Source data provided with this paper.

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**Author contributions**

L.Z., H.X. and X.L. conceived the idea and initiated this project. H.X. and Z.S. fabricated the 4L, 2L and tDB CrI$_3$ samples. H.X., X.L., Z.Y., G.Y. and H.G. built the MCD setup and carried out the MCD measurements under the supervision of L.Z. and R. He. S.H.S. and R. Hovden performed the electron diffraction and TEM measurements. S.Y., Y.F., S.T. and H.L. grew the CrI$_3$ bulk single crystals. X.L. and K.S. performed the theoretical computation and analysis. X.L. and L.Z. analysed the data, and X.L., R. He and L.Z. wrote the manuscript. All authors participated in the discussion of the results.

**Competing interests**

The authors declare no competing interests.

**Additional information**

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