

Layer-dependent ferromagnetism in a van der Waals crystal down to the monolayer limit

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Since the discovery of graphene¹, the family of two-dimensional materials has grown, displaying a broad range of electronic properties. Recent additions include semiconductors with spin-valley coupling², Ising superconductors^{3–5} that can be tuned into a quantum metal⁶, possible Mott insulators with tunable charge-density waves⁷, and topological semimetals with edge transport^{8,9}. However, no two-dimensional crystal with intrinsic magnetism has yet been discovered^{10–14}; such a crystal would be useful in many technologies from sensing to data storage¹⁵. Theoretically, magnetic order is prohibited in the two-dimensional isotropic Heisenberg model at finite temperatures by the Mermin–Wagner theorem¹⁶. Magnetic anisotropy removes this restriction, however, and enables, for instance, the occurrence of two-dimensional Ising ferromagnetism. Here we use magneto-optical Kerr effect microscopy to demonstrate that monolayer chromium triiodide (CrI₃) is an Ising ferromagnet with out-of-plane spin orientation. Its Curie temperature of 45 kelvin is only slightly lower than that of the bulk crystal, 61 kelvin, which is consistent with a weak interlayer coupling. Moreover, our studies suggest a layer-dependent magnetic phase, highlighting thickness-dependent physical properties typical of van der Waals crystals^{17–19}. Remarkably, bilayer CrI₃ displays suppressed magnetization with a metamagnetic effect²⁰, whereas in trilayer CrI₃ the interlayer ferromagnetism observed in the bulk crystal is restored. This work creates opportunities for studying magnetism by harnessing the unusual features of atomically thin materials, such as electrical control for realizing magnetoelectronics¹², and van der Waals engineering to produce interface phenomena¹⁵.

Magnetic anisotropy is an important requirement for realizing two-dimensional (2D) magnetism. In ultrathin metallic films, whether an easy axis can originate from symmetry reduction at the interface or surface is dependent on substrate properties and interface quality^{21–23}. In contrast, most van der Waals magnets have an intrinsic magnetocrystalline anisotropy owing to the reduced crystal symmetry of their layered structures. Several layered magnets exhibit effective 2D magnetic interactions even in their bulk crystal form^{24,25}. This implies that it might be possible to retain a magnetic ground state in a monolayer. In addition to enabling the study of magnetism in naturally formed crystals in the true 2D limit, layered magnets provide a platform for studying the thickness dependence of magnetism in isolated single crystals where the interaction with the underlying substrate is weak. These covalently bonded van der Waals layers prevent complex magnetization reorientations induced by epitaxial lattice reconstruction and strain²³. For layered materials, these advantages come at a low fabrication cost, since the micromechanical exfoliation technique is much simpler than conventional approaches requiring sputtering or sophisticated molecular beam epitaxy.

A variety of layered magnetic compounds have recently been investigated to determine whether their magnetic properties can be retained down to monolayer thickness^{12–14,26}. Recent Raman studies suggest ferromagnetic ordering in few-layer Cr₂Ge₂Te₆ and antiferromagnetic ordering in monolayer FePS₃^{27,28}. However, no evidence yet exists for ferromagnetism persisting down to the monolayer limit. One promising candidate is bulk crystalline CrI₃. It shows layered Ising ferromagnetism below a Curie temperature (T_C) of 61 K with an out-of-plane easy axis (Fig. 1a and b, Extended Data Fig. 1)^{10,29}. Given its van der Waals nature, we expect magnetocrystalline anisotropy, which could possibly lift the Mermin–Wagner restriction to stabilize long-range ferromagnetic ordering even in a monolayer.

In our experiment, we obtained atomically thin CrI₃ flakes by mechanical exfoliation of bulk crystals onto oxidized silicon substrates (see Methods for CrI₃ crystal growth¹⁰ and fabrication details). Given the reactivity of CrI₃ flakes, sample preparation was carried out in a glove box under an inert atmosphere. We mainly employed optical contrast based on the pixel red–green–blue (RGB) value to index the number of layers in a flake (see Methods for quantitative optical microscopy in CrI₃ and Extended Data Figs 2–4). Figure 1c is an optical micrograph of a typical multi-step CrI₃ flake on a 285-nm SiO₂/Si substrate, showing regions ranging from 1 to 6 layers in thickness. Figure 1d shows an optical contrast map of the same region illuminated by 631-nm-filtered light. The extracted optical contrast as a function of layer thickness is in good agreement with models based on the Fresnel equations (Fig. 1e). To accurately determine the correspondence between optical contrast and flake thickness, we also measured the thickness of CrI₃ flakes by atomic force microscopy, determined to be 0.7 nm per layer, after encapsulation with few-layer graphene (see Extended Data Fig. 5).

To probe the magnetic order, we employed polar magneto-optical Kerr effect (MOKE) measurements as a function of applied external magnetic field perpendicular to the sample plane (Faraday geometry). This design is sensitive to out-of-plane magnetization, and can detect small Kerr rotations, θ_K , of linearly polarized light down to 100 μ rad using an alternating-current polarization modulation technique as laid out in Extended Data Fig. 6. All optical measurements were carried out using a 633-nm HeNe laser and at a temperature of 15 K, unless otherwise specified. Figure 1f illustrates the MOKE signal from a thin bulk flake of CrI₃. The observed hysteresis curve and remanent θ_K at zero magnetic field $\mu_0 H = 0$ T (where μ_0 is the magnetic constant) are hallmarks of ferromagnetic ordering, consistent with its bulk ferromagnetism with out-of-plane magnetization. The negative remanent θ_K when approaching zero field from a positive external field is a consequence of thin-film interference from reflections at the CrI₃–SiO₂ and SiO₂–Si interfaces (see Methods for thin-film interference and Extended Data Fig. 7).

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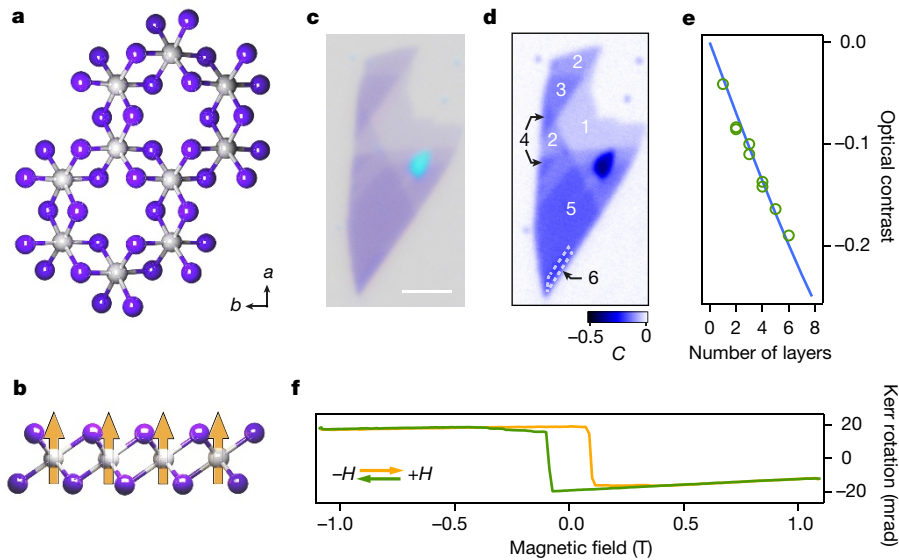


Figure 1 | Crystal structure, layer thickness identification, and MOKE of bulk CrI₃. **a**, View of the in-plane atomic lattice of a single CrI₃ layer. Grey and purple balls represent Cr and I atoms, respectively. The Cr³⁺ ions are coordinated to six I⁻ ions to form edge-sharing octahedra arranged in a hexagonal honeycomb lattice. **b**, Out-of-plane view of the same CrI₃ structure depicting the Ising spin orientation. **c**, Optical micrograph of a

representative CrI₃ flake. **d**, Calculated optical contrast map of the same flake with a 631-nm optical filter. The scale bar in **c** is 3 μm . **e**, Averaged optical contrast of the steps of the sample with different numbers of layers (circles) fitted by a model based on Fresnel's equations (solid line). **f**, Polar MOKE signal of a thin bulk CrI₃ crystal.

Remarkably, the ferromagnetic ordering remains in the monolayer limit. Figure 2a shows θ_K as a function of $\mu_0 H$ for a monolayer CrI₃ flake (inset to Fig. 2a). A single hysteresis loop in θ_K centred around $\mu_0 H = 0$ T, with a non-zero remanent Kerr rotation, demonstrates out-of-plane spin polarization. This implies Ising ferromagnetism in monolayer CrI₃. As expected, θ_K is independent of the excitation power (Fig. 2b). In the following, all data are taken with an excitation power of 10 μW . We have measured a total of 12 monolayer samples, which

show similar MOKE behaviour with consistent remanent θ_K values of about 5 ± 2 mrad at $\mu_0 H = 0$ T (Extended Data Fig. 8a). The coercive field ($\mu_0 H_c$), which is approximately 50 mT for the sample in Fig. 2a, can vary between samples owing to the formation of domain structures in some samples.

Figure 2c shows spatial maps of θ_K for another monolayer, taken at selected magnetic field values. After cooling the sample from above T_C at $\mu_0 H = 0$ T, the entire monolayer is spontaneously magnetized (in

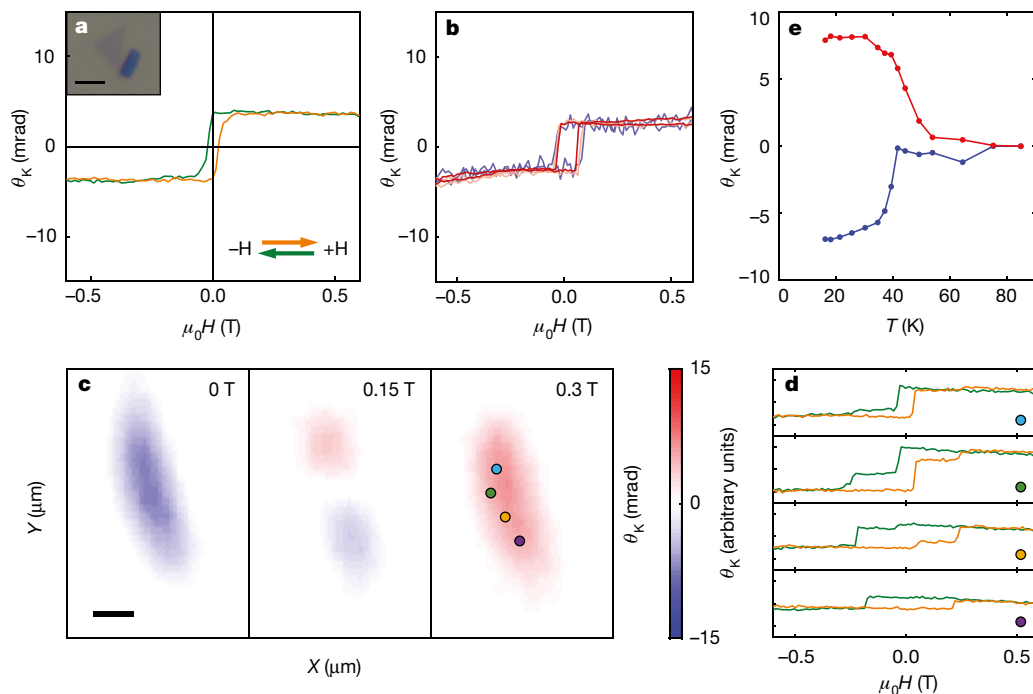


Figure 2 | MOKE measurements of monolayer CrI₃. **a**, Polar MOKE signal for a CrI₃ monolayer. The inset shows an optical image of an isolated monolayer (the scale bar is 2 μm). **b**, Power dependence of the MOKE signal taken at incident powers of 3 μW (blue), 10 μW (pink), and 30 μW (red). **c**, MOKE maps at $\mu_0 H = 0$ T, 0.15 T and 0.3 T on a different

monolayer. The scale bar is 1 μm . **d**, θ_K versus $\mu_0 H$ sweeps taken at four points marked by dots on the $\mu_0 H = 0.3$ T map in **c**. **e**, Temperature dependence of MOKE signal with the sample initially cooled at $\mu_0 H = 0$ T (blue) and 0.15 T (red).

blue, defined as spin down). As the field is increased to 0.15 T, the magnetization in the upper half of the flake switches direction (now spin up, in red). As the field is further increased to 0.3 T, the lower half of the monolayer flips and the entire flake becomes spin up, parallel to $\mu_0 H$. This observation of micrometre-scale lateral domains suggests different values of coercivity in each domain. Indeed, magnetic field sweeps (θ_K versus $\mu_0 H$) taken at discrete points ranging across both domains (Fig. 2d) show the difference in coercive field between the upper and lower half of the monolayer. Sweeps taken only on the upper domain (marked by a blue circle) show a much narrower hysteresis loop (about 50 mT) than sweeps from spots on the lower domain (orange and purple circles, about 200 mT). When the beam spot is centred between the two domains, contributions from both can be seen in the resulting hysteresis loop (green circle), a consequence of the approximately 1- μm beam spot illuminating both domains.

To determine the monolayer T_C , we perform an analysis of the irreversible field-cooled and zero-field-cooled Kerr signal. Zero-field-cooled sweeps were performed by measuring θ_K while cooling the sample in zero field. After warming up to a temperature well above T_C (90 K), the field-cooled measurement is taken upon cooling down in the presence of a small external magnetic field ($\mu_0 H = 0.15$ T). Thermomagnetic irreversibility can be observed below T_C , at which point the zero-field-cooled sweep and the field-cooled sweep diverge as illustrated in Fig. 2e. We measured the average T_C for the monolayer samples to be 45 K, slightly lower than the value (61 K) for bulk samples.

The layered structure of CrI_3 provides a unique opportunity to investigate ferromagnetism as a function of layer thickness. Figure 3a–c shows θ_K versus $\mu_0 H$ for representative 1–3-layer CrI_3 samples. All measured monolayer and trilayer samples consistently show ferromagnetic behaviour with a single hysteresis loop centred at $\mu_0 H = 0$ T (Fig. 3a and c and Extended Data Fig. 8). Both remanent and saturation values of θ_K for trilayers are about 50 ± 10 mrad, which is an order of magnitude larger than for monolayers. This drastic change in θ_K on moving from monolayer to trilayer may be due to a layer-dependent electronic structure, leading to weaker optical resonance effects at 633 nm for the monolayer than for the trilayer (see Extended Data Fig. 9 and Methods for thin-film interference and MOKE signal in CrI_3). We find that for trilayers and thin bulk samples, T_C is consistent with the bulk value of 61 K. The relatively small decrease of T_C from bulk to few-layer and monolayer samples suggests that interlayer interactions do not dominate the ferromagnetic ordering in CrI_3 . Compared with metallic magnetic thin films whose magnetic properties strongly depend on the underlying substrate³⁰, the weak layer-dependent T_C also implies a negligible substrate effect on the ferromagnetic phenomena in atomically thin CrI_3 . As such, exfoliated CrI_3 of all thicknesses can be regarded as isolated single crystals.

A further observation is that bilayer CrI_3 shows a markedly different magnetic behaviour from the monolayer (Fig. 3b). For all ten bilayer samples measured, the MOKE signal is strongly suppressed, with θ_K approaching zero (subject to slight variation between samples, see Extended Data Fig. 8b) at field values ± 0.65 T. This observation implies a compensation for the out-of-plane magnetization. Upon crossing a critical field, θ_K shows a sharp jump, depicting a sudden recovery of the out-of-plane co-parallel orientation of the spins. This new magnetic state has a saturation θ_K (40 ± 10 mrad) an order of magnitude larger than that of monolayer samples, and slightly smaller than for trilayers.

The suppression of the Kerr signal at zero magnetic field demonstrates that the ground state has zero out-of-plane magnetization. The plateau behaviour of the magnetization curve—showing three horizontal regimes between magnetic field values of -1.1 T and 1.1 T (Fig. 3b)—further implies that there are no in-plane spin components; otherwise, one would expect a gradual increase of the MOKE signal with increasing perpendicular magnetic field. Rather, our observation suggests that each individual layer is ferromagnetically ordered (out-of-plane) while the interlayer coupling is antiferromagnetic. In this case, the strength of the interlayer coupling determines the field

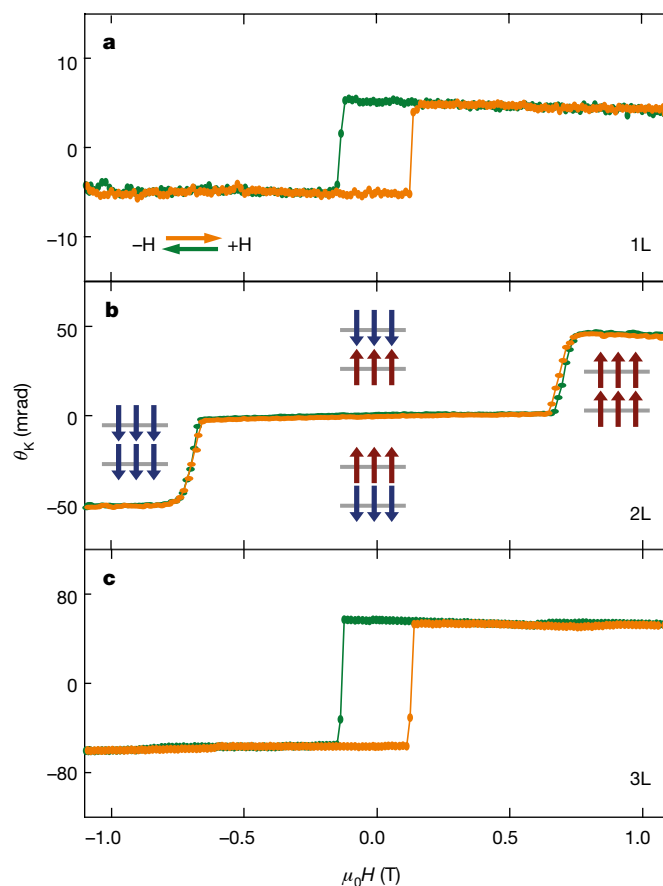


Figure 3 | Layer-dependent magnetic ordering in atomically-thin CrI_3 . **a**, MOKE signal on a monolayer (1L) CrI_3 flake, showing hysteresis in the Kerr rotation as a function of applied magnetic field, indicative of ferromagnetic behaviour. **b**, MOKE signal from a bilayer CrI_3 showing vanishing Kerr rotation for applied fields ± 0.65 T, suggesting antiferromagnetic behaviour. Insets depict bilayer (2L) magnetic ground states for different applied fields. **c**, MOKE signal on a trilayer (3L) flake, showing a return to ferromagnetic behaviour.

at which jumps between different plateaus occur, ± 0.65 T. Although the detailed mechanism of this coupling remains unclear, the different magnetic phases observed in bilayers and trilayers emphasizes the strong layer-dependent interplay between different mechanisms that stabilize magnetic ordering in the atomically thin limit.

Another bilayer feature distinct from those of monolayers is the vanishingly small hysteresis around the jumps, suggesting negligible net perpendicular anisotropy. A possible interpretation is that the shape anisotropy (which prefers in-plane spin orientation) nearly compensates for the intrinsic magnetocrystalline anisotropy (which prefers out-of-plane spin orientation) so that the overall anisotropy is close to zero.

The insets in Fig. 3b display the layer-by-layer switching behaviour that leads to plausible magnetic ground states of bilayer CrI_3 . When the magnetic field is ± 0.65 T, the magnetization of the two layers are oppositely oriented to one another. Thus, the net magnetization vanishes and bilayer CrI_3 behaves as an antiferromagnet with an exchange field of about 0.65 T. When $|\mu_0 H| > 0.65$ T, magnetization in one layer flips to align with the external magnetic field and restores out-of-plane magnetization, giving rise to the large MOKE signal. At around $|\mu_0 H| = 0.65$ T, the MOKE signal sharply increases from near zero to its saturation value within about 100 mT, suggesting an abrupt increase of out-of-plane magnetization triggered by a small change of magnetic field. Such behaviour is indicative of metamagnetism, the magnetic-field-driven transition from antiferromagnetic ordering to a fully spin-polarized state²⁰.

In summary, here we demonstrated 2D ferromagnetism in exfoliated monolayer CrI₃. The observed monolayer T_C suggests that these 2D magnets are weakly coupled to their substrates and can be regarded as isolated magnets. This is in distinct contrast with conventional metallic monolayer films, whose magnetism is strongly affected by substrate coupling. We also observed strong evidence for layer-dependent magnetic phases, from ferromagnetism in the monolayer, to antiferromagnetism in the bilayer, and back to ferromagnetism in the trilayer and bulk. We envision that the demonstration of intrinsic ferromagnetism in monolayer CrI₃ as well as its layer-dependent magnetic behaviour will provide opportunities for the investigation of quantum phenomena, such as topological effects in hybrid superconducting–ferromagnetic van der Waals heterostructures as well as the engineering of new magneto-optoelectronic devices at low temperature, such as ferromagnetic light emitters.

Online Content Methods, along with any additional Extended Data display items and Source Data, are available in the online version of the paper; references unique to these sections appear only in the online paper.

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Author Contributions X.X. and P.J.-H. supervised the project. E.N.-M. and M.A.M. synthesized and characterized the bulk CrI₃ crystal. E.N.-M. and D.R.K. fabricated the samples and analysed the layer thickness, assisted by G.C. and B.H. B.H. built the MOKE setup with help from E.S. and D.Z. G.C. and B.H. performed the MOKE measurements, assisted by K.L.S. and E.N.-M. R.C., D.X. and W.Y. provided theoretical support. B.H., G.C., E.N.-M., X.X., P.J.-H., D.X. and D.H.C. wrote the paper with input from all authors. All authors discussed the results.

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METHODS

Growth of chromium(III) iodide bulk crystals. Chromium powder (99.5%, Sigma-Aldrich) and anhydrous iodine beads (99.999%, Sigma-Aldrich) were mixed in a 1:3 ratio inside a glove box with an argon atmosphere. 1.5 g of the mixture was then loaded in a silica ampoule (16 mm of inner diameter, 19 mm of outer diameter and 550 mm in length). The ampoule was extracted from the glove box and immediately evacuated to a pressure of approximately 10^{-4} torr. Once at that pressure, the closed end was dipped in liquid nitrogen to prevent the sublimation of the iodine beads. The ampoule was then flame-sealed under dynamic vacuum and placed inside a three-zone furnace. A three-zone furnace provides the best control over the growth process by reducing nucleation, leading to the growth of large-size single isolated crystals. Following an inverted gradient step of several hours, the crystals were grown over a period of 7 days with source zone at 650 °C (containing the solid mixture), middle growth zone at 550 °C and third zone at 600 °C. Crystals formed both in the source (lustrous hexagonal platelets of several millimetres in size) and the middle (millimetre-long ribbon-like flakes) zones. The crystals were extracted from the ampoule in an argon atmosphere and stored in anhydrous conditions. The I:Cr elemental ratio was verified to be 2.8 ± 0.2 in several crystals by energy-dispersive X-ray microanalysis performed on individual crystals in a Zeiss Merlin high-resolution scanning electron microscope equipped with an electron dispersive spectroscopy probe. To confirm the crystallographic phase of the material, a few single crystals were ground, loaded into a 0.3-mm outer-diameter capillary and mounted on a Rigaku Smartlab Multipurpose Diffractometer setup in converging beam configuration with a D/tEX detector. The room-temperature X-ray diffraction patterns of both the ribbon-like and hexagonal platelets were identical and consistent with the high-temperature monoclinic AlCl_3 -type structure ($C2/m$) reported for CrI_3 , with indexed unit cell parameters of $a = 6.8735(2)$ Å, $b = 11.8859(3)$ Å, $c = 6.9944(1)$ Å and $\beta = 108.535(2)^\circ$ (Le-Bail refinement, $R_{\text{Bragg}} = 5.27\%$). SQUID magnetometry performed on the single crystals depicts a T_C of 61 K and a saturation magnetization of $3\mu_B$ per Cr atom, also in agreement with the values reported in the literature¹⁰.

Encapsulation of samples for atomic force microscopy. CrI_3 samples exfoliated under an inert atmosphere were encapsulated to preserve the CrI_3 flakes during atomic force microscopy (AFM) studies under ambient conditions. Using an all-dry viscoelastic stamping technique inside the glovebox, CrI_3 flakes were sandwiched between two layers of approximately 5-nm graphite to prevent reaction with oxygen and moisture. Encapsulated CrI_3 flakes could then be safely removed from the glove box for further study. AFM of graphite-encapsulated CrI_3 flakes was measured using a Bruker Dimension Edge atomic force microscope in tapping mode. Extended Data Fig. 5 shows AFM results for bilayer and trilayer CrI_3 flakes encapsulated in approximately 5-nm graphite layers. Their corresponding MOKE data are consistent with bare samples.

MOKE. Power-stabilized light from a 633-nm HeNe laser source was linearly polarized at 45° to the photoelastic modulator (PEM) slow axis (Extended Data Fig. 6)³¹. Transmitting through the PEM, the light was sinusoidally phase-modulated at 50.1 kHz, with a maximum retardance of $\lambda/4$. Upon phase modulation, the light was focused down onto the sample at normal incidence using an aspheric lens. At any point in time when the light was not circularly polarized, reflection off a magnetic sample would rotate the polarization axis (the major axis for elliptically polarized light) by what we define as the Kerr rotation, θ_K . This reflection was then separated from the incidence path via a laser line non-polarizing beamsplitter cube and projected onto the PEM slow axis using a second polarizer. For $\theta_K = 0$, the slow-axis component remains constant for all polarizations and hence is not time-dependent. With a non-zero θ_K , however, the slow-axis component depends on the polarization and oscillates at twice the modulation frequency, 100.2 kHz, with an amplitude that is proportional to the Kerr rotation. Therefore, to obtain θ_K , the reflection was detected using an amplified photodiode and two lock-in amplifiers: one tuned at 100.2 kHz to detect the Kerr rotation, and one tuned at the chopper frequency, 800 Hz, to normalize the Kerr signal to laser intensity fluctuations and the reflectivity of the sample.

Quantitative optical microscopy in CrI_3 . Optical microscopy images were taken using a Nikon Eclipse LV-CH 150NA optical microscope with a DS-Ri2 full-frame camera. The setup was located inside a glove box (argon atmosphere) to prevent sample degradation. The quantitative optical contrast analysis required that images were captured with a $100\times$ objective under monochromatic illumination at normal incidence. In practice, 10-nm full-width at half-maximum (FWHM) filters were used (Andover Corp.) to filter the light coming from a halogen lamp. The thickness of the flakes was determined by contact-mode AFM under ambient conditions. Given the extreme sensitivity of the samples to atmospheric moisture, the CrI_3 flakes were encapsulated between two pieces of few-layer graphite (typically 5 nm in thickness) before being extracted from the glove box. For each optical-microscopy-filtered image, individual RGB values were extracted and averaged over each flake and substrate region to give the reflected intensities of the flake

and substrate. The intensity was chosen to be exclusively the value of the channel with the highest number of counts. The experimental optical contrast value was then calculated according to the following expression (1).

$$C(d, \lambda) = \frac{I_{\text{flake}} - I_{\text{substrate}}}{I_{\text{flake}} + I_{\text{substrate}}} \quad (1)$$

Equation (1) expresses the relationship between the optical contrast C between each flake and the substrate using the reflected intensities from the flake (I_{flake}) and the substrate ($I_{\text{substrate}}$). Extended Data Fig. 1 shows an example of a contrast map of a multi-step CrI_3 flake extracted from its optical micrograph.

For flakes that have been exfoliated on a SiO_2/Si substrate, C depends on the thickness of the flake and on the illumination wavelength^{32,33}. Following the quantitative microscopy analysis proposed for graphene on SiO_2/Si substrates³⁴, C can be computed for any kind of flake by using a model based on Fresnel's equations shown in equations (2a) and (2b):

$$I_{\text{substrate}}(\lambda) = \left| \frac{r_{02} + r_{23}e^{-2i\phi_2}}{1 + r_{02}r_{23}e^{-2i\phi_2}} \right|^2 \quad (2a)$$

$$I_{\text{flake}}(\lambda) = \left| \frac{r_{02}e^{i(\phi_1+\phi_2)} + r_{12}e^{-i(\phi_1-\phi_2)} + r_{23}e^{-i(\phi_1+\phi_2)} + r_{01}r_{12}r_{23}e^{i(\phi_1-\phi_2)}}{e^{i(\phi_1+\phi_2)} + r_{01}r_{12}e^{-i(\phi_1-\phi_2)} + r_{01}r_{23}e^{-i(\phi_1+\phi_2)} + r_{12}r_{23}e^{i(\phi_1-\phi_2)}} \right|^2 \quad (2b)$$

In this calculation, the subscripts 0, 1, 2 and 3 refer to air (treated as vacuum), CrI_3 , SiO_2 and Si, respectively. The amplitude of the reflected path at the interface between media j and k is given by r_{jk} in equation (3b) and is calculated from the complex refractive indices defined in equation (3a). ϕ_j is the phase shift introduced by the interaction between light of wavelength λ and medium j with thickness d_j shown in equation (3c).

$$\tilde{n}_j(\lambda) = n_j - i\kappa_j \quad (3a)$$

$$r_{jk} = \frac{\tilde{n}_j - \tilde{n}_k}{\tilde{n}_j + \tilde{n}_k} \quad (3b)$$

$$\phi_j = \frac{2\pi\tilde{n}_j d_j}{\lambda} \quad (3c)$$

As can be noted from the previous expressions, if one wants to model C using the Fresnel equations, the complex index of refraction of the material under study must be known. The reflectivity of CrI_3 measured in vacuum at 300 K at normal incidence of a single-crystal platelet of CrI_3 has been previously reported³⁵. This data was used to calculate the phase of the amplitude reflection coefficient θ at energies in the visible range by numerical integration, according to the Kramers-Kronig relation (4a). The refractive index n and extinction coefficient κ of CrI_3 were then obtained throughout the visible range by combining equations (4b) and (4c) at each energy value. The results are plotted in Extended Data Fig. 3.

$$\theta(E) = -\frac{E}{\pi} \int_0^\infty \frac{\ln[R(E')]}{(E')^2 - E^2} dE' \quad (4a)$$

$$r(E) = \sqrt{R(E)} e^{i\theta(E)} \quad (4b)$$

$$r(E) = \frac{n(E) - 1 + i\kappa(E)}{n(E) + 1 + i\kappa(E)} \quad (4c)$$

Substituting for the complex indices of refraction³⁶ of CrI_3 , Si and SiO_2 in equations (2a) and (2b), we can calculate the expected value of C for flakes of different thicknesses as a function of the illumination wavelength. Extended Data Fig. 4a shows the contrast map for CrI_3 considering a fixed thickness of 285 nm of the SiO_2 layer in the SiO_2/Si substrate. We also present a line cut of that plot at an illumination wavelength of 635 nm in Extended Data Fig. 4b. It can be seen that the experimental data points closely follow the trend predicted by the model. Given that the method is non-destructive and can be performed inside a glove box for many different illumination wavelengths, the error in the determination of the number of layers can be reduced. This provides a fast and reliable method for the characterization of few-layer CrI_3 flakes.

Thin-film interferometry and the MOKE signal in CrI_3 . Linearly polarized light is an equal superposition of right-circularly and left-circularly polarized light (RCP,

LCP respectively). When a phase difference accrues between the RCP and LCP components, the polarization axis of the linearly polarized light rotates. This rotation can be observed in any material that exhibits circular birefringence. In magnetic samples, such as CrI₃, this birefringence arises from a non-zero magnetization, \mathbf{M} , and is known as the magneto-optical Kerr effect (MOKE) when detected in reflection geometry. A MOKE measurement then detects changes in \mathbf{M} by exploiting the functional dependence of the Kerr rotation on the magnetization, $\theta_K(\mathbf{M})$. Additional interference terms must be accounted for, however, when we discuss the Kerr rotation of a thin-film material, because reflections from the material–substrate (CrI₃–SiO₂) interface will superimpose with the reflection off the magnetic sample (Extended Data Fig. 7). As such, this motivates a model that uses the Fresnel equations (the same formalism as in Methods subsection ‘Quantitative microscopy on CrI₃’) to calculate the reflection coefficients for RCP (\tilde{r}_+) and for LCP (\tilde{r}_-) light. The phase difference between \tilde{r}_+ and \tilde{r}_- then, is the Kerr rotation:

$$\theta_K = \arg(\tilde{r}_+) - \arg(\tilde{r}_-) \quad (5)$$

To obtain the index of refraction for CrI₃ when fully spin polarized, we work in Cartesian coordinates and use a dielectric tensor of the form³⁷:

$$\epsilon = \begin{bmatrix} \tilde{\epsilon}_{xx} & iQM & 0 \\ -iQM & \tilde{\epsilon}_{xx} & 0 \\ 0 & 0 & \tilde{\epsilon}_{zz} \end{bmatrix} \quad (6)$$

where Q is the complex Voigt constant and M is the out-of-plane (parallel to the z -axis) component of the magnetization, which we assume is constant for all CrI₃ thicknesses when fully spin-polarized. This form of the dielectric tensor for a magnetic sample is valid assuming (1) that the crystal exhibits at least threefold symmetry; (2) that \mathbf{M} is parallel to the axis of rotation that gives rise to the threefold symmetry; and (3) that the axis of rotation is chosen to be the z axis. Solving for the normal modes, we obtain the eigenvalues:

$$\tilde{n}_{\pm} = \sqrt{\tilde{\epsilon}_{xx} \pm QM} \quad (7)$$

and the eigenvectors:

$$\mathbf{D}_{\pm} = (E_x \pm iE_y)\hat{z} \quad (8)$$

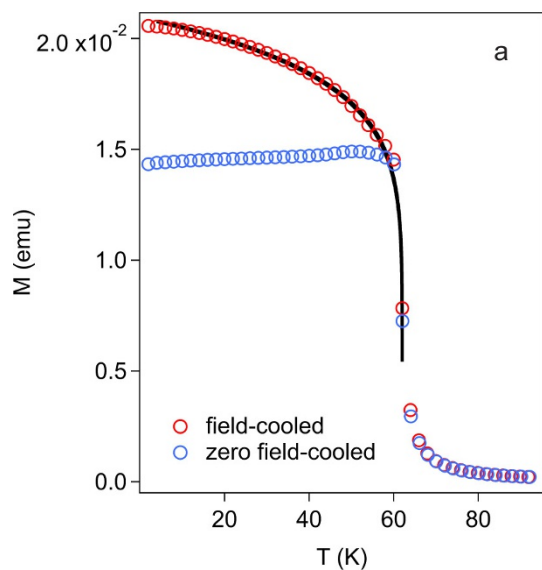
where \tilde{n}_+ and \mathbf{D}_+ denote the eigenvalue and eigenvector respectively of RCP light and \tilde{n}_- and \mathbf{D}_- denote the eigenvalue and eigenvector respectively of LCP light in CrI₃. The complex dielectric component $\tilde{\epsilon}_{xx}$, when related to the \tilde{n} defined in equation (3a), is:

$$\tilde{\epsilon}_{xx} = n^2 - \kappa^2 - 2in\kappa \quad (9)$$

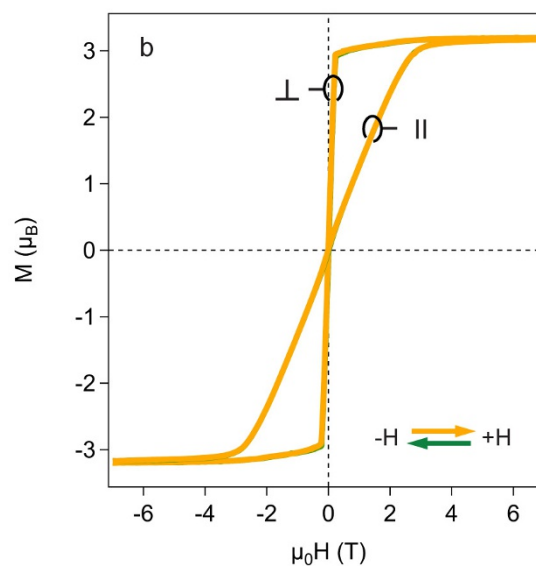
where the value of $\tilde{\epsilon}_{xx}$ was derived from the n and κ values at about 1.96 eV (modelled in Extended Data Fig. 3). It is apparent from equation (7) that we should not expect θ_K to depend linearly on \mathbf{M} and layer number. In addition, interference from reflections off the CrI₃–SiO₂ interface will give rise to a non-trivial functional form of θ_K with respect to layer thickness. There is no determination of Q in the literature for CrI₃, so we varied Q as a complex parameter and constrained it to a small range that fitted experimental θ_K values from our MOKE measurements on trilayer and bulk CrI₃. These calculations qualitatively describe the large increase in θ_K moving from monolayer to trilayer, as well as the negative θ_K seen at positive μ_0H for bulk flakes. However, this simple model does not incorporate layer-dependent electronic structure changes, seen in other atomically thin van der Waals materials such as¹⁸ MoS₂, and evident in this system as a change in magnetic ground states from monolayer to bilayer.

Data availability. The data sets generated during and analysed during this study are available from the corresponding author upon reasonable request.

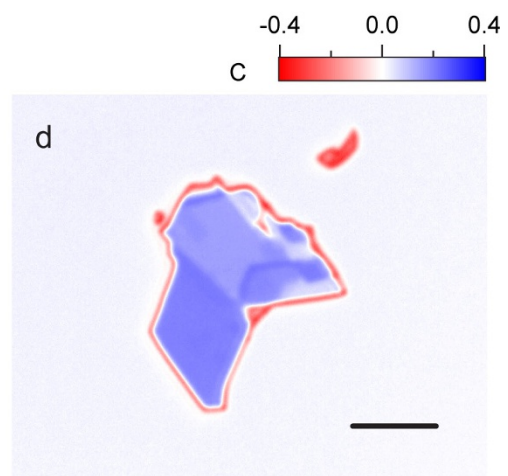
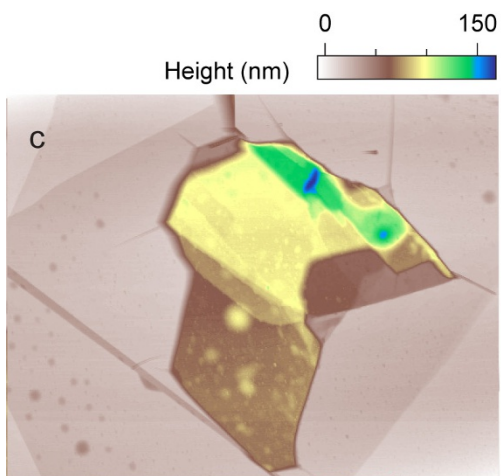
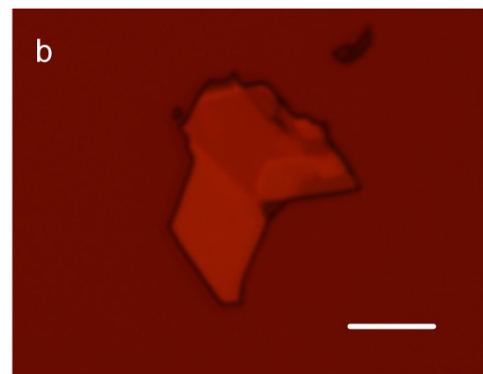
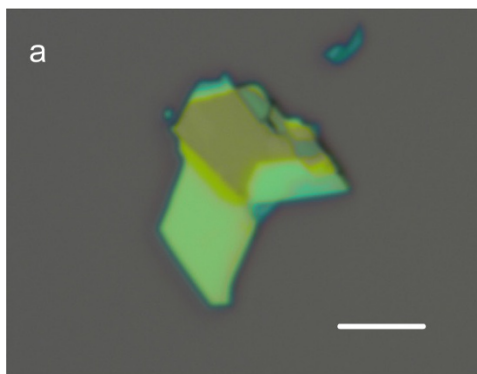
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Extended Data Figure 1 | SQUID magnetometry in bulk CrI₃. **a**, Zero-field-cooled/field-cooled temperature dependence of the magnetization of a CrI₃ bulk crystal with an applied magnetic field of 10 G perpendicular to the basal plane of the sample. The black line is a criticality fit

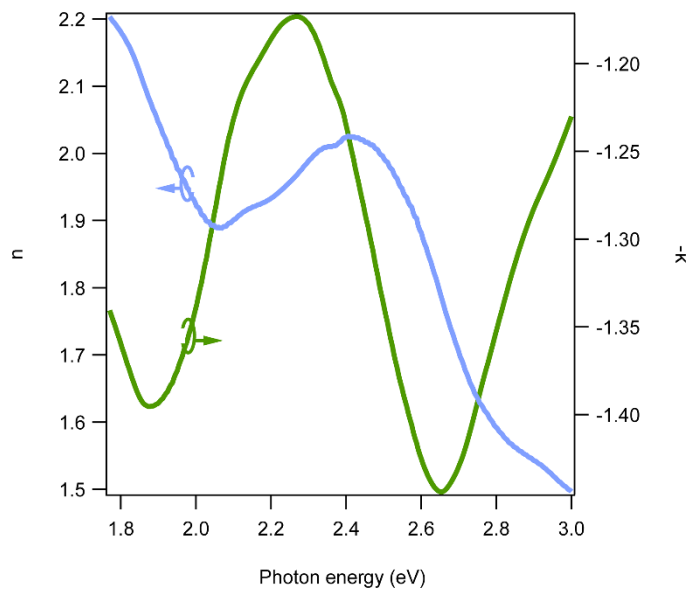


$(M = \alpha(1 - T_C/T)^\beta)$ of the data with $T_C = 61$ K and $\beta = 0.125$ (Ising universality class). **b**, Hysteresis loops of the same sample with the external magnetic field in perpendicular and parallel orientations with respect to the CrI₃ layers. (emu, electromagnetic units.)

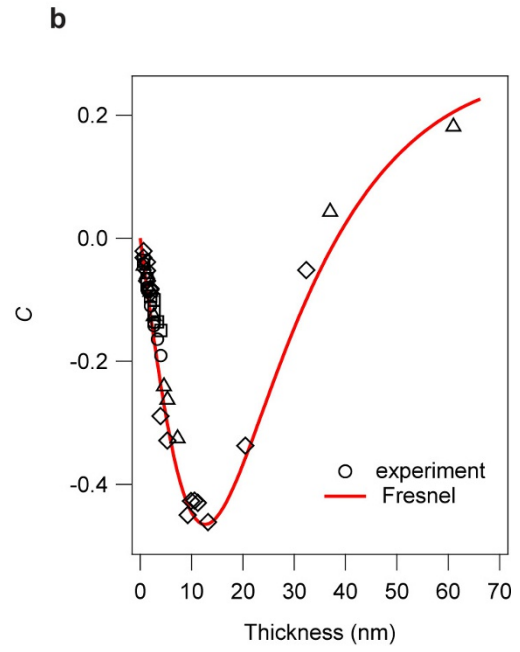
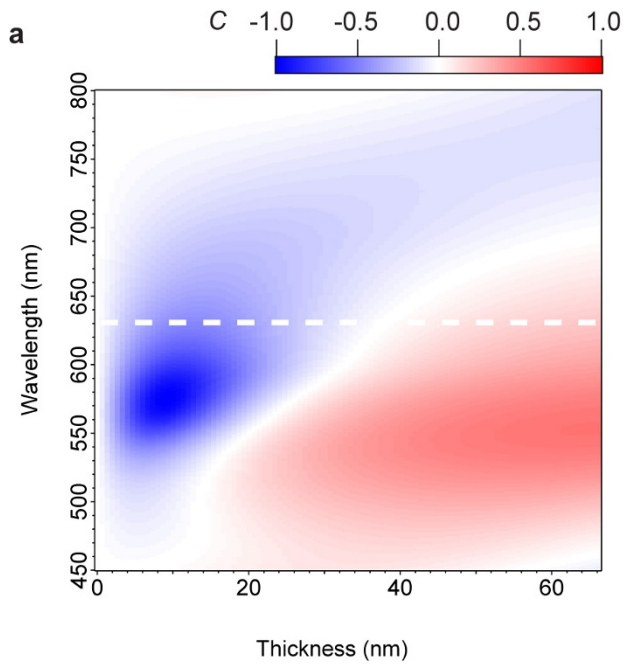


Extended Data Figure 2 | Thickness dependence of the optical contrast of the CrI₃ flakes. Optical micrographs of a CrI₃ flake illuminated with white light (a) and with 631-nm-(10-nm FWHM bandpass)-filtered light (b).

c, AFM topography image of the same sample. d, Optical contrast map extracted from the 631-nm micrograph in b. Scale bars are 5 μm .

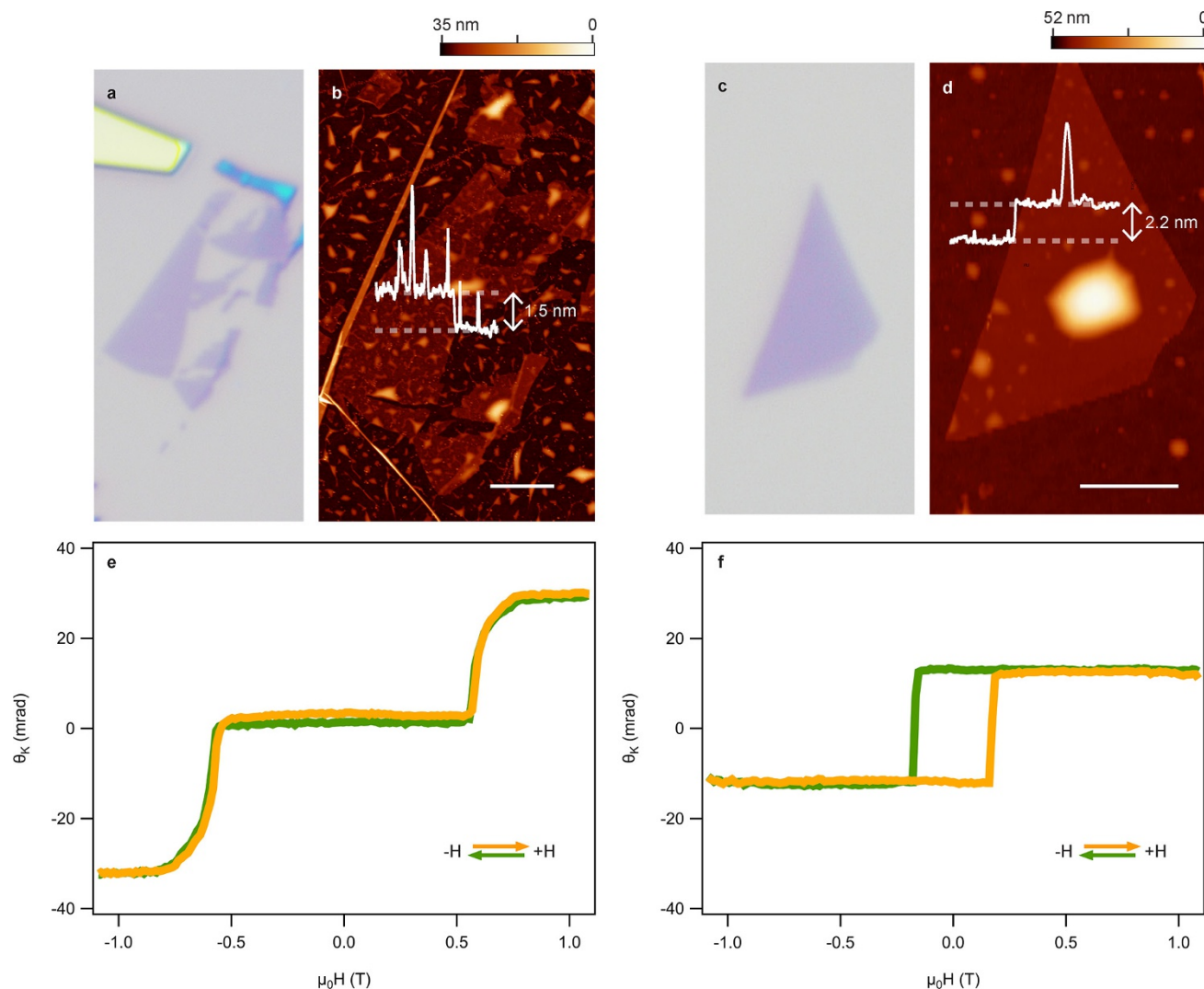


Extended Data Figure 3 | Computed index of refraction of bulk CrI₃. Real (n) and imaginary (κ) components are plotted as a function of photon energy in the visible range.



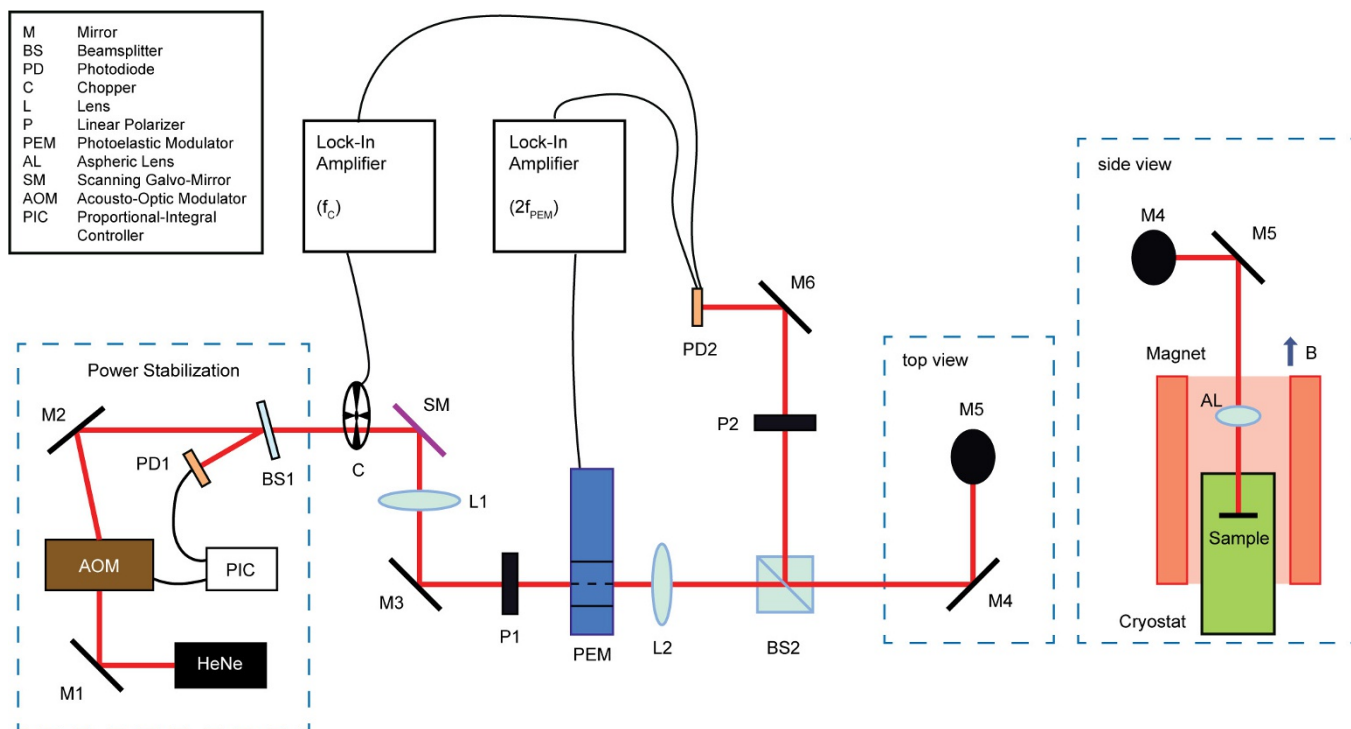
Extended Data Figure 4 | Fresnel model for the optical contrast C of CrI_3 flakes on $\text{Si}/285 \text{ nm SiO}_2$ substrates. a, Dependence of C with the number of layers for a CrI_3 flake as a function of the illumination wavelength. **b,** Comparison of the experimental data with the computed

thickness dependence of C for a red-light-illuminated sample (line cut at 631 nm as shown by the dashed line in a). The different shape markers indicate data coming from different exfoliated samples.



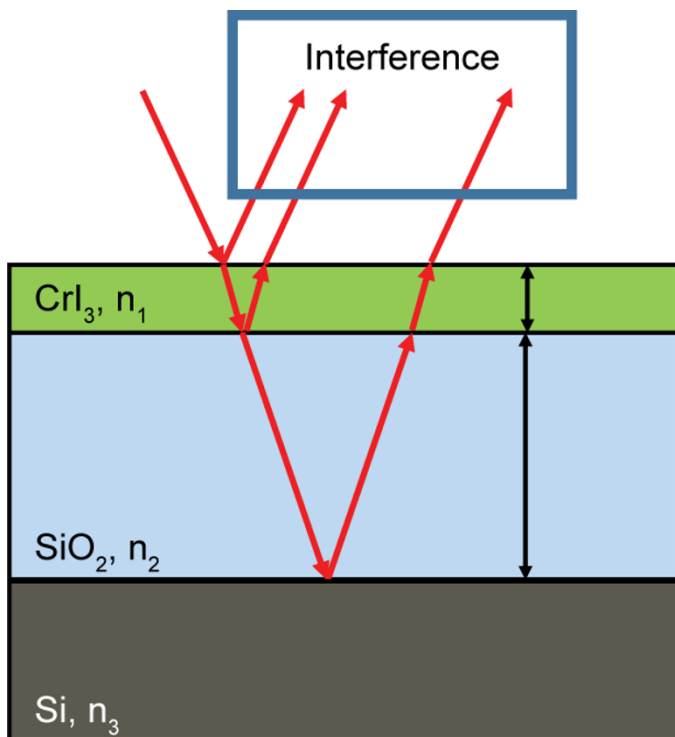
Extended Data Figure 5 | AFM and MOKE measurements of graphite-encapsulated few-layer CrI_3 . **a**, Optical microscope image of a bilayer CrI_3 flake on 285-nm-thick SiO_2 . **b**, AFM data for the CrI_3 flake in **a** encapsulated in graphite, showing a line cut across the flake with a step height of 1.5 nm. **c**, Optical microscope image of a trilayer CrI_3

flake on 285-nm-thick SiO_2 . **d**, AFM data for the CrI_3 flake shown in **c** encapsulated in graphite. A line cut taken across the flake shows a step height of 2.2 nm. Both scale bars are 2 μm . **e** and **f** show the MOKE signal as a function of applied magnetic field for the encapsulated bilayer in **b** and the encapsulated trilayer in **d** respectively.

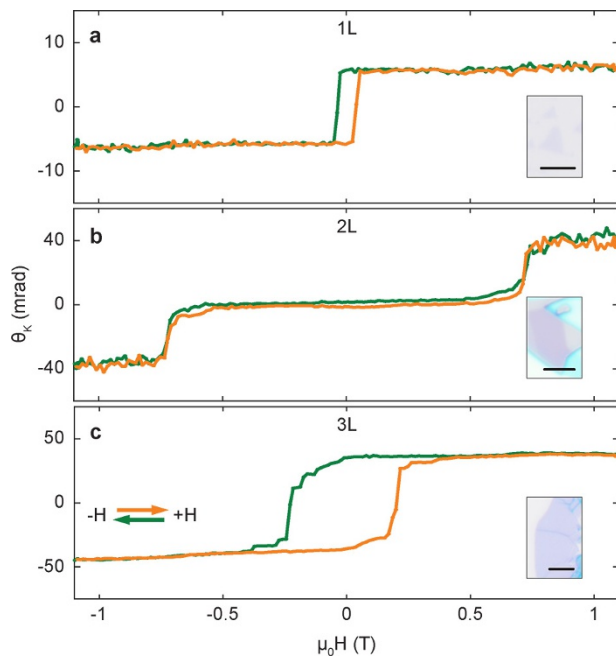


Extended Data Figure 6 | Magneto-optical Kerr effect experimental setup. Schematic of the optical setup used to measure the MOKE effect in CrI_3 samples. 633 nm optical excitation is provided by a power-stabilized HeNe laser. A mechanical chopper and photoelastic modulator provide intensity and polarization modulation, respectively. The modulated beam is directed through a polarizing beam splitter to the sample, housed in

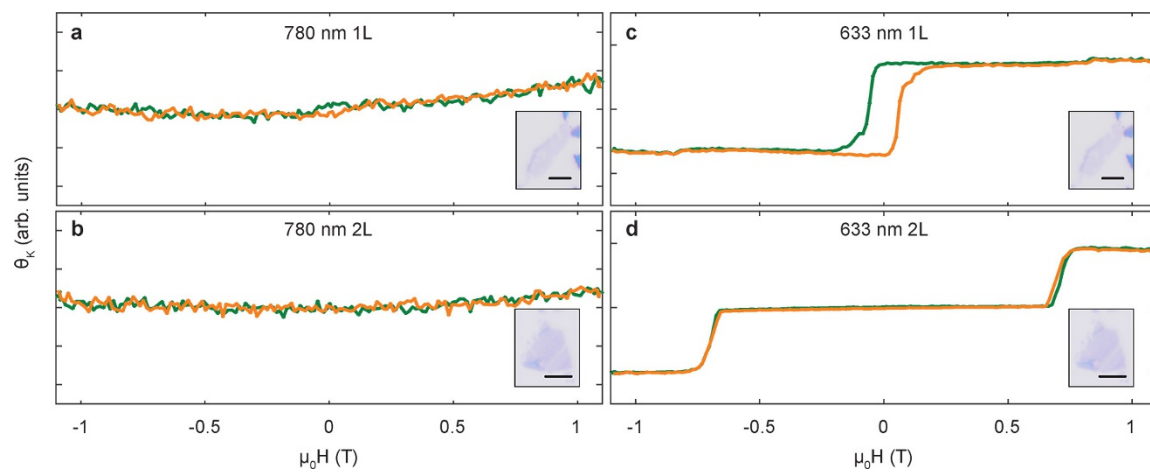
a closed-cycle cryostat at 15 K. A magnetic field is applied at the sample using a 7-T solenoidal superconducting magnet in Faraday geometry. The reflected beam passes through an analyser onto a photodiode, where lock-in detection measures the reflected intensity (at f_c) as well as the Kerr rotation (at f_{PEM}).



Extended Data Figure 7 | Thin-film interference ray diagram of CrI_3 on a silicon oxide/silicon substrate. Light incident on CrI_3 undergoes reflections at the CrI_3 - SiO_2 interface (green-blue boundary) as well as the SiO_2 - Si interface (blue-grey boundary). These reflections interfere with the initial reflection off the CrI_3 flake to produce thin-film interference that depends on the CrI_3 layer thickness, d_1 , as well as the SiO_2 thickness, d_2 . The underlying silicon wafer is assumed to be semi-infinite. The indices of refraction for CrI_3 , SiO_2 , and Si are n_1 , n_2 and n_3 respectively.



Extended Data Figure 8 | Additional data for 1–3-layer samples showing Kerr rotation as a function of the applied magnetic field. The insets show optical microscope images of the CrI₃ flakes at 100× magnification. **a**, Additional monolayer data showing ferromagnetic hysteresis and remanent Kerr signal as a function of applied magnetic field. **b**, Additional bilayer data showing field-dependent behaviour consistent with an antiferromagnetic ground state. **c**, Additional trilayer data showing field-dependent behaviour consistent with a ferromagnetic ground state, as well as a larger remanent Kerr signal than the monolayer in **a**. All scale bars are 5 μm.



Extended Data Figure 9 | Additional data for monolayer and bilayer samples under 780-nm excitation and 633-nm excitation. Insets show optical microscope images of the CrI₃ flakes at 100 \times magnification.

a, Kerr rotation as a function of applied magnetic field for a monolayer **a** (or **c**), and a bilayer **b** (or **d**) under 780-nm (or 633-nm) excitation. All scale bars are 4 μm .