Determining the phase diagram of atomically thin layered antiferromagnet CrCl$_3$

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Changes in the spin configuration of atomically thin, magnetic van der Waals multilayers can cause drastic modifications in their opto-electronic properties. Conversely, the opto-electronic response of these systems provides information about the magnetic state, which is very difficult to obtain otherwise. Here, we show that in CrCl$_3$ multilayers, the dependence of the tunnelling conductance on applied magnetic field, temperature and number of layers tracks the evolution of the magnetic state, enabling the magnetic phase diagram to be determined experimentally. Besides a high-field spin-flip transition occurring for all thicknesses, the in-plane magnetoconductance exhibits an even–odd effect due to a low-field spin-flop transition. Through a quantitative analysis of the phenomena, we determine the interlayer exchange coupling as well as the layer magnetization and show that in CrCl$_3$, shape anisotropy dominates. Our results reveal the rich behaviour of atomically thin layered antiferromagnets with weak magnetic anisotropy.

The recent discovery of magnetism in atomically thin exfoliated layers$^{1–14}$ is a major breakthrough in the field of two-dimensional (2D) materials and heterostructures$^{15–19}$. The giant tunnelling magnetoresistance observed in CrI$_3$, multilayers$^{20–23}$, for instance, shows that semiconducting van der Waals (vdW) antiferromagnets are interesting systems$^{24–27}$, with potential relevance for technology$^{28–30}$, once room-temperature operation is achieved. It is therefore important to identify—and determine experimentally—the microscopic parameters governing their behaviour. In CrI$_3$, the observed phenomenology originates from transitions in which individual layers abruptly flip their magnetization under the application of a perpendicular magnetic field$^{11–15}$ ($H$), as expected when the uniaxial magnetic anisotropy is the dominating energy scale$^{27,28}$. The case of weak anisotropy is different, as a richer variety of magnetic transitions is predicted to occur at lower field$^{29,30}$, and indeed spin-flop phases$^{31}$ are well established in bulk vdW antiferromagnets$^{32}$. However, the phase diagram of weakly anisotropic atomically thin vdW antiferromagnets, where finite-size effects can give rise to new phenomena$^{25–28}$, has not been determined yet.

Here, we investigate atomically thin multilayers of CrCl$_3$, a layered vdW antiferromagnetic semiconductor with a small anisotropy that favours spins to lie in the layers$^{39–40}$ (Fig. 1a, see Supplementary Notes 1 and 2 and Supplementary Figs. 1 and 2). We show that the sensitivity of the tunnelling current to the magnetic state tracks the phase diagram of these systems as a function of temperature and magnetic field. The measurements reveal that, down to bilayers, the antiferromagnetic alignment of spins in neighbouring layers is retained, with an easy-plane magnetic anisotropy of magnetostatic origin (that is, shape anisotropy). In an external magnetic field, a spin-flop transition is observed for all thicknesses. The dependence of the corresponding critical field on number of layers and field orientation allows us to estimate the strength of the interlayer exchange coupling and the magnitude of the sublattice magnetization. When $H$ is parallel to the layers, we reveal a prominent even–odd effect, with a spin-flop transition occurring at finite $H$ only for odd number of layers ($N$), due to the finite Zeeman energy of the magnetization of one uncompensated layer.

Tracking magnetic phase boundaries

Our experiments rely on transport measurements done on hBN-encapsulated graphite/CrCl$_3$/graphite tunnel junctions (see inset of Fig. 1b for a scheme and Supplementary Figs. 3 and 4 and Methods for details and information about device assembly). At low temperature ($T$), all devices show tunnelling transport. For thin samples, electrons from the graphite electrodes tunnel directly through the barrier, giving rise to a linear current–voltage ($I–V$) curve, as shown in Fig. 1b. For thicker devices, the current due to direct tunnelling is too small to be detected and a large bias has to be applied. The resulting $I–V$ curves are strongly non-linear, with $\ln(V)$ scaling proportional to $1/V$, as characteristic for the Fowler–Nordheim tunnelling regime$^{41}$.

We measure the tunnelling conductance, $G$, as a function of magnetic field $H$ applied parallel to the layers. Sizeable magnetoconductance is observed in all devices (see Fig. 1c), whose amplitude reaches a few hundred percent (depending on the applied bias and sample thickness), smaller than in CrI$_3$, but much larger than in non-magnetic tunnel barriers. $G$ smoothly increases with increasing $H$ and saturates at a critical value $G_c$, as expected for spins evolving from an antiferromagnetic state at $H = 0$ to ferromagnetic ordering at saturation. The value of $G_c$ depends on thickness, and increases from ~1.1 T in bilayers to ~1.9 T in eight-layer and thicker samples. To investigate magnetic transitions, we focus on the evolution of the derivative of the conductance (d$G$/d$H$) as a function of $H$ and $T$: being sensitive to changes in the relative orientation of the magnetization in the individual layers, d$G$/d$H$ strongly amplifies transitions between different phases.

A sharp jump in d$G$/d$H$, occurring at an applied field critical corresponding to the saturation field $H_c$, is observed in all devices, with Fig. 2a showing representative data measured on bi- and...
Fig. 1 | Tunnelling conductance through CrCl₃ multilayers. a, Schematic representation of the crystal structure and of the magnetic moments in adjacent CrCl₃ layers in the absence of an applied magnetic field (grey spheres represent the Cr atoms that are arranged in a honeycomb structure, while Cl atoms are represented in green). At temperatures well below the critical temperature, all spins in a given layer point in the same direction as a consequence of the strong ferromagnetic intralayer exchange coupling, while the weak interlayer coupling is antiferromagnetic and favours the spins to be antiparallel in neighbouring layers. b, Low-temperature I–V curves of tunnel barriers formed by CrCl₃ multilayers of different thickness (see values of N in the legend), measured at zero applied magnetic field. Inset: schematic representation of the graphite/CrCl₃/graphite tunnel junction devices (in actual devices, the tunnel barrier is encapsulated in between a bottom and a top hBN layers to avoid degradation). c, Low-temperature magnetoconductance of CrCl₃ multilayer tunnel barriers, as a function of magnetic field H applied parallel to the layers (normally, no specific orientation of H relative to the crystallographic directions was selected; in a few cases, such as in Supplementary Fig. 5, we intentionally compared measurements with different orientations to check that the direction of the in-plane field has no notable effect on phenomena discussed here). The applied voltage is increased on increasing the device thickness (V = 10 mV for N = 2, V = 0.7 V for N = 3, V = 1.1 V for N = 5 and V = 1.8 V for N = 8) to ensure that the tunnelling current is large enough to be measured. In b,c, the measurement temperature is 0.3 K for samples with N = 2, 3, 5 and 2 K for N = 8.

The colour maps in Fig. 2b,f illustrate the dependence of $dG/dH$ on $T$ and $H$. The magnetic field at which the jump in $dG/dH$ occurs decreases on increasing $T$ and $H$, and the maximum conductance is reached at a critical Néel temperature $T_N \approx 17$ K (not far from the bulk value, see Supplementary Note 2). The observed evolution resembles the phase boundary of a second-order transition, and is consistent with the $T$ dependence of $G$, measured at fixed $H$ (see Fig. 2c,g and Supplementary Fig. 6). As shown in Fig. 2d,h the data points obtained from either the temperature dependence of $G$ (blue circles) or from the magnetic field dependence of $dG/dH$ (empty squares) are in perfect quantitative agreement and trace the same phase boundary. Trilayers exhibit one additional feature at low field, corresponding to a minimum at finite $H = H_s$, where $dG/dH$ vanishes (Fig. 2e). On varying $T$, this low-field $dG/dH$ minimum results in the blue region in the colour map of Fig. 2f, from which we extract the temperature evolution of $H_s$ (see the triangles in Fig. 2h). Also $H_s$ decreases on increasing $T$ and eventually vanishes at $T \approx T_N$, revealing one additional boundary between different magnetic phases.

The phase boundaries observed in bi- and trilayer CrCl₃ are representative of the behaviour of all even and odd multilayers (see Supplementary Fig. 7). It is therefore useful to understand their origin in terms of the magnetic phases expected in weakly anisotropic layered antiferromagnets[15,42]. In such systems, the orientation of the sublattice magnetization is determined by the interplay of three energy scales: the Zeeman energy, the interlayer exchange coupling, $J$, and the magnetic anisotropy, $K$ (for weak anisotropy $K \ll J$, as is the case in bulk CrCl₃). In the bulk, at sufficiently low $H$, the system is in a collinear antiferromagnetic state with the spins pointing along the easy axis. On increasing $H$ a spin-flip transition takes place in which the sublattice magnetization aligns in the direction orthogonal to the applied field, with a slight canting. Indeed, in the spin-flip configuration the overall energy balance is favourable above $H_s = \sqrt{K/J}$, as the gain in Zeeman energy (proportional to $H^2$) offsets the cost in anisotropy energy (proportional to $K$). A further increase in applied field leads to more canting, and eventually to a spin-flip transition (at $H_{\text{flip}}$) into a state with uniform magnetization aligned with the applied field. In practice, in bulk CrCl₃, the in-plane magnetic anisotropy is negligible (that is, $K \approx 0$) and the spin-flip transition occurs at vanishingly small field ($H_s \approx 0$). The same is true for even-$N$ multilayers, for which the energy gain of the spin-flip phase over the collinear antiferromagnetic state reads $\Delta E_{\text{flip}} = -a_N NHJ/J + NK$ (with $a_N$ a positive, $N$-dependent factor): the first term always dominates since $K \approx 0$ and the spin-flip transition takes place at $H_s \approx 0$. In even-$N$ multilayers, therefore, only one phase boundary corresponding to the spin-flip transition is expected.

The situation is different for odd-$N$ multilayers, where the magnetization of an uncompensated layer gives a finite Zeeman contribution to the total energy that modifies the energy balance as $\Delta E_{\text{flip}} = -a_N NHJ/J + NK + \mu_B M_H$ ($M_H = 2g_a S$ is the saturation magnetization per unit cell of a single layer; $S = 3/2$ is the spin on Cr atoms). At low $H$ the Zeeman term (linear in $H$) dominates over the energetic gain associated to the spin-flip transition (quadratic in $H$), forcing the magnetization in all layers to point parallel to the applied field. The spin-flip transition then occurs at $H = H_s = J M/H_s$ (which is finite despite $K \approx 0$ and can be observed experimentally). Considerations based on energetics, therefore, indicate that even-$N$ multilayers should always exhibit only one observable phase boundary (as $H_s \approx 0$), whereas two phase boundaries can be observed in odd-$N$ multilayers. They also indicate that the field $H_s$ at which the spin-flip transition occurs in odd-$N$ multilayers should decrease on increasing thickness (approximately as $1/N$), since the effect originates from the magnetization of one individual uncompensated layer competing against the energetic contribution of all layers.

**Antiferromagnetic linear-chain model**

These predictions can be made fully quantitative using an antiferromagnetic linear-chain model to describe the state of CrCl₃ mul-
Fig. 2 | Phase diagram of bi- and trilayer CrCl₃. a,e. Magnetic field dependence of dG/dH measured on bi- and trilayer CrCl₃, respectively, at T = 0.3 K. A sharp jump at Hₛ is observed in both samples. In addition, in trilayers a minimum in dG/dH is present at finite field H. b,f. Colour plot of dG/dH as a function of temperature T and applied magnetic field H measured on bi- and trilayer devices, respectively. In both devices, the dG/dH jump at Hₛ shifts to lower fields as T is increased, and finally disappears for T ≃ 17 K, not far from the Néel temperature Tₐ of bulk CrCl₃. In the trilayer device in f, the blue region below µₜH = 0.5 T tracks the evolution with temperature of the minimum in dG/dH at H = Hₛ. c,g. Temperature dependence of tunnelling conductance of bi- and trilayer devices, respectively, measured at different values of applied field H (the field in different curves is increased by 0.2 T per step, starting from µₜH = 0 T). At each value of H, a clear feature in the T dependence allows the critical temperature Tₐ(H) to be determined (taking the derivative dG/dT makes the identification of Tₐ(H) even simpler, see Supplementary Fig. 6). The dashed line indicates the evolution of the transition temperature. d,h. Magnetic field-temperature (H-T) phase diagram of, respectively, bi- and trilayer CrCl₃. The empty black squares correspond to the value of Hₛ extracted from the measurement of G as a function of H at fixed T. The blue dots represent the value of Tₐ(H) extracted from the measurements of G as a function of T at fixed H. As discussed in the main text, these data points trace the phase boundary associated to the spin-flip transition. The triangles in h correspond to the value of Hₛ extracted from dG/dH measurements at fixed T and trace the phase boundary associated to the spin-flip transition (that in odd-N multilayers—such as trilayer CrCl₃—occurs at finite H despite the vanishing in-plane magnetic anisotropy).

tilayers, where the magnetization of each layer is represented by a ‘macro-spin’ on a site, coupled to its nearest neighbours through the interlayer exchange energy J (see Methods). At low temperature, the state of a generic multilayer—that is, the orientation of the magnetization in each layer—can be calculated as a function of applied in-plane field H by minimizing the magnetic energy per unit cell, Uₜ, of the N layer:

\[ Uₜ(\phi₁, \ldots, \phiₙ; H) = J \sum_{i=1}^{N} \cos(\phi_{i+1} - \phi_i) - \muₜH \sum_{i=1}^{N} \cos(\phi_i) \tag{1} \]

with φᵢ the angle between the iᵗʰ layer magnetization and the field applied in-plane.

To interpret our measurements, we start by calculating the linear tunnelling magnetococonductance of bi- and trilayers CrCl₃ and compare it to the experiments. We first determine the evolution of the magnetic state (see the arrows and insets in Fig. 3a,b and Supplementary Note 3). For bilayers (Fig. 3a), the magnetization in the two layers is nearly perpendicular to the field starting from H ≃ 0, progressively canting as H is increased, with the spin-flip transition occurring at H = Hₛ. For trilayers (Fig. 3b), instead, the state does not change for H up to Hₛ as the magnetization in all layers points in the direction of the applied field (either parallel or antiparallel). Only when H is increased past Hₛ do the orientation of the magnetization in each layer start to evolve and eventually aligns with the applied field at H = Hₛ. Such an evolution of the magnetic state discussed here for bi- and trilayers is characteristic of all even-N and odd-N multilayers (that is, the state of even-N multilayers starts changing from H ≃ 0, whereas changing the state of odd-N multilayers requires a finite value of H).

To obtain the tunnelling conductance we assume that each layer acts as a spin filter⁴⁰,⁴¹, transmitting mainly electrons with their spin parallel to the magnetization, so that the transmission through adjacent layers is related to the cosine of the angle between the magnetizations (see Supplementary Note 4). The calculated...
The magnetoconductance $G(H)/G_{\text{sat}}$ is represented by the continuous lines in Fig. 3a,d; the agreement with data is virtually perfect for bilayers and excellent for trilayers, showing that the experimentally observed characteristic fields $H_c$ and $H_f$ can be identified with $H_{\text{cr}}$ and $H_{\text{flop}}$. In trilayers theory predicts the conductance to be constant for $H<H_{\text{cr}}$ and indeed, the measured magnetoconductance curve is rather flat at low field. A small dip is nevertheless present around $H=H_c$ (whom magnitude is sample and bias dependent, see Supplementary Fig. 9) that gives rise to the split peak in $dG/dH$ around $H=H_c$ (blue curve in Fig. 3d). The origin of this conductance dip, present in odd-N multilayers, is currently not clear.

The behaviour of $dG/dH$ discussed for bi- and trilayers is characteristic also for thicker even- and odd-N multilayers. In all even-N multilayers, $dG/dH$ starts increasing immediately on the application of a finite field $H$, and continues to increase up to $H=H_{\text{cr}}$, where the spin-flip transition occurs. This is indeed the behaviour that we observe for $N=8$ (blue line in Fig. 3e), and that was recently reported for $N=4$ in ref. 45. In odd-N multilayers, instead, $dG/dH$ is predicted to vanish as long as $H<H_{\text{cr}}$. In devices where the $H=0$ conductance dip mentioned above is less pronounced, the measurements are consistent with such expectation, as illustrated by data taken on a $N=5$ (Fig. 3f) multilayer. Even when the conductance dip at $H=0$ has a larger magnitude and causes $dG/dH$ to peak at small $H$, in odd-N devices a minimum in $dG/dH$ (with $dH=0$) is still present at finite $H$ much before the spin-flip transition (see Supplementary Fig. 8). No such a minimum is observed in even-N multilayers. We conclude that the behaviour of $dG/dH$ is distinctly different for even-N and odd-N multilayers, that is, the experiments indicate the presence of an even-odd effect in the magnetic response of atomically thin CrCl$_3$ crystals. In odd-N multilayers, we take the value of $H_c$ to estimate the value of $H_{\text{cr}}$, the field at which the spin-flip transition occurs (see also Supplementary Fig. 8).

**Interlayer coupling and layer magnetization**

It is also revealing to compare the evolution of $dG/dH$ with magnetic field applied parallel and perpendicular to the CrCl$_3$ layers. The orange (blue) curves in Fig. 3e,f represent data taken with $H$ perpendicular (parallel) to the layers (for data taken on multilayers of different thickness see Supplementary Fig. 10). In all multilayers the spin-flip transition occurs at a higher value of $H$ when the field is applied perpendicular to the layers. The value of $H_c$, for all measured multilayers is shown in Fig. 4a for field applied in (blue circles, $H_2$) and perpendicular to (orange circles, $H_1$) the plane; the difference $\Delta H_2 = H_2 - H_1$ is shown in Fig. 4b. Remarkably, $\Delta H_2$ has nearly exactly the same value for all multilayers, and corresponds quantitatively to the saturation magnetization of an individual layer.
To understand the implications of this observation consider that the spins in the material experience an effective 'internal' field, \( H_{\text{eff}} \), which is the difference between the applied and the demagnetization fields; that is, \( H_{\text{eff}} = H - \gamma M_s \). \( (M_s = M/V \) is the magnetization per unit cell volume \( V \) and \( \gamma \) is the demagnetization factor). In the thin slab geometry of our devices, \( \gamma = 0 \) for an in-plane field and \( \gamma = 1 \) for a perpendicular field. Finding that \( \Delta H_s \approx M_s \) thus means that the spin-flip transition occurs at the same effective internal field, irrespective of the direction of the applied field \( H \). Besides allowing the saturation magnetization of individual \( \text{CrCl}_3 \) layers to be measured, this finding indicates the absence of significant magnetic anisotropy due to spin–orbit interaction. Indeed, in the presence of magnetocrystalline anisotropy, the value of effective internal field at which the spin-flip transition occurs should depend on direction (as happens, for instance, in \( \text{CrI}_3 \), refs. 18–21). We therefore conclude that the magnetic anisotropy in atomically thin \( \text{CrCl}_3 \) multilayers is dominated by shape anisotropy.

The thickness dependence of the measured transition fields \( H_1 \) and \( H_2 \) agrees quantitatively with predictions of the antiferromagnetic linear-chain model (see Supplementary Note 5). For the spin-flip transition in parallel field, the measured values of \( H_1 \) for different \( N \) are represented by the blue circles in Fig. 4a. The model predicts the transition to occur at \( H_1(N) = 2H_s \cos^2(\pi/N) \) for \( H_s(=2f_0M_s) \) is a field scale associated with the interlayer exchange coupling \( f_0 \). \( H_2 \) doubles when going from the bilayer to the bulk, a behaviour resulting from the reduced antiferromagnetic coupling of the outermost layers that miss one neighbour, which suppresses the stiffness of the system to reorient in the direction of the field. This prediction excellently reproduces the experimental points when setting \( \mu_0H_s = 0.99 \text{T} \) (blue continuous line in Fig. 4a). For a perpendicular field, the spin-flip transition occurs at \( H_2(N) = H_1(N) + M_s/V \) due to the shape anisotropy, an expression that reproduces extremely well the data points with the same value of \( H_1 \) without additional adjustable parameters (orange circles and continuous line in Fig. 4a).

The same value of \( H_1 \) also accounts quantitatively for the thickness evolution of the spin-flop field \( H_1 \). As shown in Fig. 4c, in the even-\( N \) multilayers investigated here (\( N = 2 \) and 8) and in ref. 45 (\( N = 4 \)), \( H_1 \) vanishes, due to the negligible in-plane anisotropy (see orange circles in Fig. 4c). For odd-\( N \) multilayers the measured \( H_1 \) values are finite and decrease with increasing thickness (blue circles in Fig. 4c). The antiferromagnetic linear-chain model predicts \( H_1(N) = H_1 \sin(2\pi/N) \) (blue continuous line in Fig. 4c), in remarkably good agreement with the data using the same value of \( H_1 \) extracted from the analysis of \( H_2 \). The model, therefore, reproduces the thickness evolution of all measured transition fields \( H_1 \), \( H_2 \) and \( H_3 \) using as single free parameter the interlayer exchange coupling, \( f_0 = 86 \mu \text{eV} \).

**Temperature evolution**

Finally, we extend our considerations to finite temperature. The \( T \) dependence of the layer magnetization is obtained through the relationship \( \Delta H(T) = M(T)/V \). Figure 5a shows representative results (from a five-layer device) for the temperature evolution of the measured spin-flip fields \( H_1(T) \) and \( H_2(T) \), and for their difference. All quantities monotonically decrease from their saturation values at low \( T \) and vanish at the same critical temperature \( T_N \approx 18 \text{K} \). The difference \( \Delta H_2 \) can be converted into a layer magnetization \( M \) as shown on the right axis. It can be proved theoretically (at the level of mean-field theory) that this quantity also corresponds to the sublattice magnetization of a single layer in the antiferromagnetic state at \( T = 0 \), even if \( M(T) \) is obtained in the ferromagnetic state at the spin-flip field \( H_2 \). Transport measurements, therefore, provide access to the \( T \)-dependent sublattice magnetization, a non-trivial conclusion because it is not obvious how this quantity could be measured otherwise in an atomically thin antiferromagnet.

To show that these considerations are internally consistent and quantitatively in accord with the antiferromagnetic linear-chain model used for our analysis, we look at the relation between \( M(T) \) and the spin-flip field \( H_1(T) \). The model predicts \( H_1(N, T)/H_1(T) = c_0 M(N, T)/M_{\text{sat}} \) where \( c_0 = [1 + \cos(\pi/N)] \). All quantities are known: they are either obtained directly from measurements, or can be calculated using the value of \( f_0 \) determined from the thickness dependence of \( H_1 \). To verify whether the experimental data obey this relation, in Fig. 5b we plot \( H_1(N, T)/H_1(T) \) as a function of \( c_0 M(N, T)/M_{\text{sat}} \) for several temperatures \( T \) and values of \( N \), for all devices on which we have taken systematic temperature-dependent data in parallel and perpendicular field. Indeed, all experimental points cluster around the blue, slope 1 line; that is, as predicted by theory the two quantities coincide. The model also predicts a proportionality between the spin-flop and spin-flip fields that persists to finite temperature, \( H_1(T)/H_2(T) = \tan(\pi/N)/2 \cos(\pi/N ) \), a relation well satisfied by the experimental data (Fig. 5c).

**Conclusions**

The antiferromagnetic linear-chain model therefore provides an excellent quantitative understanding of the evolution of the magnetic states of \( \text{CrCl}_3 \) multilayers, as a function of magnetic field, number of layers and temperature. This finding is unexpected,
since all theoretical predictions invoked in our analysis heavily rely on a mean-field-like approach, whose validity for 2D magnetic systems without a pronounced easy-axis anisotropy—such as CrCl$_3$ multilayers—is not straightforward. What is possibly more striking, however, is the ability to trace the complete phase diagram of a magnetic system—at a level that enables the identification of subtle phases originating from finite-size effects—from magnetococonductance experiments. Such a method offers important advantages over other techniques. Besides its simplicity, one advantage is the possibility to probe extremely small material volumes removing (or drastically limiting) the influence of inhomogeneities and domains, which plague the results of experimental techniques that require much larger sample sizes. Finding suitable techniques to probe the magnetic response of atomically thin magnets clearly represents a urgent need in the field of magnetic 2D materials. The possibility to employ tunnelling magnetoresistance to extract a multitude of information—demonstrated here for CrCl$_3$—may be applicable more broadly, which would disclose extremely promising prospects to sustain progress in this area.

Note that while finalizing the manuscript we became aware of additional investigations on the tunnelling magnetococonductance of thin CrCl$_3$ multilayers$^{[46,31,32]}$, which nonetheless do not explore the thickness and temperature evolution of spin-flip and spin-flop transitions, or analyse the data through a systematic quantitative comparison to a theoretical model.

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Methods

Crystal growth. CrCl₃ multilayers were exfoliated from bulk crystals that we grew by means of a chemical vapour transport method, using commercially available polycrystalline CrCl₃ powder (99.9%, Alpha Aesar) as a starting material. To this end, the CrCl₃ powder was inserted in a quartz tube (inner diameter = 8 mm, tube length = 13 cm) inside a glove box filled with 99.9999% Ar. The tube was subsequently evacuated down to $p \approx 10^{-4}$ mbar, sealed and inserted in a horizontal tubular furnace enabling a controlled temperature gradient to be established. The temperatures at the hot and cold ends of the tube were set to be 670 and 550°C, respectively. The tube was left in the furnace under these conditions for 7 days, after which the furnace was switched off, letting the tube cool down to room temperature. At the end of this process, violet, fairly transparent CrCl₃ platelets were found at the cold end of the tube (at the hot end, a small amount of greenish powder remains, probably Cr₂O₃ originating from oxygen present in the commercial starting material). The CrCl₃ thin crystalline platelets had a lateral size of up to 4 mm (limited by the diameter of the tube) and exhibit sharp edges, forming 60° and 120° angles, as expected from their crystal structures (Supplementary Fig. 1). The structure, stoichiometry and the magnetic response of the crystals were characterized as discussed in the Supplementary Notes 1 and 2; the results of this characterization were fully compatible with the properties of CrCl₃ known from the literature.

Sample fabrication. CrCl₃ multilayers were mechanically exfoliated from the platelet crystals discussed here above. Tunnel junctions consisting of multilayer graphene/CrCl₃/multilayer graphene were assembled using a by-now common pick-and-lift technique, in a cross geometry illustrated in the inset of Fig. 1b. To avoid degradation of thin CrCl₃ multilayers, the exfoliation of CrCl₃, and the heterostructure stacking process were done in a glove box filled with nitrogen gas, and the whole tunnelling junction was encapsulated with hBN before being taken out. Conventional electron-beam lithography, reactive-ion etching, electron-beam evaporation (10 nm/50 nm Cr/Ar) and a lift-off process were used to contact the multilayer graphene electrodes, away from the CrCl₃ tunnel junction. The thickness of the layers was determined by atomic force microscope measurements performed outside the glove box, on the encapsulated devices, which was possible because of the atomic flatness of all the layers used (including the hBN layer under the CrCl₃ flake, see Supplementary Figs. 3 and 4).

Transport measurements. Transport measurements were performed using two different cryostats from Oxford Instruments, a Heliox 4He insert with base temperature $T$ < 0.25 K and a cryofree Teslatron system with base temperature $T$ ≈ 1.5 K, using home-made low-noise electronics. For measurements performed in the Heliox system, samples were taken out of the insert and manually rotated, to change the angle between magnetic field and CrCl₃. The Teslatron cryostat, instead, is equipped with a sample rotator enabling the devices to be rotated at low temperature.

Antiferromagnetic linear-chain model. We introduce here the antiferromagnetic linear-chain model that we use to describe CrCl₃ multilayers. Several variants of this model have appeared in multiple contexts and with different names in the literature (see for example ref. 1 and references therein). The main underlying assumption is that the ferromagnetic intralayer exchange coupling is so strong (compared to the weak antiferromagnetic interlayer exchange coupling and the external magnetic field) that each layer behaves as a single unit with uniform magnetization. The temperature dependence of this layer magnetization $M(T)$ is to a large extent determined solely by the balance between ferromagnetic interactions and thermal fluctuations within a single layer. Deviations from this behaviour can be expected very close to $T_m$, when contributions from interlayer exchange interactions become more important. Still, we will not be interested in this regime as it is not the one where most of the experimental investigations have been focused.

Away from $T \to T_m$ we are then justified to adopt an approach where interlayer interactions are treated at a mean-field level, so that each layer can be considered as a macroscopic spin that is coupled antiferromagnetically to its neighbours in the adjacent layers. We can thus model any multilayer as a linear chain of macroscopic spins, with an average magnetic energy per unit cell at temperature $T$ given by:

$$U_N(T) = \sum_{i=1}^{N} M_i(T) \cdot M_{i+1}(T) - \sum_{i=1}^{N} K_i \cdot M_i(T)^2 - \mu_B \cdot H \cdot \sum_{i=1}^{N} M_i(T)$$

Here $i > 0$ is the antiferromagnetic interlayer exchange coupling (taken for simplicity to be uniform throughout the multilayer), $M_i(T)$ is the magnetization per unit cell of the $i$th layer at temperature $T$, $M_i$ is the saturation magnetization per unit cell of a single layer, $K_i > 0$ is the easy-plane anisotropy energy (assuming each layer to lie in the x-y plane) and $H$ is the external magnetic field. In general, the anisotropy energy has two main contributions, $K_T = K_{ax} + K_{ay}$, corresponding to the magnetocrystalline anisotropy ($K_{ax}$ stemming from spin–orbit coupling in the material) and the shape anisotropy ($K_{ay}$ associated with magnetostatic interactions). As we show in the main text, $K_{ax}$ is negligible and the magnetic anisotropy is dominated by $K_{ay} = \mu_B M_i^2 / V$ (being the unit cell volume of CrCl₃). Since $M_i = 2g \mu_B S$ can be easily computed from the nominal valence state of Cr atoms in CrCl₃ (corresponding to $S=3/2$) and $V$ is available from previous experiments, this means that the interlayer exchange coupling $J$ is the only free parameter in the model. Irrespective of the apparent mean-field nature of the description used to interpret the data, it is important to bear in mind that the model does not describe the strong intralayer interactions responsible for the ferromagnetic ordering of individual layers. Indeed, $M(T)$ is taken as an external parameter in the model that would need a formalism beyond the mean field to be computed theoretically.

In the zero-temperature limit, the magnetization in the $i$th layer can be expressed as $M_i = \bar{M} \cdot \mathbf{m}_i$, where $\mathbf{m}_i$ is a unit vector and the magnetic energy reads:

$$U_N = \frac{1}{2} \sum_{i=1}^{N} \mathbf{m}_i \cdot \mathbf{m}_{i+1} + \frac{K}{2} \sum_{i=1}^{N} \left( \mathbf{m}_i \cdot \mathbf{m}_i - 1 \right)^2$$

which reduces to equation (1) in the main text when the field is applied in-plane $H = (H,0,0)$ and we can write $\mathbf{m}_i = (\cos \phi_i, \sin \phi_i, 0)$. The most stable magnetic configuration $(\mathbf{m}_i)_{i=1}^{N}$ of any multilayer can be obtained by minimizing the energy $U_N$ for a given value (and orientation) of the applied magnetic field $H$. In Supplementary Note 3 we show in practice how to perform the minimization of $U_N$ to obtain the magnetic configuration of bi- and trilayers as a function of applied magnetic field.

Data availability

All relevant data are available from the corresponding authors upon reasonable and well-motivated request.

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Author contributions

Z.W., M.G. and A.F.M. conceived the work. D.D. and E.G. grew CrCl₃ crystals and performed transport characterization. T.T. and K.W. provided high-quality boron nitride crystals. Z.W. fabricated all samples and performed all transport measurements. M.G. carried out all theoretical modelling. Z.W., M.G. and A.F.M. analysed and interpreted the magnetocconductance data. All authors contributed to writing the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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