Structure of antiferromagnetic domain walls in single-crystal Cr$_2$O$_3$

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We resolve the domain-wall structure of the model antiferromagnet chromium(III) oxide using nanoscale scanning diamond magnetometry and second-harmonic-generation microscopy. We find that the 180° domain walls are predominantly Bloch-like, and can co-exist with Néel walls in crystals with significant in-plane anisotropy. In the latter case, Néel walls that run perpendicular to a magnetic easy axis acquire a well-defined chirality. We further report quantitative measurement of the domain-wall width and surface magnetization. Our results provide fundamental input and an experimental methodology for the understanding of domain walls in pure, intrinsic antiferromagnets, forming a basis for electrical control of domain-wall motion in antiferromagnetic spintronics.

One of the great unknowns of antiferromagnetism is the domain wall that separates regions with different orientation of the magnetic order parameter. The domain-wall structure influences the thermal stability [1], exchange bias [2], and magnetoresistance [3, 4] of antiferromagnets. Furthermore, the type of domain wall, Bloch or Néel, determines their response to current-induced spin torques [5–8], which is of relevance for emerging applications of antiferromagnets in spintronics [9, 10].

Unlike for ferromagnets [11], the internal structure of domain walls in antiferromagnets is not generally known. Exceptions include a few systems where antiferromagnetic order is accompanied by strain [12] or defects [13] as well as monolayer-thick films [14], but no studies for bulk, intrinsic antiferromagnets have been reported. Theoretical analysis suggests that, in the absence of in-plane magnetic anisotropy, no preference is expressed for either Bloch or Néel walls [15, 16], but no experiments have thus far been reported. The limited experimental knowledge about antiferromagnetic domain walls is due to a lack of techniques capable of spatially resolving the internal wall structure.

In this work, we use nanoscale scanning diamond magnetometry (NSDM) to investigate the spin structure of the pure intrinsic antiferromagnet Cr$_2$O$_3$. NSDM microscopy is an emerging quantum technique for the imaging of weak magnetic fields with nanometer spatial resolution (Fig. 1), with remarkable progress on antiferromagnets [4, 17, 18], multiferroics [19], and helimagnets [20]. Here, we extend NSDM to the imaging of antiferromagnetic 180° domain-wall structures. We obtain quantitative information about the domain-wall width, chirality and surface magnetization, and connect it to a model of interplaying demagnetizing and anisotropy energies. We find that both Bloch and Néel walls can be present. To the best of our knowledge, our work reports the first experimental observation of domain-wall structure in a pure, intrinsic antiferromagnet.

Cr$_2$O$_3$ is an antiferromagnetic insulator consisting of a hexagonal close packed array of oxygen anions with 2/3 of the octahedral holes occupied by chromium [21] (Fig. 1a).

Below $T_N = 307.6$ K, Cr$_2$O$_3$ forms an antiferromagnetically ordered phase, where the Cr$^{3+}$ ions organize in alternating layers of opposite magnetic polarization (green and blue spins in Fig. 1a). Because of its fundamental role in antiferromagnetism, Cr$_2$O$_3$ has served as a model system for uniaxial antiferromagnetic order [22–24], magnetoelastic coupling [25–27], and electrically controlled exchange bias [28, 29]. More recently, Cr$_2$O$_3$ has attracted attention as a candidate material for antiferromagnetic magnetoelastic random access memories [17], spin colossal magnetoresistance [30], and as generator of sub-THz spin currents [31]. Although the domain wall plays a critical role in many of these phenomena, the spin structure is unknown beyond initial theoretical work [32], presenting an important experimental test case.

We study the Cr$_2$O$_3$ domain texture of three bulk single crystals. Samples A and B are grown by the Verneuil method and polished to a surface roughness of 1–3 nm-rms. Sample C is a flux-grown platelet with an as-grown surface. In a previous study [34], we found that the spin flop transition – normally requiring a magnetic field of 5.8 T (Ref. 21) – occurs spontaneously at 150 K in sample C, pointing to an unusually strong in-plane anisotropy. In addition, this sample has a lower Néel temperature ($T_N = 304.6$ K), probably due to strain or oxygen deficiency. We create antiferromagnetic domains by repeatedly cooling samples through the transition temperature $T_N$ using magnetoelastic poling [35] or until a multi-domain state spontaneously forms. Further details about the samples are given in Ref. 33.

Domain states: Figure 2(a) shows a laser-optical second-harmonic-generation (SHG) [34] micrograph of the global domain pattern. We observe that the domains in the bulk Cr$_2$O$_3$ crystals are large, typically in the range of hundreds of micrometers, and stable below $T_N$, in agreement with earlier studies [36]. We find no correlation between the domain pattern and the in-plane crystal axes (Fig. 2a), indicating that the domain-wall location is set by the local defect or strain distribution or is completely random.

Once the domains are localized, we acquire high-
resolution magnetic imaging scans along the domain walls using NSDM microscopy (Fig. 2b). The domain wall appears as a narrow track of strong magnetic stray field in the magnetometry image; this strong field is due to the 180° reversal of uncompensated moments near the sample surface (see Fig. 1b). Fainter features within the domains reflect residual stray fields associated with surface topography [37]. To retrieve the absolute sign of the order parameter – which is not possible by optical measurements [34] – we reconstruct [33] the two-dimensional surface magnetization $\sigma_z$ from the stray field map of Fig. 2b, shown in panel c. Here, a positive sign of $\sigma_z$ (dark contrast) reflects a positive Cr$_{3+}$ surface magnetization and order parameter $L^-$ (vice versa for $L^+$). We find that the correlation between SHG contrast and surface magnetization is maintained for all domain walls on all samples (Figs. S1 and S2 in Ref. 33). Combined with the absence of strong magnetic features in the interior of domains, these findings directly confirm that the magnetic polarization of Cr$_2$O$_3$ is robust against surface roughness [18, 29], and that Cr$_2$O$_3$ always terminates with the same Cr$_{3+}$ surface magnetization for a given sign of the order parameter $L$.

**Domain-wall structure:** To investigate the internal structure of a domain wall, we acquire a large number of magnetometry images along the domain wall and analyze the magnitude and spatial profile of the stray field [38, 39]. We then compare the magnetic field along the cross section with the expected stray field from the static solution of the one-dimensional domain-wall model [15, 40, 41], with a magnetization profile given by:

$$\sigma_x(x) = \sigma_x^0 \left[ \cosh \left( \frac{x - x_0}{\Delta} \right) \right]^{-1} \cos \chi , \quad (1a)$$

$$\sigma_y(x) = \sigma_y^0 \left[ \cosh \left( \frac{x - x_0}{\Delta} \right) \right]^{-1} \sin \chi , \quad (1b)$$

$$\sigma_z(x) = \sigma_z^0 \tanh \left( \frac{x - x_0}{\Delta} \right) , \quad (1c)$$

here for a domain wall extending along the $y$ direction and centered at $x = x_0$. By computing the magnetic stray field from Eq. (1) using forward propagation [33] and fitting it to the experimental cross section, we obtain quantitative estimates for the surface moment density $\sigma_z^0$, domain-wall width $\Delta$, and twist angle $\chi$. This angle defines the orientation of the in-plane magnetization inside the wall with respect to the $x$-axis, which is $\chi = \pm \pi/2$ for a Bloch domain wall and $\chi = 0 (\pi)$ for a Néel wall with right (left) chirality. Figure 3(a,b) shows an exemplary line scan across a domain wall of sample C together with the least-squares fit. To build sufficient statistics and avoid possible cross-correlation between fit parameters,
we have analyzed of order $10^3$ line scans for each sample and validated results by a secondary data analysis (Figs. S3 and S4 in Ref. 33).

Figure 3c reports quantitative measurements of the layer magnetization $\sigma_0^2$. We find a narrow distribution of $\sigma_0^2$ values between $1.6(2)\mu_B/\text{nm}^2$ (sample C) and $2.3(2)\mu_B/\text{nm}^2$ (sample A), corresponding to between 45 – 65% of the expected layer magnetization of a perfectly ordered Cr$^{3+}$ crystal ($\sigma_0^2 = 3.64\mu_B/\text{nm}^2$, Ref. 33). The reduction is consistent with a Curie-Weiss behavior close to $T_N$ (Ref. 18), and is more pronounced for sample C due to the lower $T_N$. The narrow value distribution indicates that the Cr$^{3+}$ magnetization is well-defined and uniform across the sample surface.

Figure 4a-c plots the fit results for the domain-wall width $\Delta$ and angle $\chi$ obtained from the extensive datasets recorded on samples A-C. Each plotted $(\chi, \Delta)$ pair represents a $\sim 4\times4\mu\text{m}^2$ magnetometry scan, and color-coding reflects the propagation direction of the domain wall. For samples A and B we find all domain walls to be predominantly Bloch-like, indicated by a $\chi$ angle close to 90° (Fig. 4a,b). The domain-wall widths are not identical, but of similar magnitude $\Delta \sim 40\text{nm}$, and well in the range of $20 - 80\text{nm}$ predicted by theory [32]. Clearly, there is no correlation between $(\chi, \Delta)$ and the spatial location or propagation direction $\alpha$ of the domain wall (see panels d,e), indicating that the crystal structure plays no role in domain-wall formation. The consistency of the results from the two samples, which are grown independently by the same technique, confirms that our methods for quantifying the domain-wall structure are robust and reproducible.

Interestingly, sample C – which has an unusually strong in-plane anisotropy [34] – shows a behavior that is distinctly different from samples A and B. Most prominently, we find both Néel and Bloch walls and a pronounced dependence of the twist angle on the wall orientation. For walls that run approximately parallel to one of magnetic hard axes (dashed lines in Fig. 2a and Fig. 4d-f), the domain wall has a distinct left Néel character (blue data points in Fig. 4c). Once the angle $\alpha$ between the propagation direction and the hard axis exceeds about 9°, the wall changes to Bloch-type, and becomes similar to samples A and B. In addition, the domain-wall width increases from $\Delta = 42\text{nm}$ in the Néel to $\Delta = 65\text{nm}$ in the Bloch configuration. The correlation between $(\chi, \Delta)$ and $\alpha$ is not complete, but pervasive, suggesting that a delicate balance of interactions determines the local structure of the wall.

Discussion: The formation of distinct Bloch and Néel walls in Cr$_2$O$_3$ is intriguing, because in the absence of a demagnetizing field and in-plane anisotropy, the domain-wall energy of a collinear antiferromagnet is independent of the angle $\chi$ [15, 33, 40, 41]. Therefore, no domain-wall type is energetically favored. In Cr$_2$O$_3$, however, domain walls have a non-vanishing local magnetic moment associated with the spatially inhomogeneous order parameter [40, 42], giving rise to a small but non-zero demagnetizing field. We propose that this residual demagnetizing field, which is mostly a bulk effect, is responsible for the observation of Bloch walls in samples A and B, similar to the situation encountered in uniaxial ferromagnets [11].

The preference for Bloch walls is challenged once significant in-plane anisotropy is present (sample C). An in-plane anisotropy favors Cr$^{3+}$ spins aligned with the in-plane easy axis and for a sufficiently strong anisotropy, the domain wall is expected to change from Bloch to Néel. Due to three-fold crystal symmetry of Cr$_2$O$_3$, three in-
plane easy axes exist (that coincide with the crystal axes a, a’ and b, see Fig. 2a) leading to six preferred directions in 60° intervals. Therefore, Cr³⁺ spins will tend to align to the nearest preferred easy direction. The alignment is strongest when the domain wall is perpendicular to an easy axis, explaining the appearance of Néel walls near α ≈ 0° (blue data points in Fig. 4c). Once α becomes larger, the in-plane anisotropy torque is reduced, and the domain wall eventually changes back to a Bloch type (yellow data points). The critical angle where this change occurs is not well defined, but is roughly α ≈ 9°. At the same time as the domain-wall type changes from Néel to Bloch, the domain-wall width is expected to increase, in line with our observation (Fig. 4c). A simple theoretical argument [33] predicts a ratio of domain-wall widths of \( r = \Delta_{\text{Néel}} / \Delta_{\text{Bloch}} = 0.85 \), in reasonable agreement with the experimental result of \( r = 0.65 \pm 0.10 \) (Fig. 4c). The good overall agreement between experiment and theory motivates the conclusion that the non-vanishing magnetic moment and in-plane anisotropy determine the domain-wall structure of Cr₂O₃.

A final point that remains to be explained is the preference for left chiral Néel walls in sample C, which is also partially present in samples A and B (Fig. 4a-c). Although the asymmetry is conspicuous, it is not entirely surprising given the complex magnetoelectric properties of Cr₂O₃ [35]. Because the orientation of the spins in a left chiral Néel wall is against the stray field produced by the uncompensated magnetization of the top-most surface layers of Cr₂O₃ (Fig. 1b), the preference for left walls cannot be attributed to a magnetostatic effect, unlike the change of a Bloch wall into a Néel wall observed in the near-surface region of ferromagnets [11]. Future theoretical work shall determine whether internal effects or possibly a wall-related Dzyaloshinskii-Moriya interaction (DMI) is responsible for the domain-wall chirality. In a non-centrosymmetric environment, the DMI results in canting of the spins when \( L \) has a non-zero in-plane component [22, 43], which – unlike in bulk Cr₂O₃ – may be the case within the Cr₂O₃ domain wall.

In summary, we have resolved the spin structure of 180° domain walls in the prototype uniaxial antiferromagnet Cr₂O₃. We propose that the structure of the domain wall is determined the weak energy scales
provided by the non-vanishing magnetization of the wall, the in-plane magnetic anisotropy, and possibly the Dzyaloshinskii-Moriya interaction. Domain walls are Bloch-like in crystals with weak or negligible in-plane magnetic anisotropy, and either Bloch- or Néel-like in the crystal with larger in-plane anisotropy. In the latter case, the domain-wall type turns to Néel if the wall runs orthogonal to an in-plane easy axis, which coincides with the spin direction in the spin-flop phase of Cr$_2$O$_3$ [21].

Looking forward, our work provides important first insight into the domain walls of pure, intrinsic antiferromagnets. In particular, the possibility to induce chiral domain walls is critical for controlled domain-wall motion in emerging spintronic devices. In such systems, the structure of domain walls is often assumed a priori, without experimental verification. Because of the model character of Cr$_2$O$_3$ as a pure antiferromagnetic insulator, we expect that the methods and insight generated in our study can be readily extended to other antiferromagnets, including bulk crystals and thin films. Future studies may also include correlative imaging of antiferromagnetic and ferroelectric domain walls, so as to investigate the coupling between different types of order in multiferroic materials.

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Figure 4. Observation of Bloch and Néel walls. (a-c) Twist angle $\chi$ plotted against the domain-wall width $\Delta$ for samples A-C. Each point represents the data from a two-dimensional magnetometry scan. Error bars (±1 s.d.) are obtained by separate fits to each line of the 2D scan and computing the standard deviation (s.d.) of the fit results. Color coding reflects the propagation direction of the domain wall (see right panels). No correlation between chirality and spatial position is evident for samples A and B, whereas a clear correlation is evident for sample C. Mean angle and domain-wall widths are $(\chi, \Delta) = (106(6)^\circ, 34(5) \text{ nm})$ for sample A, $(113(11)^\circ, 45(8) \text{ nm})$ for sample B, $(143(12)^\circ, 42(6) \text{ nm})$ for sample C with $\alpha < 9^\circ$, and $(117(7)^\circ, 65(4) \text{ nm})$ for sample C with $\alpha > 9^\circ$; brackets denote standard error. (d-f) SHG images of the domain-wall regions analyzed in panels a-c. Colored squares show the scan locations. $\alpha$ is the angle between the local propagation direction of the domain wall (red solid line) and one of the magnetic hard axes (red dashed line, see Fig. 2a). Scale bars, 25 $\mu$m.
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Supplemental Material for:

“Structure of antiferromagnetic domain walls in single-crystal Cr$_2$O$_3$”

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1. MATERIALS AND METHODS

1.1. Sample preparation

We study Cr$_2$O$_3$ domain walls on three bulk single crystals. Samples A and B (Refs. 1–3) are grown by the Verneuil method and oriented using a single-crystal X-ray diffractometer. The samples are cut perpendicular to the $z$-axis or (001) orientation. Subsequently, the samples are thinned down to 70 $\mu$m. Both samples are lapped and polished, each of them following a different process. Sample A is lapped using SiC powder with 3 $\mu$m grain size on a cast-iron-lapping plate. Subsequently, the sample is polished following a two-step process. In the first step, the lapped surface is polished with a soft metal plate using diamond powder with 1 $\mu$m grain size. In the second step, a refining polishing step follows using a polyurethane polishing plate together with colloidal silicate. Here, scratches from previous mechanical treatments are removed. The sample surface is polished until it reveals a root-mean-square (rms) roughness below 1 nm. Sample B is lapped using Al$_2$O$_3$ powder and H$_2$O solution. Next, the lapped surface is diamond polished until it reveals a surface with a rms roughness below 3 nm. Sample C (Ref. 3) is a flux-grown (001) Cr$_2$O$_3$ platelet of 30 $\mu$m thickness. The flat as-grown surface presents a rms roughness below 0.5 nm. SHG images of all crystals are shown in Figs. S1 and S2. We create antiferromagnetic domains by cooling samples through the transition temperature $T_N$. For samples A and B, domains are induced by magnetoelectric poling [4]. In sample C, different domain patterns spontaneously form when the sample is cooled through $T_N$.

1.2. Second harmonic generation (SHG) measurements

SHG microscopy exploits an interference contrast of frequency-doubled optical photons in domains of opposite magnetic polarization to reveal the domain pattern [1]. A magnetic contribution to the frequency-doubled light wave coupling linearly to the antiferromagnetic order parameter $\pm L$ interferes with a frequency-doubled crystallographic background contribution which identifies the two antiferromagnetic domain states by their different brightness [5]. We use a transmission SHG setup to acquire the SHG images, in which we use a Coherent Elite Duo laser system, which emits 120 fs pulses at a repetition rate of 1 kHz. An optical parametric amplifier tunes the wavelength to excite the bulk Cr$_2$O$_3$ samples with a photon energy of 1.033 eV and a pulsed energy of 80 $\mu$J. The crystals are excited in transmission and at normal incidence by an unfocused circularly-polarized laser beam. Right-handed circularly-polarized light denote the clockwise rotation of the electric-field vector of light with respect to its propagation direction. The opposite follows for left-handed circular polarization. A camera lens is used to collect the SHG signal. Optical filters are added to select the SHG spectral wavelength, suppressing the fundamental beam and higher-harmonic contributions. SHG light is detected at room temperature with a Jobin-Yvon, back-illuminated, deep-depletion digital camera with a near-infrared detector chip of 1024×256 pixels. The
camera is cooled with liquid nitrogen to reduce thermal noise.

1.3. Nanoscale scanning diamond magnetometry (NSDM) measurements

Scanning NV magnetometry measurements are carried out on a user-facility instrument, built in-house [6], and under ambient conditions. The instrument uses 520 nm laser light and 2.76 GHz to 2.91 GHz microwave pulses to detect the NV center spin resonances. Laser illumination is kept below 90 µW to avoid laser-induced heating of the sample. The spin resonance frequency is determined by sweeping the microwave frequency and fitting a Lorentzian function to the optically-detected magnetic resonance spectrum. Four different diamond probes (QZabre LLC) of ∼22% CW ODMR contrast at a measurement count rate of ∼200 kC/s are used. The sensitivity of these probes (as determined from the average least-squares variance of the center frequency) is 1.7 µT for an integration time of 6.4 seconds per pixel. All scans are performed on the Cr₂O₃ surface pointing towards the camera in the SHG experiment.

We use both continuous and pulsed ODMR protocols [7, 8] on either transition (m_s = 0 to m_s = ±1) of the NV center. A small external bias field of ∼4 mT is applied to split the spin resonances; this small bias field is not expected to influence the Cr₂O₃ physics. To convert the measured spin resonance frequency f to units of magnetic field, we compute

\[ B_{NV} = \frac{f_0 - f}{28.02 \text{ MHz/mT}} \]

where \( f_0 \) is the mean frequency taken over the entire scan, which is approximately the frequency far from the sample surface. We recall that NV magnetometry provides one vector component of the magnetic field, \( B_{NV} = \vec{e} \cdot \vec{B} \), which is the projection of \( \vec{B} \) onto the anisotropy axis \( \vec{e} = (e_x, e_y, e_z) \) of the spin. The unit vector \( \vec{e} = (\sin \theta_{NV} \cos \phi_{NV}, \sin \theta_{NV} \sin \phi_{NV}, \cos \theta_{NV}) \) corresponds to the symmetry axis (N-V axis) of the NV center, as expressed by the laboratory frame angles \( \theta_{NV} \) and \( \phi_{NV} \). The sensor vector orientation is pre-determined for each tip using an external field sweep. The stand-off distance \( z \) between NV center and the sample surface is measured by independent calibration scans over a magnetized Co stripe before and after the Cr₂O₃ scans [9, 10]. For our probes, \( (\theta_{NV}, \phi_{NV}, z) \) is (55°, 270°, 73 ± 7 nm) for tip A, (55°, 180°, 64 ± 4 nm) for tip B, (55°, 176°, 65 ± 3 nm) for tip C and (55°, 176°, 68 ± 8 nm) for tip D.

2. DATA ANALYSIS

2.1. Definition of surface magnetization \( \sigma_z^0 \)

Antiferromagnetic order in the form of vertically alternating layers of oppositely polarized ions leads to an effective surface layer magnetization on the top and bottom surfaces of
the crystal, in analogy to the bound surface charge appearing for a polarized dielectric [11–
13]. To calculate the surface magnetization, we assign the alternating layers of opposite
polarization to two oppositely magnetized volumes, each with magnetization \( M_s = nm/V \),
vertically shifted with respect to each other by \( s \). Here, \( n \) is the number of ions per unit cell
and polarization direction, \( m \) is the magnetic moment per ion, and \( V \) is the volume of the
unit cell. Within the bulk, the magnetization of the two volumes is exactly compensated,
except in two thin layers of thickness \( s \) at the top and bottom of the body. Thus, the bulk
antiferromagnetic order appears like an magnetized surface layer at the top and bottom of
the crystal, with an effective layer magnetization of
\[
\sigma^0_z = dM_s = \frac{nms}{V} .
\]  

For thick crystals a local magnetic probe only detects the stray field of the top layer.

\( \text{Cr}_2\text{O}_3 \) has a hexagonal unit cell with a side length of \( a = 4.961 \text{ Å} \), a height of \( c = 13.6 \text{ Å} \), a
hexagonal surface area of \( A = \frac{3\sqrt{3}}{2}a^2 = 63.9 \text{ Å}^2 \), and a volume of \( V = Ac = 869.6 \text{ Å}^3 \) (Refs.
3, 13). The hexagonal unit cell is constructed from six vertically stacked \( \text{O}^{2−} \) planes. Each
\( \text{O}^{2−} \) plane has two nearest \( \text{Cr}^{3+} \) ions of opposite magnetic polarization located 0.941 Å above
or below the plane, respectively, therefore \( s = 1.882 \text{ Å} \) (see Fig. 1a and Ref. 3). Accounting
for the 12 \( \text{Cr}^{3+} \) ions per unit cell, \( n = 6 \) for each orientation. Assuming a moment of
\( m = 2.8 \mu_B \) per \( \text{Cr}^{3+} \) ion [14], we calculate a surface magnetization of
\[
\sigma^0_z = \frac{6 \times 2.8 \mu_B \times 0.188 \text{ nm}}{0.870 \text{ nm}^3} = 3.64 \mu_B/\text{nm}^2 .
\]

This is slightly less than what one would expect from one monolayer of \( \text{Cr}^{3+} \) ions, which
has a magnetization of \( m/A = 4.38 \mu_B/\text{nm}^2 \).

2.2. Transformations between surface magnetization and magnetic field

Using the relations between magnetization and magnetic stray field for two-dimensional thin
films [15], we can reconstruct the surface magnetization \( \sigma_z(x, y) \) and vector magnetic field
\( \vec{B}(x, y) \) from the measured stray field component \( B_{NV}(x, y) \). We perform transformations in
Fourier space. The magnetic vector field \( \vec{B} \) associated with the magnetization \( \vec{\sigma} \) is given by
\[
\left( \hat{B}_x, \hat{B}_y, \hat{B}_z \right) = \frac{1}{2}\mu_0 e^{-kz} \left( -k_x\hat{\sigma}_k - ik_x\hat{\sigma}_z, -k_y\hat{\sigma}_k - ik_y\hat{\sigma}_z, -ik\hat{\sigma}_k + k\hat{\sigma}_z \right)
\]  

where \( k_x, k_y \) are the in-plane \( k \)-vectors, \( k = (k_x^2 + k_y^2)^{1/2} ; \hat{\sigma}_k = (k_x\hat{\sigma}_x + k_y\hat{\sigma}_y)/k \), and hat
symbols denote Fourier transforms in \( x \) and \( y \). \( z \) is the stand-off distance of the sensor and
\( \mu_0 = 4\pi \times 10^{-7} \text{Tm/A} \). For a line scan in \( x \) direction, scanned across a domain wall extending
in \( y \) direction, the magnetic field is
\[
\left( \hat{B}_x, \hat{B}_y, \hat{B}_z \right) = \frac{1}{2}\mu_0 e^{-kz} \left( -k_x\hat{\sigma}_x - ik_x\hat{\sigma}_z, 0, -ik\hat{\sigma}_x + k\hat{\sigma}_z \right) ,
\]  

For thick crystals a local magnetic probe only detects the stray field of the top layer.
where now $k = |k_x|$. Likewise, we can recover the magnetic vector field $\vec{B}$ from the measured projection $B_{NV}$ as

$$\left( \hat{B}_x, \hat{B}_y, \hat{B}_z \right) = \frac{1}{k_{NV}} (ik_x, ik_y, -k) \hat{B}_{NV}$$

where $k_{NV} = (ie_x k_x + ie_y k_y - e_z k)$ and $(e_x, e_y, e_z)$ is the vector orientation of the sensor. Finally, under the assumption that the magnetization is fully out-of-plane ($\sigma_x = \sigma_y = 0$), we can reconstruct $\sigma_z$ from the stray field $B_{NV}$,

$$\hat{\sigma}_z = -\frac{2W \hat{B}_{NV}}{\mu_0 e^{-kz} k_{NV}}$$

where $W = W(k)$ is a suitable window function (here a Hann function) that provides a high-frequency cutoff. Although our Cr$_2$O$_3$ films do have an in-plane component in the vicinity of the domain wall, the reconstructed $\sigma_z$ still accurately reproduces the domain pattern and surface magnetization $\sigma_z^0$.

### 2.3. Magnetic field from surface roughness

Surface roughness leads to tiny stray fields at topographic steps, as sketched in Fig. 1b. The magnetic field produced at a step of height $h$ corresponds to the differential field of two magnetized layers located at $z$ and $z + h$. According to Eq. 5, the $B_z$ field of the step is given by

$$\hat{B}_z = \frac{1}{2} \mu_0 e^{-kz} kh(k \hat{\sigma}_z) .$$

For a simple order-of-magnitude estimate of the stray field, we look at the Fourier component of $\hat{\sigma}_z$ that produces the strongest $B_z$. This occurs for $k = 2/z$. For this Fourier component, the amplitude of $B_z$ is

$$B_z = \frac{\mu_0 h 2e^{-2 \sigma_z^0}}{z^2} \approx \frac{0.2707 \mu_0 h \sigma_z^0}{z^2}$$

For our Cr$_2$O$_3$ crystals, where $\sigma_z^0 \approx 2 \mu_B$/nm$^2$, and using $z = 68$ nm, we find $B_z/h \approx 1.4 \mu$T/nm. For an rms surface roughness of 3 nm-rms we therefore expect stray field fluctuations of $\approx 5 \mu$T, in good agreement with the experimental 7 $\mu$T-rms (Fig. 2d).

### 2.4. Fitting of line scans

We model the domain wall as presented in Eq. 1 in the main text. The stray field is then computed via Eq. 5. The resulting model features 7 parameters: the effective surface magnetization $\sigma_z^0$, the position of the domain wall $x_0$, its width parameter $\Delta$ and twist angle
χ, and the sensor geometry (z, θ_{NV}, φ_{NV}). Since z, θ_{NV}, φ_{NV} have been determined separately at this point, they are left fixed in the following least-squares optimization, leaving only σ_z, x_0, Δ and χ as free parameters.

The initial value of σ_z^0 is determined by estimating the surface magnetization using the two complementary methods (step height, integration of B_x) described below. The initial value for width and chirality are set to Δ = 40 nm and χ = 90°. We checked that other starting values did not significantly alter the fit results. The fitting procedure is repeated for each individual line scan.

2.5. Complementary methods for estimating σ_z^0

We use two complementary methods for estimating the Cr_2O_3 surface magnetization σ_z^0 from a stray field scans across domain walls:

*Step height in reconstructed σ_z map:* We reconstruct the surface magnetization σ_z(s) using Eqs. (5) and (7). The step height at the domain wall is 2σ_z^0.

*Integration of B_x:* We assume a domain wall extending along the y direction. We compute the B_x(x) component of the stray field from B_{NV}(x), using the known orientation of the sensor (θ_{NV}, φ_{NV}) and Eq. (6). The integrated B_x(x) is then equal to μ_0σ_z^0, irrespective of the stand-off z and the domain-wall profile and chirality. To explain this, assume an out-of-plane magnetized film with magnetization \vec{σ}(x') and a domain wall centered at x = 0 and extending along the y-direction. The step edge can have a σ_x or σ_y component. The magnetic field B_x produced by the magnetization element dx'\vec{σ}(x') is

$$dB(x) = \frac{μ_0j_y(x')tx'z}{2π[(x - x')^2 + z^2]} \quad (10)$$

where j_y(x') = [\vec{∇} × \vec{σ}]_y(x') = -[∂_xσ_z](x') is the bound current element associated with \vec{σ}(x') and t is the film thickness (t ≪ z). The total magnetic field at position x is

$$B(x) = \int_{-∞}^{∞} dx' \frac{μ_0j_y(x')tx'z}{2π[(x - x')^2 + z^2]} = -\int_{-∞}^{∞} dx' \frac{μ_0z}{2π[(x - x')^2 + z^2]} [∂_xσ_z](x') \quad (11)$$

and the integrated B(x) is

$$\int_{-∞}^{∞} dxB(x) = -\left( \int_{-∞}^{∞} dx'' \frac{μ_0z}{2π[(x'')^2 + z^2]} \right) \left( \int_{-∞}^{∞} dx' [∂_xσ_z](x') \right) \quad (12)$$

$$= -\frac{μ_0}{2} [σ_z(+∞) - σ_z(-∞)] = μ_0σ_z^0 \quad (13)$$

where we have used Fubini’s theorem and the last equation is for a domain wall where [σ_z(+∞) - σ_z(-∞)] = -2σ_z^0.
2.6. Complementary method for estimating $\Delta$ and $\chi$

For a fixed pair $(\chi, \Delta)$, we only fit $x_0$ to the data, and record the residual sum of squares (RSS). The surface magnetization is determined for each line scan by the previously introduced three complementary methods. The RSS is a measure of the likelihood. Indeed, assuming Gaussian errors, the log-likelihood is given by

$$\ln \mathcal{L} = \ln \left( \frac{1}{2\pi\sigma^2} \right)^{n/2} - \frac{1}{2\sigma^2} \text{RSS}$$

(14)

Here, $\sigma$ is the standard deviation describing the error of a single data point, and $n$ is the number of data points. We can compare the relative likelihood of two models 1 and 2 (i.e. two pairs of $\Delta$ and $\chi$) by estimating $\sigma_i^2 = \text{RSS}_i/n, i \in \{1, 2\}$, giving

$$\ln \mathcal{L}_1 - \ln \mathcal{L}_2 = -\frac{n}{2} \ln \frac{\text{RSS}_1}{\text{RSS}_2}$$

(15)

We choose model 2 as the best model (i.e. the least squares solution), so that Eq. 15 is normalized to 0. To consider the data from all scans, we sum the RSS of each line and scan, and set $n$ to be the total number of data points.

3. MODEL OF ANTIFERROMAGNETIC DOMAIN WALL

We focus on orientational 180° domain walls, as they exist in Cr$_2$O$_3$, and compare their properties between ferromagnetic (FM) and collinear antiferromagnetic (AFM) systems. The domain wall properties, i.e., the domain wall profile, domain wall width $\Delta$ and twist angle $\chi$ are governed by the interplay between exchange, anisotropy and Zeeman energies, and, for FM systems, the demagnetizing field [16].

In a first step, we use a 1D model of a domain wall and only include exchange and anisotropy energies. We show that the domain wall properties are the same for FM and AFM and that the chirality is undetermined. The exchange energy arises from the Coulomb interaction and the Pauli principle and favors parallel (antiparallel) alignment of neighbored spins in the FM (AFM) case. The exchange energy is given by:

$$e_{\text{ex}} = -J m_i m_j \cos(\Delta \phi),$$

(16)

where $J$ is the exchange constant and $m_{i,j}$ are neighboring magnetic moments. $\Delta \phi$ describes the angle between the $i$'th and $j$'th moment [17]. In the FM (AFM) case the exchange constant $J$ is positive (negative). Note that a deviation from the parallel (antiparallel) alignment increases the energy equally for the FM and AFM case.

The magnetocrystalline anisotropy favors moments that lie along specific lattice directions, the so-called magnetic easy axes. In our model, we use a system with uniaxial anisotropy, which can be described as

$$e_{\text{ani}} = K \sin^2 (\theta),$$

(17)
where \( K \) is the uniaxial constant and \( \theta \) the angle between the easy axis and the moments \([17]\).

Similar to the exchange energy, a misalignment from the easy axis is equal for the FM and AFM case. Based on the fact that exchange and anisotropy act in similar ways on FM and AFM systems, we conclude that both systems have similar domain wall properties in the absence of a demagnetizing energy. This is also observed in more elaborated models \([18–21]\) and explicitly mentioned in Refs. 17, 22, 23.

In the following we derive an analytical expression for a 1D domain wall pointing along the \( x \)-direction, following Ref. 16, p.79-82. The easy axis of the uniaxial anisotropy is pointing along the \( z \)-direction.

In order to solve the problem analytically, we begin by writing the energy density in polar coordinates \((\theta, \phi)\):

\[
e = e_{\text{ex}} + e_{\text{ani}} = A \left[ (\partial \theta / \partial y)^2 + (\sin \theta \partial \phi / \partial y)^2 \right] + K \sin^2 \theta.
\] (18)

The term of the exchange energy is given for the one-dimensional case with the exchange stiffness \( A \), which is related to the exchange constant \( J \) (the same formula without azimuthal dependency is also provided in Ref. 17, Eq. (9), specifically for the AFM case). The static equilibrium is reached when all torques acting on the moments are zero. The solution, satisfying the boundary conditions \( \theta (\pm \infty) = (0, \pi) \), is given by:

\[
\phi (y) = \chi = \text{const.} \tag{19}
\]

\[
\theta (y) = \pm 2 \arctan \left[ \exp \left( y / \Delta_0 \right) \right], \tag{20}
\]

with the domain wall width \( \Delta_0 = \sqrt{A/K} \). The total energy per unit area of the wall \( \epsilon_0 \) is given by:

\[
\epsilon_0 = 4 \sqrt{AK}. \tag{21}
\]

At this point it is worth mentioning that the angle \( \phi(y) = \chi \) is arbitrary, or in other words, Néel and Bloch domain walls, or any combination of the two, are equal in energy for both the FM and AFM case. In a next step, we consider the demagnetizing field arising due to magnetic charges at the domain wall when \( \phi = \chi \neq 0 \). We further add an in-plane anisotropy \( K_p \) in the \( xy \)-plane with the easy axis pointing at an angle \( \psi_p \) with respect to the wall plane. We find that the domain wall width \( \Delta_0 \) and \( \epsilon_0 \) change to:

\[
\Delta = \Delta_0 \left[ 1 - \frac{4K}{\mu_0 M^2} \sin^2 \chi - \left( K_p/2K \right) \sin^2 (\chi - \psi_p) \right], \tag{22}
\]

\[
\sigma = \sigma_0 + \mu_0 M^2 \Delta_0 \sin^2 \chi + 2K_p \Delta_0 \sin^2 (\chi - \psi_p). \tag{23}
\]

Let us first consider the demagnetizing energy term \( \mu_0 M^2 \Delta_0 \sin^2 \chi \). We notice that in the FM case, the domain wall energy of a Bloch wall, \( \chi \in \{0, \pi\} \), is lower than that of a Néel wall, \( \chi \in \{\pm \pi/2\} \). Therefore, in the FM case and taking into account the demagnetizing field, Bloch walls are favored over Néel walls. For a generic AFM, if the volume magnetization \( M \) is zero, no energy contribution is expected. However, a residual demagnetizing field still
persists due to the small but finite magnetic moment of the wall [19, 23]. This field promotes the formation of a Bloch wall.

Adding an in-plane anisotropy $K_p$ forces the moments to cant along the in-plane easy axis when $K_p > \mu_0 M^2$. For sufficiently large in-plane anisotropy we observe a Néel-type domain wall if the in-plane easy axis is $\psi_p \approx \pm \pi/2$, i.e., when the domain wall runs perpendicular to the in-plane easy axis. For $\psi_p \approx 0$ and $\psi_p \approx \pi$, a Bloch-type domain wall is expected.

We note that the domain wall width of an Néel wall is reduced with respect to a Bloch domain wall according to Eq. (22). To estimate the reduction in wall width for Cr$_2$O$_3$, we divide the magnetization of the polarized surface layer $\sigma_z^0 = 1.6\mu_B/\text{nm}^2$ (Sample C) by the thickness of the cleaved layer, $t = 0.188$ nm. We use the anisotropy constant $K = 13$ kJ/m$^3$ given in Ref. 24. The ratio $r$ of the width of Néel to Bloch domain wall is then

$$ r = \frac{\Delta_{\text{Néel}}}{\Delta_{\text{Bloch}}} = \frac{\Delta_0 \left[ 1 - \frac{4K}{\mu_0 M^2} \right]}{\Delta_0} = 0.85. \quad (24) $$

which is in reasonable agreement with the experimental ratio of $r = 0.65 \pm 0.10$ (Fig. 4, caption) given the simplicity of our assumptions.
Fig. S1: SHG microscopy images of sample A (left panels) and sample B (right panels). Upper panels used left-handed circular polarization, lower panels used right-handed circular polarization.
Fig. S2: SHG microscopy images of sample C. Upper panel used left-handed circular polarization, lower panel used right-handed circular polarization.
Fig. S3: Maximum likelihood estimates for domain wall width $\Delta$ and twist angle $\chi$, as explained in the Methods section. Gray dots are fit results from individual line scans. Colored contours are maximum likelihood isolines containing 75%, 50%, and 25% of datapoints. The most likely $(\chi, \Delta)$ pair is indicated by a central cross. For sample C, datasets with $\alpha > 9^\circ$ (blue) and $\alpha < 9^\circ$ (red) are analyzed separately.
Fig. S4: Maximum likelihood estimates for domain wall width $\Delta$ and twist angle $\chi$ for upper and lower bound stand-off distances $z \pm 10$ nm, where $z$ is the calibrated stand-off distance. a-c Maximum likelihood estimates for the lower bound $z - 10$ nm. d-f Maximum likelihood estimates for the upper bound $z + 10$ nm. Data points, contours and central cross are as with Fig. S3. We note that the our observation – the presence of Bloch-like walls in samples A and B, and mixed Bloch and Néel walls in sample C – is valid within the uncertainty of the sample-sensor distance. The $z \pm 10$ nm bounds are a conservative estimate, as all probes showed a calibration error of $\leq 8$ nm (see Methods).
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