Magnetochiral Spin-Polarized Tunneling in a Paramagnetic State

Seongjoon Lim¹, Fei-Ting Huang¹, Shangke Pan¹², Kefeng Wang¹, Jaewook Kim¹, Sang-Wook Cheong¹*  

¹ Rutgers Center for Emergent Materials and Department of Physics and Astronomy, Piscataway, NJ 08854, USA  
²State Key Laboratory Base of Novel Function Materials and Preparation Science, School of Material Sciences and Chemical Engineering, Ningbo University, Ningbo, Zhejiang 315211, China  
*To whom correspondence should be addressed;  
Email: sange@physics.rutgers.edu

Abstract

Crystallographic chirality can mediate various optical and electrical magnetochiral effects. Since these effects have been studied in bulk optical, transport or non-local probe setups, investigation with a local probe is necessarily the next step towards further understanding of the intriguing phenomena closer to the quantum regime. We observed a spin-polarized scanning tunneling microscopy (SP-STM) contrast in the chiral domains of Co₁/₃NbS₂ in a paramagnetic state, which is unexpected in the conventional SP-STM mechanism. This spin-polarized tunneling, depending on the local structural chirality, is argued to be an inverse magnetochiral effect due to a dynamic coupling between tunneling electrons and chirality. In addition, using the standard STM, we also find magnetochiral nonreciprocal tunneling in the presence of external magnetic fields, considered as the inverse process. Our results demonstrate a new application of SP-STM in detecting the dynamic interaction of tunneling electrons with broken crystallographic symmetries.
Magnetochiral effects can arise from the broken mirror symmetry of chiral materials interacting with various moving entities such as photon$^1$, phonon$^2$, or electron$^3$. Lately, chirality-induced spin selection, where flow of photoelectrons gets spin-polarization through magnetochiral interaction, has been studied intensively on chiral molecules$^4$–$^7$. In crystalline materials, where crystallographic chirality interplays with various exotic magnetic orders such as skyrmion$^8$, polar magnetism$^9$, multiferroicity$^{10}$, and chiral topological spin texture$^{11,12}$, a similar effect has been found that electric conduction through a chiral crystal, e.g. trigonal Te$^{13}$–$^{15}$, can directly induce magnetic moment. Interestingly, the THz-range Faraday effect of chiral Te in the presence of electric current was reported four decades ago, and can be simply explained in terms of the induced magnetization by a current$^{16}$. These effects can also be understood based on the symmetry operational similarity (SOS) between a magnetic moment and a chiral structure placed in a quasi-equilibrium electric conduction$^{17,18}$. Although the chirality-induced spin selection has a classical analogy of the magnetic-field generation by flowing electric current through a conducting coil, the quantum mechanical understanding is not intuitive and there is still much room for improvement$^4$–$^5$. Specifically, there still lacks any detailed investigation of the magnetochiral interaction at the atomic level where quantum mechanical tweak often acts in a counter-intuitive way. Moreover, whether the magnetochiral interaction can be associated with local probing techniques such as STM is a crucial question$^{19}$ that might provide superior spatial resolution compared to the existing bulk optical or transport techniques in chirality studies$^{20}$–$^{22}$. However, it is highly unclear how the accumulated effect in bulk contributes to the surface measurement with an atomic spatial-resolution, or even whether the spin-polarization induced by the magnetochiral interaction is a quantity that can be detected with SP-STM.
SP-STM is a powerful tool to visualize various magnetic orders in, e.g. ferromagnets, antiferromagnets, helimagnets, and skyrmion systems, at the atomic scale. In addition, considering various interactions that magnetism can have with other order parameters, its use may be not limited to visualizing magnetic orders. In this paper, we report our discovery of a novel spin-polarization mechanism in the quantum tunneling due to the magneto-chiral interaction of electrons with a chiral structure and demonstrate the detection of the atomic chiral environment with SP-STM. As an exemplary material, we chose Co$_{1/3}$NbS$_2$, one of the chiral intercalated transition metal dichalcogenides, where we found, for the first time, the numerous structural chiral domains outlined by a network of topological vortex/antivortex. The existence of micron-scale structural chiral domain boundaries makes this system unique to detect the change of magneto-chiral interaction across the domain boundaries. Moreover, Cr$_{1/3}$NbS$_2$ (isostructural to Co$_{1/3}$NbS$_2$) has been reported to show magneto-chiral interactions in a bulk transport setup, i.e. chirality-induced spin transport and anomalous nonreciprocal electrical transport. Specifically, Co$_{1/3}$NbS$_2$, compared with Cr$_{1/3}$NbS$_2$, is advantageous since it shows paramagnetic magnetism at temperatures above the antiferromagnetic transition temperature of ~26 K, thus we can eliminate the influence of magnetic ordering at the liquid nitrogen temperature and test solely the intrinsic magneto-chiral effect. Furthermore, a study of the magneto-chiral interaction in chiral van der Waals materials could suggest a novel mechanism that controls spin degrees of freedom with electrical means for two-dimensional spintronics.

**Structural Chiral Domains and Topological Vortices**

The crystal structure of Co$_{1/3}$NbS$_2$ consists of 2H-NbS$_2$ matrix and intercalated Co ions occupying one of three Nb-atop sites, A, B, and C. The Co ions are arranged in such a way that the
intercalation site changes alternately along the $z$ direction, e.g. AB-type stacking. In Fig. 1a, we draw the AB-stacked and BA-stacked structures with opposite chiralities. The planar displacements of S ions determine the chirality of the structures as shown with red or blue triangular arrows. In relation with the fictitious cleaving planes (the horizontal black lines), the top-most NbS$_2$ layer has upper and lower Co ion lattices, and the first and second characters in a stacking sequence denote the intercalation site of the upper and lower Co lattices, respectively. Throughout this paper, we will use Upper (Lower) Co to refer the top-most (subsurface) Co lattice. There exist six combinations of stacking sequence, the chirality of which can be identified from the permutation sequence (i.e. [AB, BC, CA] and [BA, CB, AC] have the opposite chiralities). The structure belongs to one of the Sohncke space groups (P6$_3$22) that can bear the opposite chiralities within one space group$^{31}$. The chiral $P6_322$ structure is obtained from the high-temperature $P6_3/mmc$ prototype through Co ordering and S displacements in Co$_{1/3}$NbS$_2$, accompanied by the cell tripling. Ref. 27 reported the first discovery that in chiral Fe$_{1/3}$TaS$_2$, the six-types of domains are interlocked around a topological vortex/antivortex core, and adjacent domains always show the opposite chirality$^{27}$. Our superlattice dark-field transmission electron microscopy (DF-TEM) image of Co$_{1/3}$NbS$_2$ manifests clear micron-scale bright and dark regions, corresponding to opposite-chirality (i.e., left-handed and right-handed) chiral domains, as shown in Fig. 1b. The DF-TEM image also shows that similar with Fe$_{1/3}$TaS$_2$, Co$_{1/3}$NbS$_2$ displays topological vortex domains: depending on the sign of the vorticity, a topological defect is either a topological vortex or antivortex, and these vortices and antivortices tend to be paired in a few-micrometers scale, which can be manipulated through the cooling rate across $T_c$. (see Supplementary Information, SI Fig. S1) Fig. 1c represents the atomic model of the chiral domains around a vortex core. Note that there are two types of domain boundaries, solid and dashed lines, where we have a change of
intercalation sites only in Upper or Lower Co, respectively. We will similarly refer them as Upper and Lower domain boundaries. Then, it is evident that a cleaved surface only exposes three Upper domain boundaries while rest three Lower domain boundaries exist underneath the top-most NbS$_2$ layer, which is an important structural aspect in STM images. In Fig. 1d, we show the magnetic susceptibility revealing antiferromagnetic ordering below 26 K, which is consistent with the previous result$^{32}$. Since all the following experiments are performed at the liquid nitrogen temperature, we expect no spin-polarized signal from ordered Co magnetic moments, which is in the paramagnetic state.

**Chiral Structural Domains in STM/SP-STM**

Co$_{1/3}$NbS$_2$ is cleaved at a sample stage cooled by liquid nitrogen to minimize the effect of thermal diffusion. (SI note 1) Because Co ions reside in the van der Waals gap, they can remain on either side of exposed surfaces, so two types of surface, e.g. Co-/NbS$_2$-types, can be produced. Fig. 2a shows the STM topographies, the simulated images, and FFTs of the two types. The primary difference is the predominant periodicity shown in the images as well as in FFT. The Co-type surface is dominated by \((\sqrt{3} \times \sqrt{3})R30^\circ\) super-structure peak (red circles) while NbS$_2$-type one reveals more obvious \((1 \times 1)\) peak (white circles) of the host unit cell. Although both types show mixed traces of two periodicities, the type of surface can be identified from the stronger peaks in FFT (especially at low biases). The coexistence of the two types becomes more obvious when it comes to a boundary between the two types as shown in Fig. 2b.

In order to compare STM and SP-STM on chiral domains, we have used two Pt/Ir-alloy tips that are treated on non-magnetic Cu(111) and magnetic Cr(001) surface, respectively. The SP-STM tip
is coated with Cr from the Cr(001) surface, and its magnetic sensitivity is verified by the layer-by-layer contrast of Cr(001) antiferromagnetic order\textsuperscript{33}. (SI note 2) It is known that the magnetization direction of antiferromagnetic Cr tips tends to be randomly oriented, so it shows both in-plane and out-of-plane sensitivity\textsuperscript{34}. Throughout this paper, we assume the dominant out-of-plane sensitivity along the chiral axis, which is the expected induced magnetization direction by symmetry\textsuperscript{17}. (It is also consistent with the observed two levels of contrast, which will be discussed later.) Fig. 2c and 2d show the topographies of topological vortices in STM and SP-STM on Co-type surfaces. It is immediately noticeable that STM shows only “three” Upper domain boundaries depicted by aligned Co deficiencies, while SP-STM shows “domain contrast” exhibiting “six” domains and six boundaries for one vortex. Considering the fact that Co lattices exhibit localized orbitals well separated by the delocalized electrons of NbS\textsubscript{2}\textsuperscript{35}, the appearance of only three Upper domain boundaries in STM seems reasonable. The observed Co-deficient Upper domain boundaries are consistent with the fine atomic-structure model of domain boundaries around a topological vortex core. (SI note 3) On the other hand, the significant domain contrast in SP-STM is obviously not from magnetic order at this paramagnetic temperature, and we attribute it to magnetochiral interaction as will be discussed later. Note that the domain contrast becomes clearer in a larger size scan as it averages out the Co deficiencies. (Fig. 2d inset)

To examine the Co lattice shift, we obtained atomic images near boundaries. The STM image in Fig. 3a shows lined-up Co deficiencies that form an Upper domain boundary (gray solid line). As the intercalation sites, A, B, and C, have a relative translational shift that corresponds to 1/3 of the unit cell length (along both of the \(a\) and \(b\) directions), Co lattice manifests a \(2\pi/3\) phase shift upon changing intercalation site. The shift is evident in the expanded image in Fig. 3d, which is consistent with the atomic model of an Upper domain boundary given in Fig. 3g. Note that this
atomic-scale configuration of a domain boundary is also consistent with the extinction rule for the antiphase boundaries in super-lattice DF-TEM image\textsuperscript{27}. (SI note 3) To our best knowledge, this is the first atomic observation of topological vortex domain boundaries in chiral 1/3 intercalated transition metal dichalcogenides that can reveal the topological nature of vortex structures. Regarding the topological nature, we emphasize that the S displacement vectors, in modulus to unit cell translation, exhibits a $2\pi/3$ rotation between adjacent domains, and thus enclosing a topological vortex core results in an overall $4\pi$ phase shift, i.e. the topological charge of $\pm 2^{36}$. (SI note 4) The completion of full rotation in the order parameter space (i.e. the presence of topological charge) is closely related to the robustness of the defect structure (i.e. topological protection), especially, to the prohibited domain boundary switch. (SI Fig. S4)

Domain boundaries obtained with SP-STM show more complicated and non-trivial features. The main difference is that it shows contrast-change corresponding to the chirality (i.e. domain contrast), which is shown as depressed height in the middle domain of Fig. 3b. As a result, both the Upper and the Lower domain boundaries are traced by gray solid and dashed lines, respectively. The Co lattice shift of the Upper domain boundary (Fig. 3e) follows the same manner as in the case of STM and is consistent with Fig. 3g again. However, in case of the Lower domain boundary, there is no shift as shown in Fig. 3f, and it matches with the model of Fig. 3h. Instead of phase shift, the domain boundary exhibits a step-like topographic change as depicted in the height profile in Fig. 3c. Note that the alternating contrast shows a direct correlation with the chirality and is consistent with the expected current-induced magnetization along the electric current direction in transport setup\textsuperscript{19} and within SOS concept\textsuperscript{17}. Emphasize that instead of phase shift, the change of atomic “chiral” environment makes the spin-polarized signal only appearing in SP-STM, not in STM.
Inverse Magnetochiral Effect

The spectroscopic signature of the magnetochiral interaction has been checked by obtaining tunneling spectra across a domain boundary in SP-STM. Fig. 4a shows a series of spin-polarized tunneling spectra across a Lower domain boundary. While the modulation by atomic corrugation is visible throughout the spectra, there is a definitive change of spectral intensity across the boundary. To examine the change in more detail, the spectral intensities from two representative biases are depicted in Fig. 4b. The changes of spectral intensity that follow step-like guides are obvious, and the step-like feature is a result of adding the opposite magnetochiral interaction to the base tunneling spectra (gray lines). Therefore, the domain contrast in topography is due to the enhanced/suppressed tunneling probability upon chirality change. Next, we check the dependence on the electric field direction. Although it can be seen briefly in the inversion of the step-like guides in Fig. 4b, it is depicted fully by comparing the spectra obtained from the two points (red and blue stars in Fig. 4a). The two spectra in Fig. 4c indicate zero bias as the crossing point where the magnetochiral interaction vanishes. We can further eliminate the “non-magnetochiral portion” by getting polarization \( P = (G_\uparrow - G_\downarrow)/(G_\uparrow + G_\downarrow) \) where \( G_\uparrow(G_\downarrow) \) depicts the tunneling conductance of parallel(antiparallel) spin-polarized tunneling (Fig. 4c inset), which denotes zero bias as its crossing point. In other words, the effect is exactly antisymmetric about the zero bias. Generally, in any magnetic ordered states, spin polarization of tunneling current reflects the difference between opposite spin states, and the zero-crossing point in polarization appears at a somewhat arbitrary energy level, as can be found in many examples in the comprehensive review by Wiesendanger\(^{37}\). The coincidence of the crossing point of spin polarization with zero bias is a
crucial characteristic, which is remarkably different from the conventional mechanism for SP-STM contrasts.

We try to understand our observation based on the comparison with the previously known two magnetochiral phenomena: (A) current-induced magnetization in bulk chiral crystal\cite{13,19} and (B) spin-polarization in photoelectron transmission of chiral molecules\cite{5,6}. First, our result has a similarity with (A) in the sense that the material has chirality in bulk structure but is different from (A) since our measurement detects atomic local regions. The observation of spin-polarized current in chiral Cr\textsubscript{1/3}NbS\textsubscript{2} with bulk transport setups\cite{19} was claimed to be associated with the nonlocal measurement of diffused spins, which is distinct from our results with local tunneling current. This remarkable difference makes it difficult to employ the theories relevant to (A)\cite{14,15,38,39} directly. The local-probing nature is in line with (B), dealing transmitted photoelectrons interacting with an individual chiral molecule. However, a perturbative approach\cite{5} in (B) utilizing a small spin-orbit coupling of light atoms composing chiral molecules cannot be applied to Co\textsubscript{1/3}NbS\textsubscript{2}, where d-orbital bands of Co/Nb dominate the electronic structure near the Fermi energy\cite{40}. Moreover, the nature of tunneling electrons and photoelectrons\cite{6} should be very much different. As a result, we argue that a new theoretical framework is required for the unprecedented atomic observation of the magnetochiral interaction, which we will refer as the inverse magnetochiral effect. (the name is used in contrast to the various magnetochiral effects exhibiting nonreciprocal change of quantities induced by magnetic field\cite{1,3}) We pay particular attention to the common characteristics among our result and the cases in (A) and (B). Firstly, the induced spin-polarization should be reversible by changing chirality. Secondly, since the effect is caused by interaction between non-equilibrium electric carrier and chiral structure, the effect should disappear when electric current vanishes and reverses when current flows backwards\cite{5,14,15,38}. We emphasize that both of these are
already shown in the tunneling spectra analysis by the reversal upon chirality change and the zero-bias crossing point. Thus, we argue that all these magnetochiral phenomena have something in common at the fundamental level.

**Magnetochiral Nonreciprocal Tunneling**

Lastly, we test the cross-coupling between tunneling electrons and magnetism. In, for example, linear magnetoelectric systems, the magnetoelectric coupling depends on the strength of linearly cross-coupled term (i.e. $E \cdot H$) in free energy$^{41}$, and gives a mutual relationship that induces one by another. A similar relationship can be assumed for the inverse magnetochiral effect. In other words, “electric tunneling induced by magnetic field” and “magnetization induced by tunneling electron” are all possible in chiral systems. However, any permanent electric current induced by magnetic field in an isolated system is thermodynamically prohibited, unless it is, for example superconducting. Instead, when there is a current flow by an external electromotive force, the magnetochiral interaction is in the form of enhancement/suppression of electric current. This phenomenon has already been reported in single wall carbon nanotube$^{42}$, chiral MnSi$^3$, or helically deformed Bi crystal$^{43}$ as known as electrical magnetochiral anisotropy. The chirality-dependent resistance-change under magnetic field has an analogue with tunneling probability change in the presence of external magnetic field in a STM setup although it is not straightforward to match tunneling probability and resistance. We will refer this effect as magnetochiral nonreciprocal tunneling. Note that it does not require any magnetic probe to see this effect. The STM image in Fig. 4d demonstrates the emergence of domain contrast upon application of a magnetic field, which confirms the existence of the magnetochiral nonreciprocal tunneling effect. Although, from the
viewpoint of SOS, it was predicted that the magnetic field applied to a chiral structure could exhibit nonreciprocity\textsuperscript{17}, it has never been associated with electric tunneling or scanning probe microscopy.

In conclusion, we have demonstrated that the inverse magnetochiral effect can be exploited to observe chiral domains and domain boundaries using SP-STM. The inverse magnetochiral effect is reversible readily by changing chirality or the electron tunneling direction, which is in a clear distinction with the conventional SP-STM contrasts from different orientations of ordered local spins. Thus, SP-STM can, now, be utilized to reveal atomic-scale chiral structures, in addition to atomic spin structures. Moreover, the magnetochiral nonreciprocal tunneling, an inverse phenomenon, gives rise to domain contrast upon application of external magnetic field without using a magnetized probe. Our observations provide a new paradigm to unveil quantum-level interrelationship between electric tunneling and crystallographic chirality. We also would like to remark that the novel spin-polarization mechanism using a chiral van der Waals material can be an innovative spin-polarizing technique in two-dimensional spintronics, where spin-injection has been relied on integration of ferromagnetic materials or circularly polarized light so far\textsuperscript{30}.
Methods

Crystal growth

Single crystals of $\text{Co}_{\frac{1}{3}}\text{NbS}_2$ were grown by a chemical vapor transport reaction method in the presence of iodine as a transport agent. About 0.2 g mixture of cobalt powder (Alfa Aesar, 99.99%), niobium powder (Alfa Aesar, 99.9%), and sulfur piece (Alfa Aesar, 99.999%) with a molar ratio Co: Nb: S = 0.7:1:2 were sealed in a quartz tube with 100 mg of iodine (Alfa Aesar, 99.99%) under vacuum. Then, the quartz tube was placed under an optimum temperature gradient in a two-zone tube furnace. The hot and cold zones were kept at 950°C and 800°C, respectively. After about 4 weeks, the tube furnace was turned off and the quartz tube was cooled to room temperature naturally. Crystals in hexagonal shape were collected both at the hot and cold end of the quartz tube. X-ray diffraction data on ground powder specimens were taken at room temperature with Cu K$_\alpha$ ($\lambda = 0.15418$ nm) radiation in a Malvern Panalytical X’Pert 3 powder diffractometer.

Transmission Electron Microscopy

Specimens for TEM experiments were prepared by Ar-ion milling and studied using a JEOL-2010F TEM. To unveil the chiral domains, DF-TEM images were taken by selecting $g_{\pm} = \pm 222$ along the [101] zone axis. All images are raw data.

Scanning Tunneling Microscopy

STM measurements were performed using a Unisoku ultra-high vacuum SPM system (USM-1500) equipped with a cleaving stage capable of cryogenic cooling. Cu(111) and Cr(001) samples cleaned by repeated cycles of sputtering and annealing have been used as reference samples as well as a tip treatment base. Two Pt/Ir alloy tips were prepared by electron bombardment heating in ultra-high vacuum, and treated separately on Cu(111) and Cr(001) surface to be used for normal STM and SP-STM, respectively. The treatment was done by tip shaping mechanism implemented
in the controller (Nanonis) typically with indentation into the surface with a negative bias to the tip and pulling out with a few nm/s of speed. Several repetitions of the procedure result in Cr transfer to the tip. The magnetization of Cr-coated tips are checked by tunneling spectroscopy across a step edge with the alternating contrast of antiferromagnetic ordering of Cr(001) as described in SI Note 2. Co$_{1/3}$NbS$_2$ sample is glued to a sample plate by silver epoxy (Epotek H20E) and a metal post is attached to the top with the same epoxy. Then the sample is cleaved at 80 K in the cleaving stage after precooling of 1 hour. All the STM and SP-STM measurements are acquired at 78 K. The tunneling spectroscopy measurement is obtained by modulation of bias and demodulation of tunneling current using lock-in technique.

**STM Simulation**

Density functional theory Calculations were performed using GPAW package$^{44}$ within the PAW formalism implemented in Atomic Simulation Environment$^{45}$ (ASE). We used PBE exchange-correlation functional$^{46}$ with SCF convergence criteria of $4.0 \times 10^{-8}$ eV$^2$/el. (for eigenstates), $5.0 \times 10^{-4}$ eV (energy), and $10^{-4}$ electrons (electron density). Gamma-centered k-point mesh of $(2 \times 2 \times 2)$ was used to sample the $k$-space, and 0.2 Å real space grid was used for wavefunction expansion in the finite-difference scheme. The surface of Co$_{1/3}$NbS$_2$ was modeled with four slabs of 2H-NbS$_2$ with embedded intercalants in between. The top-most Co lattice was added/removed according to the type of surface that is being simulated with 10 Å of vacuum added on top. The STM simulations were performed using the local density of states obtained from the density functional theory calculations following the Tersoff-Hamann approach$^{47}$ as implemented in the GPAW package.
Author contributions
S.C. initiated the study; S.L. performed STM and simulation; F.H. performed the TEM; S.P. and K.W. grew the crystals; J.K. performed susceptibility measurement; S.L. and S.C. analyzed the data and wrote the paper.

Acknowledgement
This work was supported by the center for Quantum Materials Synthesis (cQMS), funded by the Gordon and Betty Moore Foundation’s EPiQS initiative through grant GBMF6402, and by Rutgers University.
Figure 1. Structural Chiral Domains and Topological Vortices in \( \text{Co}_{1/3}\text{NbS}_2 \) (a) Atomic model of \( \text{Co}_{1/3}\text{NbS}_2 \) with opposite chiralities. After cleaving (horizontal black lines), the top-most \( \text{NbS}_2 \) layer has upper (orange circle) and lower (dashed orange circle) Co ions, which are referred as Upper and Lower Co, respectively. (light-blue polyhedral, light blue: Nb, yellow: S) There are six possible stacking sequences, and two of them, AB (upper, right-handed) and BA (lower, left-handed), are shown. The local chiral environments of Co ions are depicted by displacements of S ions as shown as red or blue triangular arrows. (b) DF-TEM observation of a topological vortex-antivortex pair. Six chiral domain boundaries merge into a topological vortex/antivortex while any two adjacent domains have the opposite chirality. (scale bar: 200 nm) (c) Atomic model of domain boundaries around a topological vortex. Domain boundaries having lattice shift in Upper/Lower Co lattice are referred as Upper/Lower domain boundary (Solid/Dashed Lines), respectively. (d) Temperature dependence of the magnetic susceptibility revealing the antiferromagnetic phase transition at 26 K. Several measurement schemes upon field/zero-field cool (FC/ZFC) or field along \( c \)-axis/\( ab \)-plane (\( H_c/H_{ab} \)) are shown.
Figure 2. STM and SP-STM of Chiral Domains around a Topological Vortex (a) STM topography/simulated image/FFT of Co- (upper) and NbS$_2$- (lower) type surface. The Co-type surface exhibits principal periodicity of ($\sqrt{3} \times \sqrt{3}$)$R30^\circ$ superstructure (red circles) while the NbS$_2$-type exhibits (1 × 1) lattice (white circles). (scale bars: 0.7 nm) (b) STM topography of a transition area between Co- and NbS$_2$-type surfaces. (c) STM observation of a topological vortex. (5 mV, 5 pA, scale bar: 10 nm) Among the six chiral domain boundaries, only three Upper domain boundaries are revealed as three dark lines. (d) SP-STM observation of a topological vortex. (-50 mV, 10 pA, scale bar: 10 nm) Domain contrast is generated by the inverse magnetochiral effect and it is locked to the structural chirality which makes alternating contrast between adjacent domains. (inset: large area SP-STM image, scale bar: 20 nm)
Figure 3. Atomic Configurations of Upper and Lower Domain Boundaries

(a) Upper domain boundary (solid line) in STM topography. (b) Upper (solid) and Lower (dashed line) domain boundaries in SP-STM topography. Domain contrast appears as suppressed height in the middle domain region due to the inverse magnetochiral effect. (scale bars: 5 nm) (c) Height profile from the black line in (b) depicting the step-like change of topographic height about 22 pm. (d,e,f) Closeup views of the three areas depicted by black rectangles in (a) and (b). The shift of Co lattice between two different intercalation sites (blue and red) is visible in the Upper domain boundaries (d and e), while no shift is observed in the Lower domain boundary (f). (scale bars: 0.5 nm) (g,h) Atomic model of an Upper (g) and a Lower (h) domain boundary showing the shift of Co lattice only in the Upper and the Lower Co lattice, respectively.
Figure 4. Reversibility of the Magnetochiral Interaction. (a) spin-polarized tunneling spectra across a Lower domain boundary. (upper) SP-STM topography of a region where the spectra are taken. (lower) spatial variation of tunneling spectra across the center domain boundary. (vertical dashed line) (b) Two tunneling spectra that correspond to positive and negative sample biases ($\pm 40$ mV) extracted from the horizontal dashed lines in (a). The opposite magnetochiral interaction in two domains gives suppression (enhancement) of tunneling spectrum in the left (right) domain in the positive bias, while the effect is reversed in the negative bias. (c) Bias dependence of spin-polarized tunneling spectra obtained from two positions, red and blue stars in (a), in the opposite chiral domains. Inset: polarization ratio calculated from the spectra (All four combinations of domain chiralities and electron tunneling directions are depicted as schematics in b and c) (d) domain contrast revealed by magnetochiral nonreciprocal tunneling effect in STM upon application of external magnetic field. (out-of-plane magnetic field 0.2 T, scale bar: 50 nm)
References

1. Rikken, G. L. J. A. & Raupach, E. Observation of magneto-chiral dichroism. *Nature* **390**, 493–494 (1997).

2. Nomura, T. *et al.* Phonon Magnetochiral Effect. *Phys. Rev. Lett.* **122**, 145901 (2019).

3. Yokouchi, T. *et al.* Electrical magnetochiral effect induced by chiral spin fluctuations. *Nat Commun* **8**, 866 (2017).

4. Yang, X., van der Wal, C. H. & van Wees, B. J. Spin-dependent electron transmission model for chiral molecules in mesoscopic devices. *Phys. Rev. B* **99**, 024418 (2019).

5. Dalum, S. & Hedegård, P. Theory of Chiral Induced Spin Selectivity. *Nano Lett.* **19**, 5253–5259 (2019).

6. Gohler, B. *et al.* Spin Selectivity in Electron Transmission Through Self-Assembled Monolayers of Double-Stranded DNA. *Science* **331**, 894–897 (2011).

7. Pop, F., Auban-Senzier, P., Canadell, E., Rikken, G. L. J. A. & Avarvari, N. Electrical magnetochiral anisotropy in a bulk chiral molecular conductor. *Nature Communications* **5**, 3757 (2014).

8. Mühlbauer, S. *et al.* Skyrmion Lattice in a Chiral Magnet. *Science* **323**, 915–919 (2009).

9. Johnson, R. D. *et al.* MnSb 2 O 6 : A Polar Magnet with a Chiral Crystal Structure. *Physical Review Letters* **111**, (2013).

10. Johnson, R. D. *et al.* Cu3Nb2O8: A Multiferroic with Chiral Coupling to the Crystal Structure. *Phys. Rev. Lett.* **107**, 137205 (2011).

11. Inoshita, T., Hirayama, M., Hamada, N., Hosono, H. & Murakami, S. Topological semimetal phases manifested in transition metal dichalcogenides intercalated with $3d$ metals. *Phys. Rev. B* **100**, 121112 (2019).

12. Schröter, N. B. M. *et al.* Chiral topological semimetal with multifold band crossings and long Fermi arcs. *Nature Physics* **15**, 759–765 (2019).
13. Furukawa, T., Shimokawa, Y., Kobayashi, K. & Itou, T. Observation of current-induced bulk magnetization in elemental tellurium. *Nature Communications* **8**, 1–5 (2017).

14. Şahin, C., Rou, J., Ma, J. & Pesin, D. A. Pancharatnam-Berry phase and kinetic magnetoelectric effect in trigonal tellurium. *Phys. Rev. B* **97**, 205206 (2018).

15. Yoda, T., Yokoyama, T. & Murakami, S. Orbital Edelstein Effect as a Condensed-Matter Analog of Solenoids. *Nano Lett.* **18**, 916–920 (2018).

16. Vorob’ev, L. E. *et al.* Optical activity in tellurium induced by a current. *JETP Lett.* **29**, 441–445 (1979).

17. Cheong, S.-W. SOS: symmetry-operational similarity. *npj Quantum Mater.* **4**, 1–9 (2019).

18. Cheong, S.-W. Trompe L’oeil Ferromagnetism. *npj Quantum Materials* **5**, 1–8 (2020).

19. Inui, A. *et al.* Chirality-Induced Spin-Polarized State of a Chiral Crystal CrNb3S6. *Phys. Rev. Lett.* **124**, 166602 (2020).

20. Cireasa, R. *et al.* Probing molecular chirality on a sub-femtosecond timescale. *Nature Phys* **11**, 654–658 (2015).

21. Neufeld, O. *et al.* Ultrasensitive Chiral Spectroscopy by Dynamical Symmetry Breaking in High Harmonic Generation. *Phys. Rev. X* **9**, 031002 (2019).

22. Li, W. *et al.* Circularly polarized light detection with hot electrons in chiral plasmonic metamaterials. *Nature Communications* **6**, 8379 (2015).

23. Elmers, H. J. *et al.* Submonolayer Magnetism of Fe(110) on W(110): Finite Width Scaling of Stripes and Percolation between Islands. *Phys. Rev. Lett.* **73**, 898–901 (1994).

24. Heinze, S. *et al.* Real-Space Imaging of Two-Dimensional Antiferromagnetism on the Atomic Scale. *Science* **288**, 1805–1808 (2000).

25. Haze, M., Yoshida, Y. & Hasegawa, Y. Experimental verification of the rotational type of chiral spin spiral structures by spin-polarized scanning tunneling microscopy. *Sci Rep* **7**, 1–5 (2017).

26. Romming, N. *et al.* Writing and Deleting Single Magnetic Skyrmions. *Science* **341**, 636–639 (2013).
27. Horibe, Y. et al. Color Theorems, Chiral Domain Topology, and Magnetic Properties of Fe₅TaS₂. *J. Am. Chem. Soc.* **136**, 8368–8373 (2014).

28. Aoki, R., Kousaka, Y. & Togawa, Y. Anomalous Nonreciprocal Electrical Transport on Chiral Magnetic Order. *Phys. Rev. Lett.* **122**, 057206 (2019).

29. Baričić, N. et al. High-pressure study of transport properties in Co0.33NbS₂. *Phys. Rev. B* **84**, 075157 (2011).

30. Avsar, A. et al. Colloquium: Spintronics in graphene and other two-dimensional materials. *Rev. Mod. Phys.* **92**, 021003 (2020).

31. Flack, H. D. Chiral and Achiral Crystal Structures. *Helv. Chim. Acta* **86**, 905–921 (2003).

32. Parkin, S. S. & Friend, R. H. 3d transition-metal intercalates of the niobium and tantalum dichalcogenides i. Magnetic properties. *Philosophical Magazine B: Physics of Condensed Matter; Statistical Mechanics, Electronic, Optical and Magnetic Properties* **41**, 65–93 (1980).

33. Kleiber, M., Bode, M., Ravlić, R. & Wiesendanger, R. Topology-Induced Spin Frustrations at the Cr(001) Surface Studied by Spin-Polarized Scanning Tunneling Spectroscopy. *Phys. Rev. Lett.* **85**, 4606–4609 (2000).

34. Schlenhoff, A., Krause, S., Herzog, G. & Wiesendanger, R. Bulk Cr tips with full spatial magnetic sensitivity for spin-polarized scanning tunneling microscopy. *Appl. Phys. Lett.* **97**, 083104 (2010).

35. Nakayama, M., Miwa, K., Ikuta, H., Hinode, H. & Wakihara, M. Electronic Structure of Intercalation Compounds of CoₓNbS₂. *Chem. Mater.* **18**, 4996–5001 (2006).

36. Huang, F.-T. & Cheong, S.-W. Aperiodic topological order in the domain configurations of functional materials. *Nature Reviews Materials* **2**, 17004 (2017).

37. Wiesendanger, R. Spin mapping at the nanoscale and atomic scale. *Rev. Mod. Phys.* **81**, 1495–1550 (2009).
38. Yoda, T., Yokoyama, T. & Murakami, S. Current-induced Orbital and Spin Magnetizations in Crystals with Helical Structure. *Scientific Reports* **5**, 1–7 (2015).

39. Massarelli, G., Wu, B. & Paramekanti, A. Orbital Edelstein effect from density-wave order. *Phys. Rev. B* **100**, 075136 (2019).

40. Polesya, S., Mankovsky, S. & Ebert, H. Electronic and magnetic properties of the 2H-NbS2 intercalated by 3d transition metal atoms. *Zeitschrift für Naturforschung B* **74**, 91–98 (2019).

41. Fiebig, M. Revival of the magnetoelectric effect. *J. Phys. D: Appl. Phys.* **38**, R123–R152 (2005).

42. Krstić, V., Roth, S., Burghard, M., Kern, K. & Rikken, G. L. J. A. Magneto-chiral anisotropy in charge transport through single-walled carbon nanotubes. *The Journal of Chemical Physics* **117**, 11315–11319 (2002).

43. Rikken, G. L. J. A., Fölling, J. & Wyder, P. Electrical Magnetochiral Anisotropy. *Phys. Rev. Lett.* **87**, 236602 (2001).

44. Mortensen, J. J., Hansen, L. B. & Jacobsen, K. W. Real-space grid implementation of the projector augmented wave method. *Phys. Rev. B* **71**, 035109 (2005).

45. Larsen, A. H. *et al.* The atomic simulation environment—a Python library for working with atoms. *J. Phys.: Condens. Matter* **29**, 273002 (2017).

46. Perdew, J. P., Burke, K. & Ernzerhof, M. Generalized Gradient Approximation Made Simple. *Phys. Rev. Lett.* **77**, 3865–3868 (1996).

47. Tersoff, J. & Hamann, D. R. Theory of the scanning tunneling microscope. *Phys. Rev. B* **31**, 805–813 (1985).