

Experimental realization of an intrinsic magnetic topological insulator

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Intrinsic magnetic topological insulator (TI) is a stoichiometric magnetic compound possessing both inherent magnetic order and topological electronic states. Such a material can provide a shortcut to various novel topological quantum effects but remains elusive experimentally so far. Here, we report the experimental realization of high-quality thin films of an intrinsic magnetic TI—MnBi₂Te₄—by alternate growth of a Bi₂Te₃ quintuple-layer and a MnTe

bilayer with molecular beam epitaxy. The material shows the archetypical Dirac surface states in angle-resolved photoemission spectroscopy and is demonstrated to be an antiferromagnetic topological insulator with ferromagnetic surfaces by magnetic and transport measurements as well as first-principles calculations. The unique magnetic and topological electronic structures and their interplays enable the material to embody rich quantum phases such as quantum anomalous Hall insulators and axion insulators in a well-controlled way.

A topological insulator (TI) is non-magnetic, carrying gapless surface electronic states topologically protected by the time-reversal symmetry (TRS) (1, 2). Many exotic quantum effects predicted for TIs, however, need the TRS to be broken by acquired magnetic order (3). A remarkable example is the quantum anomalous Hall (QAH) effect, a zero-magnetic-field quantum Hall effect that had been sought after for over two decades until it was observed in a magnetic TI with ferromagnetic (FM) order induced by magnetic dopants (3-6). The experimental realization of the QAH effect paved the road for hunting many other novel quantum effects in TRS-broken TIs, for example topological magnetoelectric (TME) effects and chiral Majorana modes (3, 8, 9). However, magnetically doped TIs are notorious “dirty” materials for experimental studies: the randomly distributed magnetic impurities induce strong inhomogeneity in the electronic structure and magnetic properties, and the sample quality is sensitive to the details of the molecular beam epitaxy (MBE) growth conditions (10-12). Such a complicated system is often a nightmare for some delicate experiments such as those on chiral Majorana modes and topological quantum computation, and the strong inhomogeneity is believed to contribute to the extremely low temperature (usually <100 mK) required by the QAH effect (13). An ideal magnetic TI is an intrinsic one, namely a stoichiometric compound with orderly arranged and exchange-coupled magnetic atoms which features a magnetically ordered ground state, but becomes a TI when the TRS recovers above the magnetic ordering temperature. A thin film of such an intrinsic magnetic TI could be a

congenital QAH insulator with homogeneous electronic and magnetic properties, and presumably higher QAH working temperature. Yet few experimental progresses were achieved in this direction in spite of several interesting theoretical proposals raised in the past years (*14-16*).

Some stoichiometric ternary tetradymite compounds, which can be considered as variants of well-studied Bi_2Te_3 family 3D TIs, have been found to be also 3D TIs (*17*). A simplest system is XB_2T_4 where X is Pb, Sn or Ge, B is Bi or Sb, and T is Te or Se. Such a compound is a layered material with each septuple-layer (SL) composed of single atomic sheets stacking in the sequence T-B-T-X-T-B-T. If X were a magnetic element, there would be chance that XB_2T_4 is an intrinsic magnetic TI. A few works observed $\text{MnBi}_2\text{Te}(\text{Se})_4$ in multi-crystalline samples, or as the second phase or surface layer of $\text{Bi}_2\text{Te}(\text{Se})_3$, without figuring out their topological electronic properties (*18-20*). Interestingly, a SL of $\text{MnBi}_2\text{Te}(\text{Se})_4$ on $\text{Bi}_2\text{Te}(\text{Se})_3$ was reported to be able to open a large magnetic gap at the topological surface states of the latter (*20, 21*).

In this study, we found that high-quality MnBi_2Te_4 films can be fabricated in a SL-by-SL manner by alternate growth of 1 quintuple-layer (QL) of Bi_2Te_3 and 1 bilayer (BL) of MnTe with MBE. Amazingly, MnBi_2Te_4 films with the thickness $d \geq 2$ SL show Dirac-type surface states—a characteristic of a 3D TI. Low temperature magnetic and transport measurements as well as first-principles calculations demonstrate that MnBi_2Te_4 is an intrinsic antiferromagnetic (AFM) TI, composed of ferromagnetic SLs with a perpendicular easy axis which are coupled antiferromagnetically between neighboring SLs. Remarkably, a thin film of such an AFM TI thin film with FM surfaces is expected to be an intrinsic QAH insulator or axion insulator depending on the film thickness.

To prepare a MnBi_2Te_4 film, we first grow a 1 QL Bi_2Te_3 film on a Si(111) or $\text{SrTiO}_3(111)$ substrate (*22, 23*). Mn and Te are then co-evaporated onto Bi_2Te_3 surface with the coverage corresponding to a MnTe BL with the sample kept at 200°C. Post-annealing at the same temperature for 10 minutes is carried out to improve the crystalline quality. This leads to the formation of a SL of MnBi_2Te_4 (see the schematic

in Fig. 1A) (20), as experimentally proved and theoretically explained below. Then on the MnBi₂Te₄ surface, we grow another QL of Bi₂Te₃ which is followed by deposition of another BL of MnTe and post-annealing. By repeating this procedure, we can grow a MnBi₂Te₄ film SL by SL in a controlled way, in principle up to any desired thickness.

The MnBi₂Te₄ film shows sharp 1×1 reflection high-energy electron diffraction streaks (Fig. S1) indicating its flat surface morphology and high crystalline quality. The X-ray diffraction (XRD) pattern (Fig. 1B, taken from a 7 SL MnBi₂Te₄ film) exhibits a series of peaks (marked by blue arrows), most of which can neither be attributed to Bi₂Te₃ nor to MnTe. From the positions of these XRD peaks, we can estimate the spacing between the crystalline planes to be ~1.36 nm, very close to the inter-SL distance of bulk MnBi₂Te₄ (1.356 nm) predicted by our first-principles calculations.

High resolution scanning transmission electron microscopy (STEM) was used to characterize the real-space crystalline structure of a MnBi₂Te₄ film (5 SL). The high-angle annular dark field (HAADF) images (Figs. 1C and 1D) clearly show the characteristic SL structure of XB₂T₄ compounds, except for the region near the substrate where stack faults and dislocations are observed. Figure 1E displays the intensity profile along an atomic row across two SLs (cut 1 in Fig. 1C). One can see the atomic contrast varies a lot at different positions in a SL. The contrast of an atom in a HAADF-STEM image is directly related to its atomic number. The intensity distribution along a SL is thus well consistent with the Te-Bi-Te-Mn-Te-Bi-Te sequence. The electron energy loss spectroscopy (EELS) (Fig. 1F) reveals the Mn L_{2,3} edges at ~645 eV. The intensity distribution curve of EELS at 645 eV (the pink line in Fig. 1F) taken along cut 2 in Fig. 1C shows a peak at the middle atom of each SL, which also agrees with the MnBi₂Te₄ structure.

In-situ angle-resolved photoemission spectroscopy (ARPES) was used to map the electronic energy band structure of the MBE-grown MnBi₂Te₄ films. Figures 2A-2D show the ARPES bandmaps of the MnBi₂Te₄ films with the thickness $d = 1, 2, 5$, and

7 SL, respectively, with the sample temperature at ~ 25 K. The spectra were taken around Γ point along the M- Γ -M direction of the Brillouin zone. The spectra of the $d = 1$ SL sample (Fig. 2A) shows a bandgap with Fermi level cutting the conduction band. The films with $d \geq 2$ SL all show similar band structures (Figs. 2B-2D). One can always observe a pair of energy bands with nearly linear band dispersion crossing at Γ point forming a Dirac cone. Figures 2E and 2F show the momentum distribution curves (MDCs) and the constant-energy contours of the 7 SL sample, respectively, which exhibits an archetypal Dirac-type energy bands. It is worth to note that the Dirac-type bands are quite different from the topological surface states of Bi_2Te_3 (24, 25). The band dispersion observed here is rather isotropic, as shown by the nearly circular constant-energy contours even at the energy far away from the Dirac point, which is distinct from the strongly warped Bi_2Te_3 topological surface states (25, 26). The Dirac point observed here is located right in the band gap, in contrast with the Bi_2Te_3 case where the Dirac point is below the valance band maximum. Moreover, the Fermi velocity near Dirac point is $5.5 \pm 0.5 \times 10^5$ m/s, obviously larger than that of Bi_2Te_3 surface states ($3.87 \sim 4.05 \times 10^5$ m/s in different directions) (25). Therefore the Dirac-type bands can only be attributed to MnBi_2Te_4 , and, as demonstrated below, are also the topological surface states of a 3D TI.

The orderly and compactly arranged Mn atoms in MnBi_2Te_4 are expected to give rise to a long-range magnetic order at low temperature. Figure 3A displays the magnetization (M) — magnetic field (H) curves of a 7 SL MnBi_2Te_4 film measured with superconducting quantum interference device (SQUID) at different temperatures (T_s). The linear diamagnetic background contributed by the substrate and capping layer has been subtracted (the raw data are shown in Fig. S2). The unit of M is the magnetic moment (μ_B) per in-plane unit cell (2D U.C.), i.e. the average magnetic moment of each Mn atom multiplied by the number of SLs. H is applied perpendicular to the sample plane. With decreasing temperature, hysteresis appears in the M - H curves and grows rapidly, exhibiting a typical FM behavior. The Curie temperature (T_C) is 20 K according to the temperature (T) dependence of the remnant

magnetization [$M_r = M(0 \text{ T})$] shown in Fig. 3B. The M - H curve measured with in-plane magnetic field has much smaller hysteresis than the curve measured with perpendicular one (see Fig. 3A inset, which were taken from another 7 SL MnBi₂Te₄ sample). Therefore the magnetic easy axis is along the c direction [perpendicular to the (001) plane]. Estimated from the saturation magnetization $M_s = 8 \mu_B/2D \text{ U.C.}$, the Mn atomic magnetic moment is about $1.14 \mu_B$ which is much smaller than $5 \mu_B$ expected for Mn²⁺ ions. It suggests that Mn²⁺ ions in the material may have a more complex magnetic structure than a simple uniform ferromagnetic configuration.

The ferromagnetism of 7 SL MnBi₂Te₄ film is also demonstrated by Hall measurements. Figure 3D displays the Hall resistance (R_{yx}) vs. H curves of a 7 SL film grown on SrTiO₃(111) substrate measured at 1.6 K under different gate-voltages (V_{gs}). The SrTiO₃ substrate is used as the gate dielectric for its huge dielectric constant (~20000) at low temperature (27). The curves exhibit hysteresis loops of the anomalous Hall effect (AHE) with a linear background contributed by the ordinary Hall effect (OHE). The slope of the OHE background reveals that the sample is electron-doped with the electron density $n_e \sim 1.1 \times 10^{13} \text{ cm}^{-2}$, which basically agrees with $n_e \sim 8 \times 10^{12} \text{ cm}^{-2}$ derived from the Fermi wavevector ($k_F \sim 0.07 \text{\AA}^{-1}$) of the ARPES-measured Dirac-type band. The hysteresis loops of the AHE confirm the ferromagnetism of the film with perpendicular magnetic anisotropy. The T_C obtained from the R_{yx} - T curve is similar to that given by SQUID data (Fig. 2B). The H_c of the R_{yx} - H hysteresis loops is however larger than that of the M - H loops. Tuning the chemical potential of the film by applying different V_{gs} , we observe obvious change in the anomalous Hall resistance. The sensitivity of the AHE to the chemical potential suggests that the AHE is mainly contributed by the Berry curvature of the energy bands induced by intrinsic magnetism of the material instead of magnetic impurities or clusters (28).

Noticeably, 6 SL MnBi₂Te₄ film shows different magnetic properties from 7 SL one. As shown in Fig. 3C, the hysteresis (M_r and H_c) in the M - H curve of a 6 SL film is rather small even at 3 K, and M_s decreases slowly with increasing temperature.

Clearly the film is not dominated by long-range FM order. The M - H curves of the MnBi_2Te_4 films from 4 SL to 9 SL are displayed in Fig. 3E which will be analyzed below based on our theoretical results.

Next we discuss the structure, magnetism and topological electronic properties of MnBi_2Te_4 with the above experimental observations and our first-principles calculation results. To understand the mechanism for the formation of MnBi_2Te_4 , we calculated the energies of a MnTe BL adsorbed on a Bi_2Te_3 QL (Fig. 4A left) and a MnBi_2Te_4 SL (Fig. 4A right). The calculations show that the latter one has 0.51 eV/unit lower total energy and is thus energetically more stable. The result is easy to understand in terms of valence states. By assuming Te^{2-} , the former structure gives unstable valence states of Mn^{3+} and Bi^{2+} which tend to change into more stable Mn^{2+} and Bi^{3+} by swapping their positions. The atom-swapping induced stabilization thus explains the spontaneous formation of MnBi_2Te_4 with a MnTe BL grown on Bi_2Te_3 .

We calculated the energies of different magnetic configurations of MnBi_2Te_4 (Fig. S3) (23). It was found that the most stable magnetic structure is FM coupling in each SL and AFM coupling between adjacent SLs (i.e. A -type AFM), whose easy axis is out-of-plane (Fig. 4B). In MnBi_2Te_4 , Mn atoms are located at the center of slightly distorted octahedrons that are formed by neighboring Te atoms. The FM intralayer coupling induced by Mn-Te-Mn superexchange interactions is significantly stronger than the AFM interlayer coupling built by weaker Mn-Te \cdots Te-Mn super-superexchange interactions. Similar A -type AFM states were predicted to exist in other magnetic XB_2T_4 compounds (29).

Figure 4C shows the calculated band structure of a 7 SL MnBi_2Te_4 film. We can observe Dirac-like energy bands around Γ point, which basically agrees with the ARPES data, except for a gap (~ 52 meV) at the Dirac point. All the films above 4 SL show similar band feature with nearly identical gap values at the Dirac point, implying that the gapped Dirac cone is an intrinsic surface feature of the material. Purposely tuning down the SOC strength in calculations, the gap first decreases to zero and then increases (inset of Fig. 4C), which suggests a topological phase

transition and thus the topologically non-trivial nature of the gap. Actually our calculations on the system reveal that bulk MnBi₂Te₄ is a 3D AFM TI with Dirac-like surface states that are gapped by the FM (001) surfaces with out-of-plane magnetization (29, 30).

As illustrated in Fig. 4D and confirmed numerically, the gapped surface states can be described by an effective Hamiltonian $H(\mathbf{k}) = (\sigma_x k_y - \sigma_y k_x) + m_z \sigma_z$, where σ is the Pauli matrix with $\sigma_z = \pm 1$ referring to spin up and down, m_z is the surface exchange field (2, 3). For films thicker than 1 SL, hybridizations between top and bottom surfaces are negligible. Thus, their topological electronic properties are determined by the two isolated surfaces, which have the same (opposite) m_z for odd (even) number of SLs and half-integer quantized Hall conductance of $e^2/2h$ or $-e^2/2h$ depending on the sign of m_z . Therefore, odd-SL MnBi₂Te₄ films are intrinsic QAH insulators with Chern number $C = 1$; meanwhile even-SL films are intrinsic axion insulators ($C = 0$) that behave like ordinary insulators in dc measurements but can show topological magnetoelectric effects in ac measurements (3). However, when the TRS is recovered above T_C , the exchange splitting of the bands gets vanished while the SOC-induced topological band inversion remains unaffected. MnBi₂Te₄ thus becomes a 3D TI showing gapless topological surface states which are exactly the band structure observed in the ARPES measurements performed at 25 K (above T_C).

The theoretically predicted magnetic configuration of MnBi₂Te₄ (Fig. 4B) is supported by our magnetic measurements. For an odd-SL AFM MnBi₂Te₄ film, whatever the exact thickness, the net magnetic moment is only of 1 SL. It explains why the atomic magnetic moment of Mn estimated from the 7 SL MnBi₂Te₄ film (1.14 μ_B) is much smaller than 5 μ_B . The measured $M_s = 8 \mu_B$ per 2D U.C. may have contributions from both the FM surfaces (supposed to be 5 μ_B) and the AFM bulk which can give magnetic signals via canting or disorder. With the AFM arrangement of neighboring FM SLs, MnBi₂Te₄ films are expected to show oscillation in its magnetic properties as the thickness changes between even and odd SLs. We indeed observed even-odd oscillation in their magnetic properties as shown in Figs. 3E and

3F. The remnant magnetization (M_r), which characterizes long range ferromagnetic order, is larger in odd-SL films than in even-SL ones. H_c shows similar oscillation below 7 SL, but increases monotonously in thicker films. It is because in an AFM film with FM surfaces, the Zeeman energy in magnetic field (E_z) is only contributed by the FM surfaces and thus invariant with film thickness, while the magnetocrystalline anisotropy energy (E_{MCA}), which is contributed by the whole film, increases with thickness and thus becomes more difficult to be overcome by E_z . Besides, as shown in the 6 SL film (Fig. 3C) and other even-SL films, M_s is less sensitive to temperature than in odd-SL films. For a comparison, the differences between the M - H curves measured at 3 K and those measured above T_C are displayed in the bottom column of Fig. 3E, which shows a clear even-odd oscillation (Fig. 3F). A rapid increase of M_s with decreasing temperature below T_C is typical of ferromagnetic order. The magnetic signal from AFM canting, on the other hand, decreases or keeps nearly constant with decreasing temperature. So the odd-SL films obviously have more FM features.

The large inter-SL distance (~ 1.36 nm) is expected to give a weak AFM coupling between neighboring SLs which can be aligned into FM configuration in a magnetic field of several tesla (31). We carried out a Hall measurement of a 7 SL MnBi₂Te₄ film with H up to 9 T. As shown in Fig. 3G (the linear background of the OHE has been subtracted from the R_{yx} - H loop), besides a small hysteresis loop at low field contributed by the FM surfaces, R_{yx} resumes growing above ~ 2 T and is saturated at a higher plateau above 5 T. The phenomenon is typical of a layered magnetic material and presumably results from an AFM-to-FM transition (see the schematic magnetic configuration shown by the blue arrows in Fig. 3G). The FM configuration may drive the system into a magnetic Weyl semimetal phase (29, 30).

In spite of the above evidences for an *A*-type AFM order of MnBi₂Te₄, there are still some observations which we have not yet fully understood. For example, the even-SL films show larger M_s than odd-SL ones above T_C , which is particularly clear in comparing the 6 SL (Fig. 3C) and 7 SL (Fig. 3A) data at 30 K. We also notice that overall M_s shows a maximum around 6 SL and 7 SL at 3 K, regardless of even- or

odd-SLs. Another confusion is that the magnetic properties revealed by Hall effect measurements are not fully consistent with those revealed by magnetization measurements: R_{yx} - H loops always show larger H_c than M - H loops, and oscillatory behaviors are barely observed in the AHE data of the films of different thicknesses. These phenomena should result from the interplays between the complex magnetic structures and topological electronic properties of the unique layered magnetic material and require a comprehensive study combining various techniques to clarify (31, 32). Besides, we found that MnBi₂Te₄ films are relatively easy to decay at ambient condition: M_s of a sample decreases significantly after it is exposed in air for couple of days. This may also complicate the magnetization and magneto-transport measurement results. Finding an effective way to protect the material is crucial for the experimental investigations on this system and for the explorations of the exotic topological quantum effects in it.

Acknowledgments: The authors thank Wanjun Jiang and Jing Wang for stimulating discussions. We are grateful to the National Science Foundation of China, Ministry of Science and Technology of China, and the Beijing Advanced Innovation Center for Future Chip (ICFC) for financial support.

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Figures Captions

Fig. 1. MBE growth and structural characterizations of MnBi₂Te₄ films. (A) Schematic illustrations of the MBE growth mechanism of 1 septuple layer (SL) MnBi₂Te₄ thin film. (B) XRD pattern of a MnBi₂Te₄ (MBT) film grown on Si(111). (C) Cross-sectional HAADF-STEM image of a 5 SL MnBi₂Te₄ film grown on a Si (111) substrate. (D) Zoom-in view of (C) with the structural model of MnBi₂Te₄. (E) Intensity distribution of HAADF-STEM along cut 1 in (C). (F) EELS spectra mapping along cut 2 in (C). The pink curve shows the intensity distribution of the Mn L_{2,3}-edge along cut 2 in (C).

Fig. 2. Energy band structures of MnBi₂Te₄ films measured by ARPES. (A-C) ARPES spectra of 1, 2, 5, and 7 SL MnBi₂Te₄ films measured near the Γ point, along the M- Γ -M direction. (D) Momentum distribution curves (MDCs) of the 7 SL film from E_F to -0.38 eV. The red triangles indicate the peak positions. (E) Constant energy contours of the 7 SL film at different energies. All the ARPES data were taken at 25 K.

Fig. 3. Magnetic and magneto-transport properties of MnBi₂Te₄ films. (A) Magnetization vs. magnetic field (M - H) curves of a 7 SL MnBi₂Te₄ film measured with SQUID at 3 K (red), 10 K (orange), 15 K (green), and 30 K (blue), respectively. H is perpendicular to the sample plane. The inset shows M - H curves measured with H perpendicular to (red) and in (blue) the sample plane (a different 7 SL MnBi₂Te₄ sample). (B) Temperature dependences of the remnant magnetization (M_r) and zero magnetic field Hall resistance (R_{yx}^0) of a 7 SL film, which give the Curie temperature (T_C). (C) M - H curves of a 6 SL MnBi₂Te₄ film measured with SQUID at 3 K (red), 10 K (orange), 15 K (green), and 30 K (blue), respectively. H is perpendicular to the sample plane. (D) R_{yx} - H curves measured at 1.6 K at different gate voltages. (E) M - H curves of 4, 5, 6, 7, 8, and 9 SL MnBi₂Te₄ films measured at 3 K and right above T_C (top column) and the differences between the curves at the two temperatures (bottom column).

column). **(F)** Thickness dependences of M_r at 3 K, M_r difference at 3 K and above T_c (top panel) and H_C (bottom panel). **(G)** R_{yx} - H curve of a 7 SL MnBi₂Te₄ film measured at 1.6 K with H up to 9 T. The blue arrows indicate the magnetic configurations at different H . Each arrow represents the magnetization vector of a SL.

Fig. 4. First-principles calculation results of MnBi₂Te₄. **(A)** Lattice structures of a MnTe bilayer adsorbed on a Bi₂Te₃ quintuple layer (left) and a MnBi₂Te₄ SL (right). Valence states of atoms were labelled by assuming -2 for Te. Atom swapping between Mn and Bi results in stable valence states, thus stabilizing the whole structure. **(B)** Atomic structure of layered MnBi₂Te₄, whose magnetic states are ferromagnetic within each SL and antiferromagnetic between adjacent SLs. Insets show Te-formed octahedrons together with center Mn. **(C)** Band structure of a 7-SL MnBi₂Te₄ film, which is an intrinsic QAH insulator (band gap \sim 52 meV), as proved the dependence of band gap on the strength of SOC (inset). **(D)** Schematic band structure of MnBi₂Te₄ (001) surface states, showing a gapped Dirac cone with spin-momentum locking. The energy gap is opened by the surface exchange field (m_z), which gets vanished when paramagnetic states are formed at high temperatures.

Figure 1

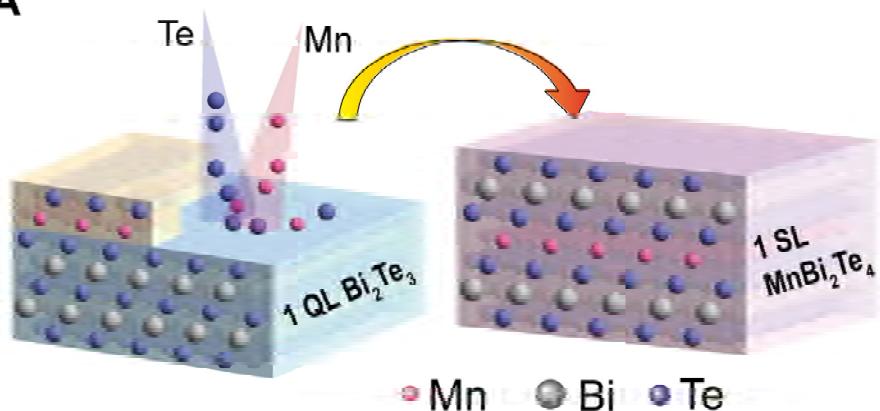
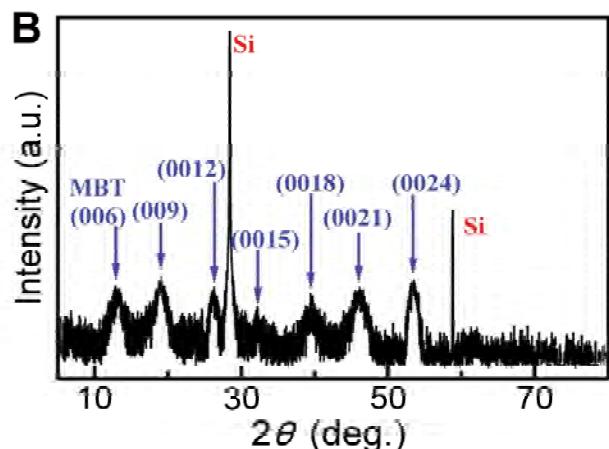
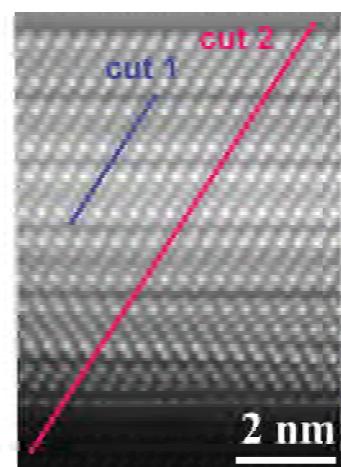
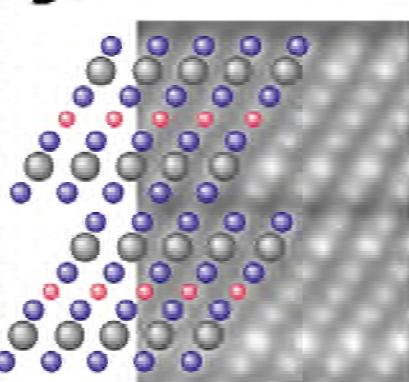
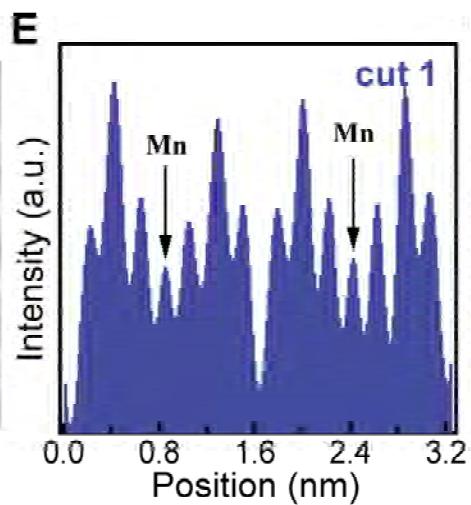
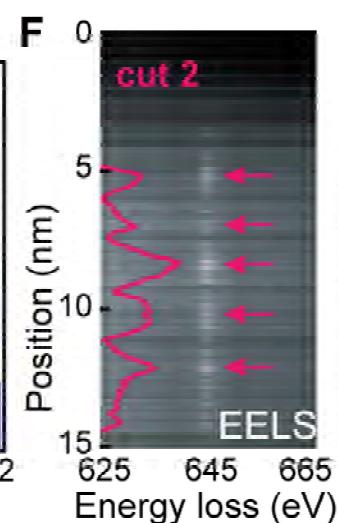
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Figure 2

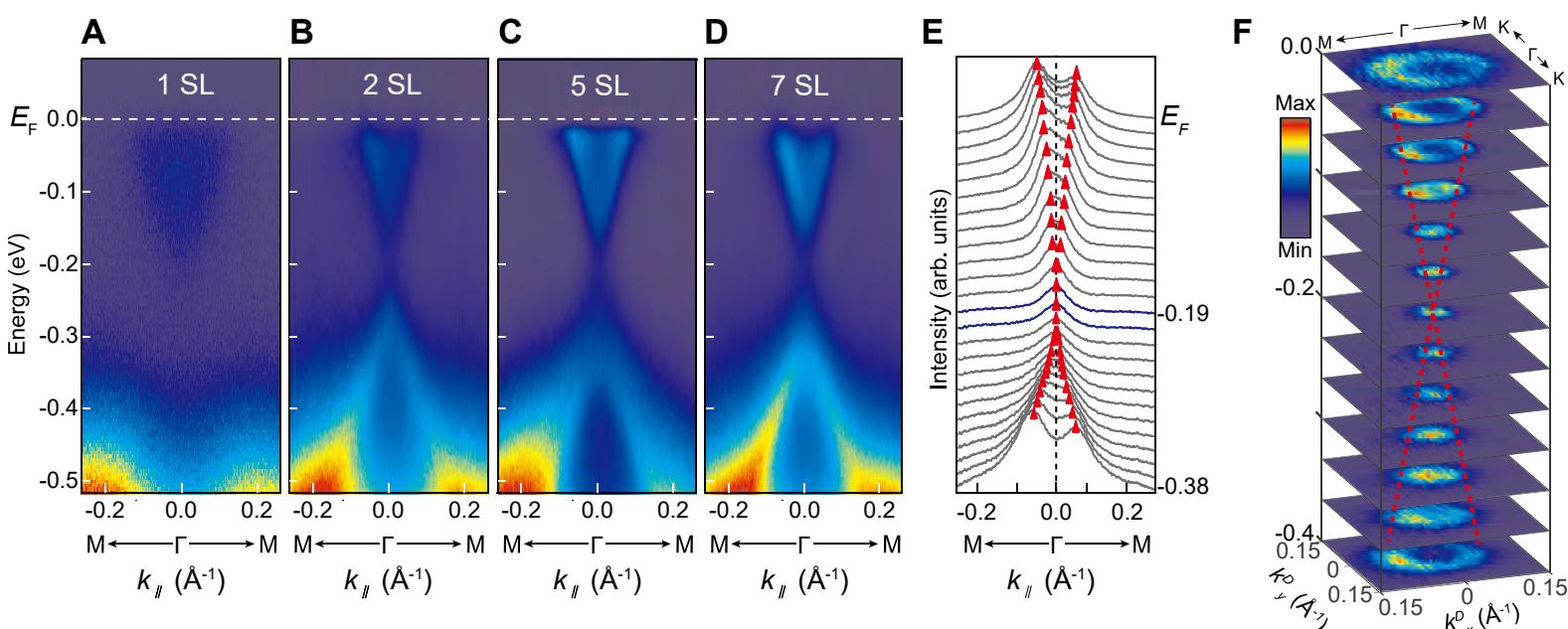


Figure 3

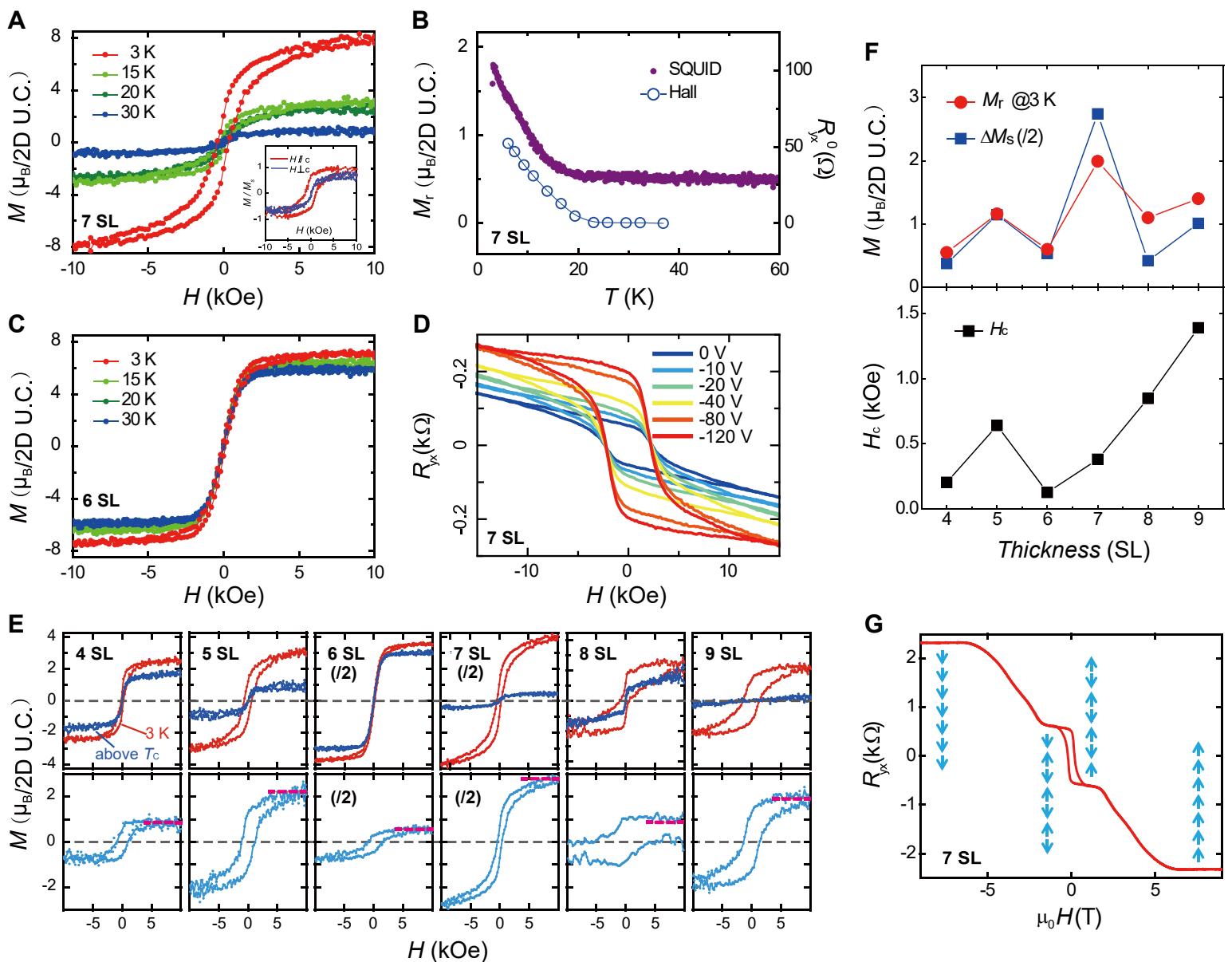


Figure 4

