I. INTRODUCTION

A central theme of contemporary condensed matter research explores the notion of topology and symmetry to generate novel quantum phenomena and device concepts. In this work, we show that under certain synthetic conditions, a van der Waals single-crystalline compound MnSb1.8Bi0.2Te4 exhibits a net ferromagnetic state with a Curie temperature of 26 K, in contrast to the fully compensated antiferromagnetic order observed previously for other members of the Mn(Sb,Bi)2Te4 family. We employ magneto-transport, bulk magnetization, x-ray and neutron scattering studies to illustrate the structural, magnetic, and electrical properties of MnSb1.8Bi0.2Te4. Our structural analyses reveal considerable Mn-Sb site mixing and suggest a recently proposed mechanism, where Mn occupying the Sb site mediates a ferromagnetic coupling between Mn layers [Murakami et al., Phys. Rev. B 100, 195103 (2019)], could be at play. Close comparisons made to an antiferromagnetic compound MnSb2Te4 illustrate the subtle magnetic interactions of the system and the important role played by local chemistry. The appearance of an unusual anomalous Hall effect in MnSb1.8Bi0.2Te4 at low temperatures hints at a magnetic ground state different from other members of this family. Our results are an important step in the synthesis and understanding of magnetism in materials with topological characteristics.

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Recent research activities have identified Mn(Bi,Sb)2Te4, a vdW magnetic family with strong spin-orbit coupling, to be promising candidates in realizing the QAHE above dilution refrigerator temperatures [21–32]. Mn(Bi,Sb)2Te4 can be regarded as consisting of a Mn-Te layer inserted into the quintuple layer of (Sb,Bi)2Te4 [Fig. 1(a)]. Here moments carried by an ordered Mn layer can create an internal magnetic field without introducing random disorder. In the most studied compound MnBi2Te4, the interlayer Mn coupling was found to be A-type antiferromagnetic (AFM) [21–23,30,31,33–35]. The AFM ground state was also favored in other compositions of the compound family [31] with the exception of a recent report of ferrimagnetism in polycrystalline MnSb2Te4 [36]. Few-layer devices exfoliated from antiferromagnetic MnBi2Te4 crystals show the QAHE effect [24]. A ferromagnetic bulk can stabilize the effect at higher temperature and open the door to other interesting possibilities such as type-II Weyl semimetals with broken time reversal symmetry [36,37].

In this work, we show that under certain synthetic conditions, single-crystalline MnSb1.8Bi0.2Te4 can be stabilized into a phase with a net ferromagnetic moment and a Curie temperature of TC = 26 K. We present transport, magnetometry, and neutron diffraction measurements that
illustrate the properties of this state, which we denote as the “FM” state. Refinements of x-ray and neutron diffraction data reveal considerable Mn-Sb site mixing, which supports a recent Mn layer to Mn layer ferromagnetic coupling mechanism discussed in Ref. [36]. Interestingly, our sample also exhibits a sizable unconventional anomalous Hall effect that signals the possibility of additional magnetic structure at very low temperatures. Further understanding of the rich magnetic orders the Mn(Bi,Sb)2Te4 family exhibits and their correlation with local chemistry opens up possibilities of engineering magnetic and topological phenomena in this vdW family with potential prospects in device applications.

II. EXPERIMENTAL METHODS

MnSb1.8Bi0.2Te4 single crystals exhibiting the FM state were synthesized using a flux method. A mixture of high-purity Mn powder (99.95%), Bi shot (99.999%), antimony shot (99.999%) and Te ingot (99.999+%) with the molar ratio of Mn:Sb:Bi:Te = 1:9:1:16 was loaded into an Al2O3 crucible and sealed in an evacuated quartz tube. The mixture was heated up to 900°C for 12 h to promote homogeneous melting and then slowly cooled down to 595°C at a rate of 2°C/h and dwelled at this temperature for 24 h. It is then further cooled down to 400°C within 3 h and then immediately heated back to 625°C in 1 h and dwelled at this temperature for another 2 h. We then move the sample quickly from the furnace to a centrifuge to remove the excess flux before letting it cool down in the centrifuge. The treatment of cooling down the melt from 900 to 400°C before going back to 625°C is critical to the synthesis of the FM state in MnSb1.8Bi0.2Te4. Cooling the melt directly to 625°C without this step leads to an AFM ground state similar to prior results [31]. A MnSb1.85Bi0.15Te4 crystal synthesized via this method displays very similar Curie temperature and saturation moment to the MnSb1.8Bi0.2Te4 discussed in the main text (see Fig. S3 of the Supplemental Material (SM) [38]). We also attempted synthesizing MnSb2Te4 crystals using the same method, but obtained only crystals exhibiting the AFM ground state. MnSb2Te4 crystals studied here were prepared using a flux method similar to that used in Ref. [31]. X-ray diffraction (XRD) and scanning transmission electron microscopy measurements confirm the septuple-layer rhombohedral structural phase in both materials. Measurements presented in the main text are obtained on crystals and exfoliated flakes with greater than 95% purity in the primary Mn(Sb,Bi)2Te4 phase (see Figs. S1 and S2 of the Supplemental Material [38]). Powdered polycrystalline samples of both MnSb1.8Bi0.2Te4 and MnSb2Te4 are grown with the same methodologies and are verified to have the same magnetic ground states as their single-crystal counterparts. They are used for neutron powder diffraction (NPD) studies. Rietveld analysis on the NPD was performed using a combination of the FULLPROF and GSAS-II programs (see details in Sec. S1 of the SM [38–41]). Figure 1(a) illustrates a schematic sideview of the layer stacking in the Mn(Bi,Sb)2Te4 family. Micrometer-sized flakes are exfoliated from selected crystals and transferred using a polypropylene carbonate stamp to prepatterned electrodes inside a glovebox filled with argon gas. The finished device is covered with a droplet of Poly(Methyl MethAcrylate) before being transferred to a cryostat. Figure 1(b) shows an optical image of a typical Hall bar device. Flakes of similar color tone measure 100–300 nm in thickness in an atomic force microscope. Transport measurements are performed in a pumped 4He cryostat with a magnetic field up to 9 T using standard low-frequency techniques. Magnetometry measurements are performed in a superconducting quantum interference device magnetometer from 2 to 300 K. Single-crystal elastic neutron scattering measurements are performed using the BT-4 triple-axis spectrometer (TAS) and NPD measurements were made on the BT-1 powder diffractometers at the NIST Center for Neutron Research (NCNR). The BT-1 measurements were made using 60° collimation with both Cu(311) and Ge(311) monochromators. TAS measurements were taken with an instrument configuration of open-pg-pg-40’s-pg-40’-100’ where pg refers to pyrolytic graphite.

III. RESULTS AND DISCUSSION

Figure 1(c) plots the Hall resistance \( R_{xy}(H) \) obtained on a MnSb1.8Bi0.2Te4 device. A small \( R_{xy} \) component is removed from the data though an asymmetric step that averages the upswing of \( R_{xy}(H) \) and the downswing of \(-R_{xy}(-H)\). Measurements were taken at a series of fixed temperatures ranging from 2 to 60 K. Traces plotted here represent the typical behavior in different temperature ranges. As the \( T = 60 \) K trace shows, \( R_{xy}(H) \) is a straight line from \(-9 \) to \( 9 \) T at high temperatures. An anomalous Hall effect starts to develop at \( T < 46 \) K, where the slope \( dR_{xy}/dT \) taken at \( R_{xy} = 0 \) (illustrated by a green dashed line for the \( T = 2 \) K upswipe trace) becomes larger than the slope taken at high field (a black dashed line in the inset). The difference of the two originates from a nonzero magnetization \( M \) of the sample since \( R_{xy}(H) = R_{0}H + R_{m}M \) [42]. At sufficiently high field where \( M \) saturates, the slope \( dR_{xy}/dT \) yields the normal Hall coefficient \( R_{0} = 1/\mu_{e} \). \( R_{0} \) follows a cos \( \theta \) dependence as the external field tilts away from the c axis of the crystal (Fig. S4), which confirms the two-dimensional nature of the mobile carriers and yields a hole carrier density of \( n_{h} = 6.3 \times 10^{13}/\text{cm}^{2} \). This translates to a doping level of \( \sim 10^{15}/\text{cm}^{2} \) per septuple layer and puts the Fermi level in the bulk valence band of MnSb1.8Bi0.2Te4 [31]. \( R_{0} \) is approximately \( T \) independent, as demonstrated in Fig. S4 of the SM [38]. In contrast, the slope \( dR_{xy}/dT \) taken at \( R_{xy} = 0 \), called the low-field slope from now on, increases rapidly with decreasing temperature and reaches a broad maximum around 12–20 K. Its \( T \) dependence is plotted in Fig. 2(a) as magenta squares. Similar measurements are performed on a MnSb2Te4 device and the results are plotted as blue squares.

In addition to the anomalous Hall effect, \( R_{xy}(H) \) becomes hysteretic at temperatures below \( \sim 23 \) K. Data at 2 and \( 15 \) K are plotted to show the two different shapes of the hysteresis loop. Hysteresis is also observed in bulk magnetization measurements of the parent MnSb1.8Bi0.2Te4 crystal. Figure 1(d) plots the \( M(H) \) data at \( T = 2 \) K, from which we extracted a remanent magnetization of \( M_{r} = 0.6 \mu_{B}/\text{Mn} \), a coercive field of \( H_{c} = 310 \) Oe, and a saturated \( M \) of \( 1.8 \mu_{B}/\text{Mn} \) [inset of Fig. 1(d)]. Both the \( H_{c} \) and \( R_{xy} \) decrease with increasing temperature and vanish at \( T > 23 \) K, supporting the establish-
ment of the FM state in this temperature vicinity (see Fig. S6 of the Supplemental Material [38]).

To further explore the magnetic properties of MnSb1.8Bi0.2Te4, we plot in Figs. 2(a) and 2(b) the $T$-dependent magnetic susceptibility $\chi(T)$, extracted from the low-field slope of the Hall resistance $dR_{xy}/dH$ and DC magnetometry measurements, respectively. In a magnetic system, the low-field slope $dR_{xy}/dH$ includes the contribution from the out-of-plane magnetic susceptibility $\chi = dM/dH$. In Fig. 2(a), $dR_{xy}/dH$ ascends rapidly at $T \sim 25$ K, reaches a maximum value of 11 $\Omega$ T around 12–20 K, which is more than 100 times larger than the normal Hall coefficient $R_0 = 0.1 \Omega$ T of this device, before dropping again at lower temperatures. In other words, the low-field slope $dR_{xy}/dH$ is dominated by the magnetic response of the system and effectively measures the $\chi(T)$ of the microscope device. The magnetometry studies conducted on bulk crystals tell a similar story. Figure 2(b) plots the temperature-dependent linear susceptibility $M/H(T)$ obtained under both zero-field-cooling (ZFC) and several field-cooling (FC) conditions using several different fields as labeled in the plot. The 50 Oe ZFC data (solid black line) strongly resemble the low-field slope $dR_{xy}/dH$ shown in Fig. 2(a), suggesting that our samples behave homogeneously from the $\mu$m to the mm length scale. Both support the onset of a FM order at a Curie temperature of $T_C \sim 26$ K obtained by fitting the neutron scattering data below. At $T < 12$ K, both the low-field slope $dR_{xy}/dH$ and the low-field ZFC $M/H$ data show a pronounced drop that deviates from a conventional FM. More complex magnetic phases may emerge in this temperature range [34,43–45]. We aim to understand its nature with additional measurements and analyses [46].

Figures 2(c) and 2(d) compare neutron scattering results obtained on our MnSb1.8Bi0.2Te4 and MnSb2Te4 single crystals. Upon cooling, the $(1 0 1)$ and $(1 0 4)$ nuclear reflection

![Image](image.png)

**FIG. 1.** (a) Schematic stacking order of Mn(Sb,Bi)2Te4. (b) An optical image of a typical MnSb1.8Bi0.2Te4 device in a Hall bar geometry. (c) The Hall resistance $R_{xy}(H)$ on a MnSb1.8Bi0.2Te4 device at selected temperatures as labeled in the plot. Arrows indicate the field-sweep direction. The green dashed line illustrates the low-field slope $dR_{xy}/dH$ taken at $R_{xy} = 0$. The inset shows the full-range down sweep of $R_{xy}(H)$ at 2 K. The black dashed line illustrates the high-field slope $dR_{xy}/dH$. (d) $M(H)$ of a MnSb1.8Bi0.2Te4 crystal. The coercive field $H_c = 310$ Oe. $M(H)$ to 7 T showing a saturated magnetization of $\sim 1.8 \mu_B$/Mn. Inset: $M(H)$ at 7 T showing a saturated magnetization of $\sim 1.8 \mu_B$/Mn. (e) The anomalous Hall component $\Delta R_{xy}(H)$ for the device shown in (b) at selected temperatures. $\Delta R_{xy}(H)$ is obtained by subtracting the normal Hall contribution $R_{H0}$ from the measured $R_{xy}(H)$. The dashed lines illustrate the process of obtaining the saturation field $H_s$ for the $T = 30$ K trace. Fig. S5 of the SM illustrates the determination of $H_s$ at $T < 22.5$ K, when hysteresis is present [38]. Up and down sweeps produce the same $H_s$.

![Image](image.png)

**FIG. 2.** (a) The low-field slope $dR_{xy}/dH$ as a function of temperature in MnSb1.8Bi0.2Te4 (magenta circles) and MnSb2Te4 (blue squares). The dashed lines are a guide to the eye. (b) The temperature-dependent magnetic susceptibility $M/H$ of a MnSb1.8Bi0.2Te4 sample measured at three external fields $H = 50$, 100, and 1000 Oe under both ZFC and FC conditions as labeled in the graph. $H \parallel c$. The molar magnetic susceptibility $\chi_m = MV_0/H$, where $V_0$ is the volume per gram mole. 1 emu/(mol Oe) $= 4 \pi \times 10^{-6}$ m$^3$/mol. (c) The main panel shows the temperature-dependent elastic neutron scattering centered at the $(1 0 1)$ reflection in MnSb1.8Bi0.2Te4 (c) and the $(1 0 2.5)$ reflection in MnSb2Te4 (d). Mean-field fits (solid lines) yield $T_C = 26.3$ K in (c) and $T_C = 19.5$ K in (d). The insets show scans along the $(1 0 L)$ direction at 45 and 45 K with the * symbol marking the $(1 0 1)$ reflection in MnSb1.8Bi0.2Te4 and the $(1 0 2.5)$ reflection in (c) and the $(1 0 2.5)$ reflection in (d). Larger-range scans from $(1 0 4)$ to $(1 0 4)$ on both compounds are shown in Fig. S7 to show a clear increase of the $(1 0 4)$ peak with decreasing temperature and the lack of twinning in the MnSb1.8Bi0.2Te4 samples [38]. Error bars in (c) and (d) represent one standard deviation.
peaks in MnSb_{1.8}Bi_{0.2}Te_4 gained intensity with no peak appearing at the (1 0 2.5) position (Fig. 2c) inset and a larger range scan from (1 0 −4) to (1 0 4) in Fig. S7 of the SM [38]). In contrast, the (1 0 2.5) peak appeared at low temperatures in our MnSb_{2}Te_4 sample while the amplitude of the nuclear reflections remained unchanged (Fig. 2d) inset and a larger range scan from (1 0 −4) to (1 0 4) in Fig. S7 of the SM [38]). The (1 0 2.5) peak is associated with the development of the A-type AFM phase in MnBi_{2}Te_4 in previous reports [30,32]. The neutron data clearly indicate a different magnetic order in our samples, that is, AFM in MnSb_{2}Te_4 and FM in MnSb_{1.8}Bi_{0.2}Te_4. Mean-field fits to the temperature-dependent scattering amplitude at the (1 0 1) and (1 0 2.5) positions yield a Curie temperature of $T_C \sim 26$ K and a Néel temperature of $T_N \sim 20$ K for the MnSb_{1.8}Bi_{0.2}Te_4 and MnSb_{2}Te_4 samples, respectively. Further, we show in Fig. 2a the low-field slope $d R_{xy}/d H$ we obtained on a MnSb_{2}Te_4 device (solid blue squares). It is consistent with an AFM phase with $T_N \sim 19.5$ K, and is in excellent agreement with previous susceptibility measurements of this material [31].

Diverse observations including the FM and AFM order we identified in our MnSb_{1.8}Bi_{0.2}Te_4 and MnSb_{2}Te_4 crystals, respectively, the literature results of mostly AFM order in the majority of the Mn(Bi,Sb)_{2}Te_4 crystals synthesized [21,23,30,31,33], and a very recent report of a ferrimagnetic ground state with $T_C \sim 25$ K in polycrystalline MnSb_{2}Te_4 [36] all together paint a much more nuanced picture than others’ highlight the sensitivity of the magnetic interactions to the sign of $\gamma$. The intrinsic AFM or FM coupling between adjacent Mn layers competes closely in energy in Sb-rich compositions [31,33]. In Ref. [36], the authors noted that mixing between Mn and Sb sites can alter the interlayer Mn-Mn exchange coupling from AFM to FM via a ferrimagnetic configuration that aligns Mn moment occupying the Sb site in the opposite direction. Through the refinement of x-ray and NPD data, we have also found a considerable amount of antisite defects in our MnSb_{1.8}Bi_{0.2}Te_4 and MnSb_{2}Te_4 samples, with approximately 41(1)% and 26(1)% of Mn occupying nominal (Bi,Sb) sites in MnSb_{2}Te_4 and MnSb_{1.8}Bi_{0.2}Te_4, respectively. The inclusion of Bi seems to suppress the presence of the antisite defects, which is consistent with the significantly fewer antisite defects (3%) and the universal AFM ground state found in the end compound MnBi_{2}Te_4 [32]. A full discussion of the local and long-ranged defects observed in our samples can be found in Sec. S1 of the SM [38,47]. In the literature, Mn-doped Bi_{2}Te_3 is known to have a FM ground state [48–50]. Samples studied here are screened by XRD to have less than 5% intergrowth of the (Sb,Bi)_{2}Te_3 phase. In addition, we have explicitly tested the behavior of a flake exfoliated from a crystal with significant secondary Bi_{2}Te_3 intergrowth. The results are presented in Fig. S11 of the SM [38]. This device exhibits a Curie temperature of $\sim 11$ K and its transport and magnetotransport behavior closely resembles that of Mn-doped Bi_{2}Te_3 [48–50], but are very different from that of the MnSb_{1.8}Bi_{0.2}Te_4 and MnSb_{2}Te_4 devices. No excess anomalous Hall signal discussed in Fig. 4 was observed. These results rule out the possibility of an FM state originating solely from the ferromagnetic coupling of Mn occupying the Sb site.

The different magnetic orders exhibited by our samples and others’ highlight the sensitivity of the magnetic interactions of the system to the details of the local defect chemistry. As described in the Methods section, we are able to grow MnSb_{1.8}Bi_{0.2}Te_4 crystals that are either FM or AFM using different thermal treatments before quenching, which could conceivably lead to different local defect chemistry that supports different magnetic orders. Further understanding of this process and the identification of synthesis conditions that lead to FM order in a wide range of alloy compositions will be an important goal of future studies. In the remainder of the paper, we continue to describe the properties of MnSb_{1.8}Bi_{0.2}Te_4 samples that display the FM characteristics, focusing on its transport characteristics and the appearance of an excess anomalous Hall signal at yet lower temperatures of $T < 12$ K.

The sensitivity of $R_{xy}$ to the magnitude of $M$ enables us to determine the saturation field $H_s$ and construct an $H_s$-$T$ phase diagram. To do this we first determine the anomalous Hall signal $\Delta R_{xy}(H) = R_{xy}(H) - R_0(H)$. The results for the device shown in Fig. 1(b) at several temperatures are shown in Fig. 1(e). The saturation field $H_s$ is defined as the field at which the extension of the slope at $R_{xy} = 0$ reaches the saturated value of $\Delta R_{xy}$, as illustrated by the dashed lines for the $T = 30$ K trace. Figure S5 of the SM illustrates the process of determining $H_s$ at low temperature when hysteresis is present [38]. Figure 3(a) plots the resulting $H_s$-$T$ diagram. $H_s$ reaches a minimum of 0.030 T near $T_C$. The onset of a hysteresis loop leads to a small increase of $H_s$ with decreasing

FIG. 3. (a) Moment saturation field $H_s$ vs $T$ in MnSb_{1.8}Bi_{0.2}Te_4 obtained from $\Delta R_{xy}(H)$ data shown in Fig. 1(e) and Fig. S5. Solid symbols are data below $T_C$. Open symbols are data above $T_C$ and use the right axis. (b) Lower panel: Temperature-dependent magnetoresistance $R_{xy}(T)$ taken at fixed magnetic field as labeled in the plot. The black dashed lines divide the curves into three regions according to the sign of $dR/dT$. The boundary points are plotted in the upper panel of (b). The symbols follow the notation of (a). (c) Normalized magnetoresistance $MR = [R_{xy}(H) - R_{xy}(0)]/R_{xy}(0) \times 100$% at selected temperatures. Arrows indicate field-sweep direction. Note the change of the y scale in different panels.
FIG. 4. The excess anomalous Hall effect in MnSb$_{1.8}$Bi$_{0.2}$Te$_4$. (a) $R_{xy}(H)$, which is the same as $\Delta R_\chi(H)$ in Fig. 1(e) at $T = 2$ K and with a tilt angle $\theta = 56^\circ$ as illustrated in the inset. The magenta dashed line is a Langevin fit to the anomalous Hall effect with an uncertainty of 0.9 mOe. The area shaded in green indicates the excess Hall effect contribution $R_{xy}^e(H)$ at $\theta = 0^\circ$ and $T = 2$, 5, and 10 K. Data obtained from up and down sweeps are shifted horizontally to coincide at $H = 0$. (c) $R_{xy}^e(H)$ at $T = 2$ K and selected tilt angle $\theta$. $R_{xy}^e(\theta)$ peak is marked by the * symbol. (d) The angular dependence of $\Delta R_{xy}^e$ at $T = 2$ K. The right axis labels the effective magnetic field $H_{\text{eff}} = R_{xy}^e/\Omega_1$.

temperature at $T < T_C$. Remarkably, at low temperature, $H_a$ is only 0.084 T in MnSb$_{1.8}$Bi$_{0.2}$Te$_4$, in comparison to 0.42 T in MnSb$_{2}$Te$_4$ (Fig. S9 of the SM [38]) and more than 7 T in MnBi$_{2}$Te$_4$ (Fig. S10 of the SM [38]), despite similar ordering temperatures of ~20 K in all three materials. This observation strongly attests to the FM order in MnSb$_{1.8}$Bi$_{0.2}$Te$_4$. The small $H_a$ here is associated with the alignment of the FM domains in an external field, rather than the spin-flop transition of individual Mn moment. In addition, we see that the anomalous Hall effect extends into the paramagnetic phase [open circles in Fig. 3(a)], indicating FM fluctuations are already important at $T \geq T_C$.

Next, we demonstrate the impact of magnetic order on the transport characteristics of MnSb$_{1.8}$Bi$_{0.2}$Te$_4$. Figure 3(b) plots $R_{xy}(T)$ traces taken at a series of fixed magnetic fields. We track the sign change of $dR_{xy}/dT$ as a function of $T$ and $H$ and plot the results on an $H$-$T$ map, similar to the $H$-$T$ diagram shown in Fig. 3(a). At temperatures above ~50 K, $R_{xy}(T)$ exhibits the expected metallic $T$ dependence, i.e., $dR_{xy}/dT > 0$ with no dependence on the magnetic field. An expanded $R_{xy}(T)$ from 2 to 100 K is given in Fig. S8 of the SM [38]. As $T$ approaches $T_C$, strong spin fluctuations lead to a slightly insulating $T$ dependence, i.e., $dR_{xy}/dT < 0$, similar to the situation in MnBi$_{2}$Te$_4$ [30]. A positive $dR_{xy}/dT$ is found again when the moments align spontaneously or under a sufficiently large external field, likely due to the reduction of magnetic scatterings that involve a spin-flip/flop. The onset of another insulator-like regime at $T < 12$ K coincides with the drop of $\chi$ in Fig. 2(a), and the onset of an excess anomalous Hall signal in Fig. 4. We are working to understand its origin.

Figure 3(c) plots the normalized magnetoresistance (MR) of a MnSb$_{1.8}$Bi$_{0.2}$Te$_4$ device at selected temperatures. In MnSb$_{1.8}$Bi$_{0.2}$Te$_4$, MR is always negative. Its magnitude increases with decreasing temperature and becomes hysteretic with the onset of the FM order. Alignment of all moments in an external field results in a large reduction of nearly 20% at low temperature. Comparing to similar measurements on MnSb$_{2}$Te$_4$ and MnBi$_{2}$Te$_4$ (Figs. S9 and S10 of the SM [38]), we see that electrical transport in Sb-rich compositions is much more influenced by the magnetic order, likely because the Mn orbitals are located in the valence band and couple more closely to the hole carriers in MnSb$_{1.8}$Bi$_{0.2}$Te$_4$ and MnSb$_{2}$Te$_4$ [31,33].

Finally, we report the appearance of an excess anomalous Hall signal in MnSb$_{1.8}$Bi$_{0.2}$Te$_4$ that is beyond the conventional AHE. This signal concentrates in the circled areas in the $\Delta R_{xy}(H)$ plot shown in Fig. 1(e). Following the literature [11,13,14], we fit the conventional AHE component $R_{\chi}^A$ with a Langevin function [magenta dashed line in Fig. 4(a)] and use $R_{\chi}^T$ to denote the excess signal (green shaded area). Figures 4(b) and 4(c) plot $R_{\chi}^T(H)$ obtained at several temperatures and different tilt angles of the external field, respectively. The raw $\Delta R_{xy}(H)$ plots are given in Fig. S12 of the SM [38]. The magnitude of $R_{xy}(H)$ decreases rapidly with increasing temperature and approaches the fitting uncertainty (~0.9 mOe) at $T > 10$ K. The signal peaks at $H \sim +0.7$ T and persists to several tesla. Figure 4(d) plots the angle dependence of the peak value $R_{\chi}^T(\theta)$ at $T = 2$ K. $R_{\chi}^T(\theta)$ is nonmonotonic and reaches a maximum of 0.1 Ω around $\theta = 60^\circ$. The value corresponds to a fictitious field of $H_{\text{eff}} = R_{\chi}^T/\Omega_1 \sim 1$ T; i.e., the excess anomalous Hall signal in our MnSb$_{1.8}$Bi$_{0.2}$Te$_4$ sample has considerable strength.

This intriguing excess anomalous Hall signal points to excess Berry curvature of the material, which disappears with increasing magnetic field. The room-temperature crystal structure of Mn(Sb,Bi)$_2$Te$_4$ is centrosymmetric, which does not allow for a Dzyaloshinskii-Moriya interaction (DMI) term in the Hall effect [4–9]. We have not identified a clear symmetry-breaking structural transition at low temperatures though this possibility cannot be ruled out. Excess Berry curvature can also occur in systems with noncollinear, frustrated, or spatially modulated magnetic textures such as clustered spin glass, frustrated magnets, or noncollinear AFMs [51–53]. Our refinement analysis has revealed considerable Mn-Sb site mixing. The second magnetic sublattice produced by Mn occupying the Sb sites could conceivably play a role in generating a noncollinear magnetic structure. Our ongoing work seeks to further examine this possibility.

IV. CONCLUSION

In summary, we combine electrical transport, bulk magnetometry, and neutron diffraction studies to show evidence of a ferromagnetic ground state with a Curie temperature of 26 K in MnSb$_{1.8}$Bi$_{0.2}$Te$_4$. Our work is an encouraging step towards realizing a ferromagnetic topological insulator. Its vdW geometry opens up possibilities of forming heterostructures and gate tuning. Studies that illuminate the synthesis conditions of
different magnetic phases in the Mn(Sb,Bi)$_2$Te$_4$ family will greatly facilitate future explorations of their topological and magnetic properties.

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