

Evidence for a Magnetic-Field-Induced Ideal Type-II Weyl State in Antiferromagnetic Topological Insulator $\text{Mn}(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_4$

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The discovery of Weyl semimetals (WSMs) has fueled tremendous interest in condensed matter physics. The realization of WSMs requires the breaking of either inversion symmetry (IS) or time-reversal symmetry (TRS). WSMs can be categorized into type-I and type-II WSMs, which are characterized by untilted and strongly tilted Weyl cones, respectively. Type-I WSMs with breaking of either IS or TRS and type-II WSMs with solely broken IS have been realized experimentally, but a TRS-breaking type-II WSM still remains elusive. In this article, we report transport evidence for a TRS-breaking type-II WSM observed in the intrinsic antiferromagnetic topological insulator $\text{Mn}(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_4$ under magnetic fields. This state is manifested by the electronic structure transition caused by the spin-flop transition. The transition results in an intrinsic anomalous Hall effect and negative *c*-axis longitudinal magnetoresistance attributable to the chiral anomaly in the ferromagnetic phases of lightly hole-doped samples. Our results establish a promising platform for exploring the underlying physics of the long-sought, ideal TRS-breaking type-II WSM.

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I. INTRODUCTION

Weyl semimetals (WSMs) have become a forefront research topic in contemporary condensed matter physics [1–3]. They provide model platforms for studying concepts in high-energy physics, such as the magnetic chiral anomaly effect. Moreover, they provide a means of realizing potentially useful exotic quantum states of technological relevance, e.g., the quantum anomalous Hall insulator [4]. In WSMs, singly degenerate, linearly dispersed bands cross at Weyl nodes in momentum space; these nodes always appear in pairs with opposite chirality

and can be understood as a monopole and an antimonopole of Berry flux. When the Weyl nodes are located at or near the chemical potential, their diverging Berry curvature can give rise to distinct properties such as a large intrinsic anomalous Hall effect (AHE) and an anomalous Nernst effect if time-reversal symmetry (TRS) is broken [5–8]. In addition, the bulk-edge correspondence principle leads WSMs to possess unique surface Fermi arcs [9–11], which can also generate exotic phenomena such as quantum oscillations arising from unconventional magnetic orbits [12,13] and a bulk quantum Hall effect through the formation of Weyl orbits [14]. Further understanding of Weyl fermion physics requires an *ideal* WSM in which all Weyl nodes should be symmetry related and at the same energy level (at or near the chemical potential), without interference from any other bands [1–3,15]. However, although ideal Weyl states have been long pursued, they have been realized only in bosonic systems (i.e., photonic crystals [16]). An ideal fermionic WSM with distinct exotic properties is still lacking. The Weyl nodes of the WSMs discovered to date are either not close to the chemical

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potential, or not at the same energy level, or interfered with by other bands.

WSMs can be divided into two categories: type-I and type-II WSMs. Type-I WSMs, which feature untilted Weyl cones, can be further categorized into inversion-symmetry-breaking WSMs and TRS-breaking WSMs. The inversion-symmetry-breaking type-I WSM state was first discovered in TaAs-class materials [10,11,17–19], and the TRS-breaking type-I WSM was recently demonstrated in several magnetic materials such as GdPtBi [20], Co₃Sn₂S₂ [5,6,21–24], and Co₂MnGa [25,26]. Unlike type-I WSMs, type-II WSMs are characterized by strongly tilted Weyl cones that violate Lorentz invariance [27]. The inversion-symmetry-breaking type-II WSM was found in several nonmagnetic materials such as (W/Mo)Te₂ [27–30], LaAlGe [31], and TaIrTe₄ [32,33]; however, the TRS-breaking type-II WSM still remains elusive. YbMnBi₂ has been claimed to be a member of this category [34], but it is debated whether it breaks TRS [35]. In this article, we report experimental evidence for a recently predicted, magnetic-field-induced ideal TRS-breaking type-II Weyl state with only one pair of Weyl nodes in MnBi₂Te₄ [36,37].

MnBi₂Te₄ has been demonstrated as the first intrinsic antiferromagnetic (AFM) topological insulator (TI) [36–38]. The combination of spontaneous magnetization and nontrivial band topology in this material allows access to a variety of exotic topological quantum states in its 2D thin layers, including the quantum anomalous Hall insulator [39], the axion insulator [40], and the $C = 2$ Chern

insulator [41]. In addition, MnBi₂Te₄ is also predicted to host a long-sought, ideal type-II TRS-breaking WSM when its AFM order is coerced into FM order by a magnetic field parallel to the c axis [36,37]. Theoretical studies show that the interlayer hybridization, combined with the protection of C_3 rotational symmetry, induces band crossings in the FM phase, resulting in a single pair of strongly tilted, type-II Weyl cones with the Weyl nodes above the chemical potential [36,37]. Here, we provide transport evidence for such an ideal WSM state using magnetotransport measurements in Mn(Bi_{1– x} Sb _{x})₂Te₄. By tuning the Sb:Bi ratio, we observe an electronic structure transition driven by the spin-flop transition. The transition leads to an AHE that we attribute to an intrinsic origin and a large negative c -axis longitudinal magnetoresistance that is consistent with the chiral anomaly. All these results provide strong support for the existence of the ideal Weyl state in the FM phase of this material.

II. RESULTS AND DISCUSSION

Prior studies of Mn(Bi_{1– x} Sb _{x})₂Te₄ [42,43] show that the carrier density is minimized near $x \sim 0.3$, where the carrier type also changes from electron to hole. Although there is only a small difference between the Néel temperatures (T_N) of MnBi₂Te₄ ($T_N = 25$ K) and MnSb₂Te₄ ($T_N = 19$ K) [42,43], Sb substitution for Bi induces striking changes in the magnetic properties of the Bi compound. Undoped MnBi₂Te₄ undergoes a spin-flop transition upon increasing

TABLE I. Information of the Mn(Bi_{1– x} Sb _{x})₂Te₄ samples used in this study, including Sb content x , carrier type, SdH oscillation frequency f_{osc} , and carrier densities estimated from Hall coefficient and f_{osc} . The Hall carrier density is extracted from the linear background of ρ_{yx} in FM phase at 2 or 0.7 K. For the lightly hole-doped samples (S3B and S3C) which exhibit strong nonlinear field dependence in ρ_{yx} due to the coexistence of electron and hole pockets, the Hall carrier densities given in the table represent the difference between the hole and electron density, $n_h - n_e$, estimated from the slope of ρ_{yx} in the high field limit (approximately 40 T), where $\rho_{yx} \approx [B/(n_h - n_e)e]$ (see the text).

Sample label	Sb content, x (measured by EDS)	Carrier type	Carrier density $\times 10^{20} \text{ cm}^{-3}$		
			n_{Hall}	n_{SdH}	f_{osc} (T)
S1A	0	Electron-doped (e)	1.3		
S2A	0.22		0.29	0.12	102
S3A	0.26		0.034	0.07	60
S3B	0.26	Hole-doped (h)	0.038		
S3C	0.26		0.089		
S3F	0.26		0.097 ^a		
S3G	0.26		0.039	0.04	36
S4A	0.39		0.17	0.10	85
S5A	0.79		0.55		
S5B	0.79		0.55 ^a		
S6A	0.52		0.25	0.19	161
S3D	0.26	For ARPES experiment, from batch S3			
S3E	0.26				

^aThe samples used for the c -axis transport measurements (S3F and S5B) are cleaved from the pieces with known Hall carrier densities.

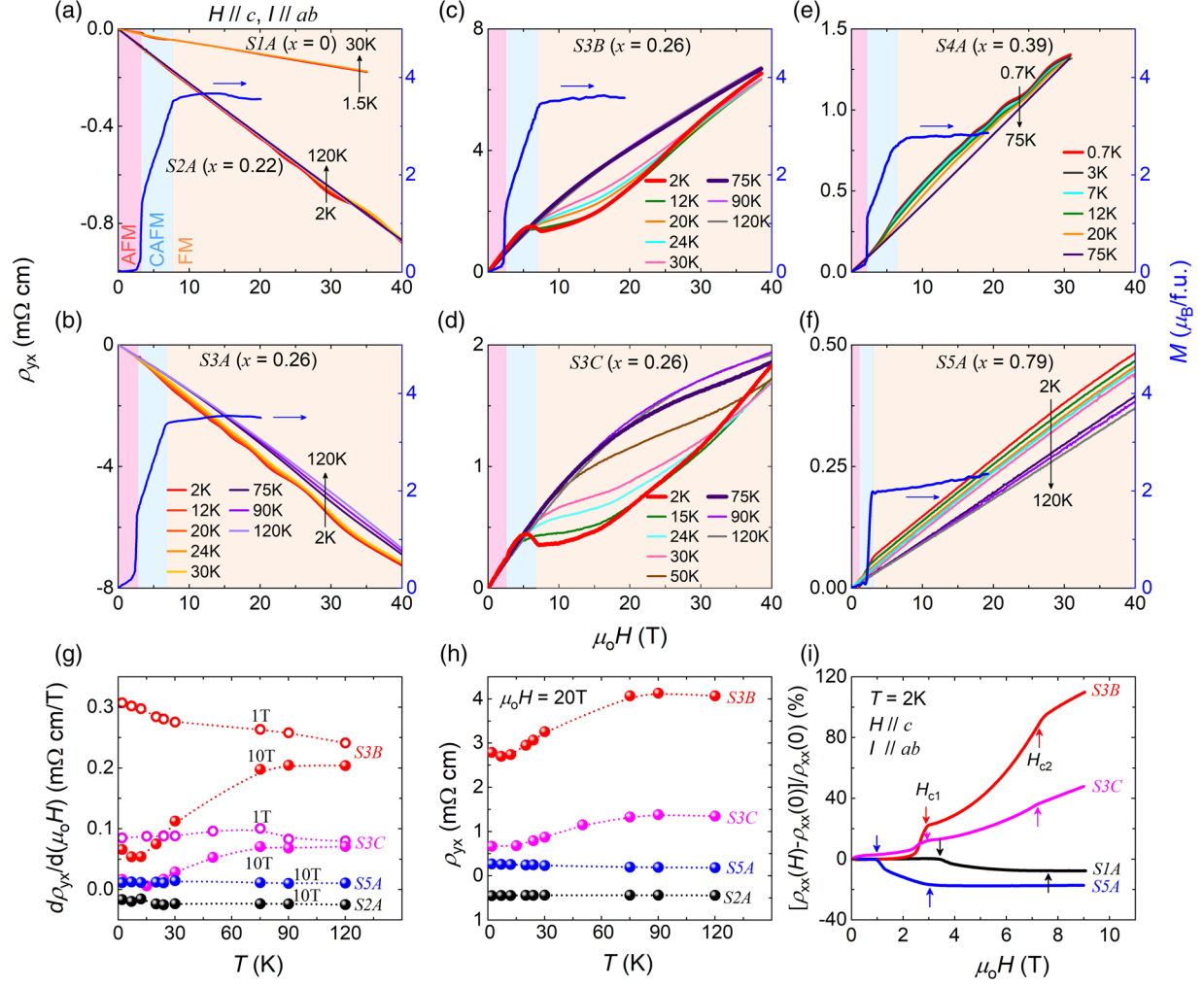


FIG. 1. Magnetic and transport properties of $\text{Mn}(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_4$. Magnetic field dependence of Hall resistivity ρ_{yx} at various temperatures and magnetization M at 2 K for $\text{Mn}(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_4$ with (a) $x = 0$ and 0.22, (b)–(d) 0.26, (e) 0.39, and (f) 0.79. All these measurements are conducted with the currents applied along the ab plane and the magnetic fields perpendicular to the plane. The rose, blue, and orange regions refer to the AFM, CAFM, and FM phase regions, respectively, determined by magnetization [blue curves in (a)–(f)]. The heavily (a),(f) and moderately (e) doped samples exhibit linear magnetic field dependence for ρ_{yx} or almost linear ρ_{yx} background, while the lightly doped samples S3A, S3B, and S3C show nonlinear field dependence in ρ_{yx} (b)–(d). The S3A, S3B, and S3C samples used in this study are taken from the same growth batch but display either electronlike (b) or holelike (c),(d) transport behavior in $\rho_{yx}(H)$ owing to inevitable chemical inhomogeneity as discussed in the text. (g) Temperature dependences of the Hall resistivity slope $d\rho_{yx}/d(\mu_0 H)$ at 1 and 10 T for the lightly hole-doped (S3B, S3C), heavily electron-doped (S2A), and heavily hole-doped (S5A) samples. (h) Temperature dependence of Hall resistivity ρ_{yx} at 20 T for the lightly hole-doped (S3B and S3C), heavily electron-doped (S2A), and heavily hole-doped (S5A) samples. (i) The transverse in-plane magnetoresistivity for lightly hole-doped (S3B and S3C), heavily electron-doped (S1A), and heavily hole-doped samples (S5A). The downward and upward arrows refer to the two magnetic transitions upon increasing the magnetic field along the c axis, i.e., the AFM-to-CAFM transition at H_{c1} and the CAFM-to-FM transition at H_{c2} .

the magnetic field along the c axis, manifested by the AFM-to-canted-antiferromagnetic (CAFM) transition at H_{c1} and the CAFM-to-FM transition at H_{c2} [44]. Both H_{c1} and H_{c2} are suppressed by increasing Sb content and tend to merge as x approaches 1 [43]. Moreover, the Sb substitution for Bi also leads to a nearly linear decrease of the Mn saturation moment, from $3.56\mu_B/\text{Mn}$ for MnBi_2Te_4 to $1.57\mu_B/\text{Mn}$ for MnSb_2Te_4 [43]. This result can be attributed to magnetic fluctuations arising from competing

magnetic interactions. There are two possible origins of competing magnetic interactions. One is the competition between the dominant intralayer FM interaction and the direct AFM exchange interaction of the nearest Mn-Mn neighbors [43]. The other is the competition between the interlayer AFM interaction and the ferrimagnetic order mediated by the Mn-(Bi,Sb) antisite defects [45–47]. In this article, we focus on investigating the magnetotransport properties of $\text{Mn}(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_4$ under high magnetic

fields (up to 41.5 T) and seek transport evidence for the predicted type-II WSM.

Single crystals of $\text{Mn}(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_4$ used in this work are grown by a flux method and characterized using x-ray diffraction (see Sec. III). The composition analyses using energy-dispersive x-ray spectroscopy (EDS) show that the measured Sb content x deviates from the nominal value. The measured x values for each batch of samples as well as other sample information are given in Table I. EDS analyses also show that the variation of the Bi/Sb ratio between different spots of an identical crystal is small, with the largest difference being about 2.2%. We do not observe any clustering of Bi and Sb in the EDS mapping, suggesting that both Bi and Sb distributions are relatively homogenous; Supplemental Material, Sec. S1 [48], shows representative data collected on a sample from the $x = 0.26$ batch. From isothermal magnetization measurements at 2 K [blue curves in Figs. 1(a)–1(f)], we find that the evolution of magnetism of $\text{Mn}(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_4$ with Sb content is consistent with prior studies [42,43]. The AFM, CAFM, and FM phases determined by the magnetization data for each sample are color coded in Figs. 1(a)–1(f). We conduct high magnetic field Hall resistivity ρ_{yx} measurements for representative samples with various x and find that the magnetic field and temperature dependences of ρ_{yx} exhibit drastic changes with increasing Sb content. The slope of ρ_{yx} is negative for $x < 0.26$ [Fig. 1(a)] but positive for $x > 0.26$ [Figs. 1(e) and 1(f)], indicating that $x = 0.26$ is the critical concentration for the carrier type change [Figs. 1(b)–1(d)].

As shown in Figs. 1(b)–1(d), while samples $S3A$, $S3B$, and $S3C$ are all taken from the same batch, sample $S3A$ is lightly electron doped, but samples $S3B$ and $S3C$ are lightly hole doped. Such strong sample dependences of transport properties, which are not observed in $x > 0.26$ (or < 0.26) samples, can be attributed to the fact that the $x = 0.26$ samples have chemical potentials close to the band edge, which makes their magnetotransport properties extremely sensitive to the electronic structure transition induced by the spin-flop transition, as discussed below.

To locate the position of the chemical potentials in the $x = 0.26$ samples, we perform angle-resolved photoemission spectroscopy (ARPES) measurements (see Sec. III) on two samples ($S3D$ and $S3E$) selected from the $x = 0.26$ batch along the Γ - K direction and find that their chemical potentials are indeed close to the conduction or valence band edges. Samples $S3D$ and $S3A$ are originally from the same piece. As shown in Fig. 2(a)(i), the chemical potential of sample $S3D$ is close to the bottom of the bulk conduction band, in good agreement with the light electron doping probed in the transport measurements of sample $S3A$ (see Table I). However, for sample $S3E$, we measure two spots and find that its chemical potential is pushed into the bulk gap at spot 1 [Fig. 2(a)(ii)] but near the top of the bulk valence band at spot 2 [Fig. 2(a)(iii)]. For spot 2, we also construct constant energy maps within two different energy

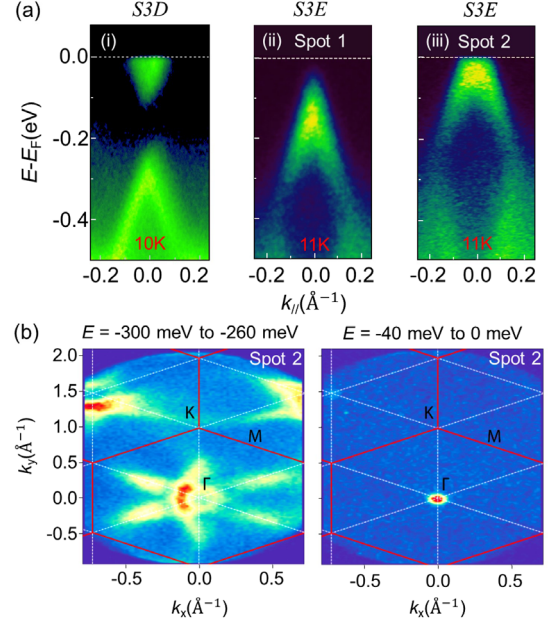


FIG. 2. Band structures of $\text{Mn}(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_4$. (a) ARPES spectra measured on (i) sample $S3D$ using the photon energy of 21.218 eV and (ii),(iii) sample $S3E$ using the photon energy of 81 eV. Spectra in (ii) and (iii) are measured on two different spots of sample $S3E$. All the spectra in (a) are measured below $T_N = 25$ K along the Γ - K direction [see (b)]. (b) ARPES constant energy maps at spot 2, acquired by integrating over -300 to -260 meV (left) and -40 to 0 meV (right). A pointlike Fermi surface centered at Γ can be seen on the k_x - k_y plane.

windows (-300 to -260 meV and -40 to 0 meV) [Fig. 2(b)], showing that the projection of the Fermi surface on the k_x - k_y plane is centered at the Γ point. Since ARPES is a surface-sensitive probe, the difference of the ARPES spectra between spots 1 and 2 does not necessarily imply that the bulk involves large electronic inhomogeneity. If the measured sample is bulk insulating, small doping differences at the surface from surface adsorbents can cause large changes to the Fermi level at the surface. This phenomenon is often seen in topological insulators (e.g., see Ref. [49]). Given that sample $S3E$ is taken from the $x = 0.26$ batch and shows the chemical potential inside the gap at spot 1, its bulk is either insulating or lightly doped. As such, the ARPES spectra difference between spot 1 and 2 in Figs. 2(a)(ii) and 2(a)(iii) is most likely attributable to the small doping difference induced by surface adsorbents. This conjecture is verified by transport measurements on samples $S3B$ and $S3C$, which are also from the $x = 0.26$ batch. From the carrier density derived from the Hall resistivity data of the paramagnetic (PM) state at 75 K (see Fig. 3), the Fermi wave vector k_F is estimated to be approximately 0.012 and 0.015 $\text{\AA}^{-1}$ for samples $S3B$ and $S3C$, respectively, indicating that the average chemical potential of these samples is much closer to the top of the bulk valence band as compared to the scenario seen in Fig. 2(a)(iii). These lightly hole-doped

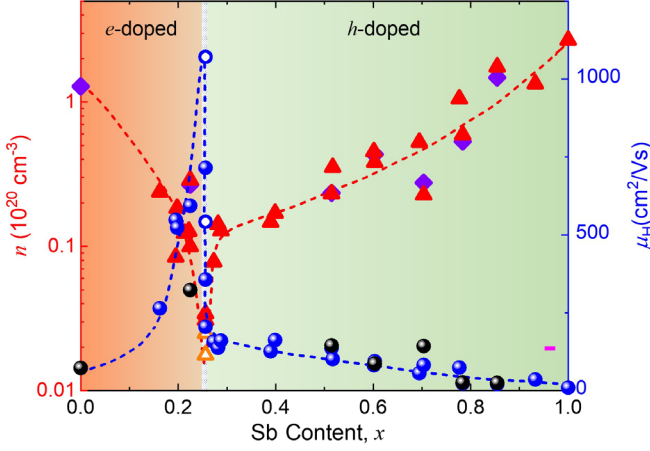


FIG. 3. Composition dependence of carrier density n and transport mobility μ_H of $\text{Mn}(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_4$. For heavily and moderately doped samples showing linear or almost linear ρ_{yx} (Fig. 1 and Supplemental Material, Fig. S1 [48]), n is obtained from the linear slope of $\rho_{yx}(H)$ at $T = 2$ K for the FM phase (purple diamond) and 75 K for the PM phase (red triangle). The black and blue circles represent transport mobility $\mu_H [= R_H/\rho_{xx}(0)]$ for the FM and PM phases, respectively. For lightly hole-doped samples (from the $x = 0.26$ batch) with nonlinear $\rho_{yx}(H)$, n (empty orange triangle) and μ_H (empty blue circle) are extracted from the two-band model fit (see Supplemental Material, Sec. S2 [48]), and only the data of dominant bands are presented here. The magenta bar represents the maximal statistical error of the measured Sb content by EDX.

samples enable us to observe remarkable signatures of the predicted Weyl state, as presented below.

We use the magnetic field dependence of ρ_{yx} to estimate the carrier densities of the samples shown in Fig. 1. The results of this analysis are summarized in Fig. 3; note that the red triangles and purple diamonds in Fig. 3 represent the carrier densities of the samples in the PM states at 75 K and FM states at 2 K, respectively (see more ρ_{yx} data of additional samples in Supplemental Material, Fig. S1 [48]). The heavily electron-doped (S1A) or hole-doped (S5A) samples exhibit a linear magnetic field dependence of ρ_{yx} in both the FM and PM phases [Figs. 1(a) and 1(f)]. The moderately doped samples (S2A and S4A) exhibit quantum oscillations in ρ_{yx} in the FM phase, with their background of ρ_{yx} showing slight deviation from the linear field dependence [Fig. 1(e)]. The carrier densities of these heavily or moderately doped samples are, thus, estimated directly using the linear slope of ρ_{yx} (i.e., Hall coefficient R_H); this estimation shows little or no variation between the PM and FM states (Fig. 3). In strong contrast, in the lightly hole-doped samples S3B and S3C [Figs. 1(c) and 1(d)], ρ_{yx} has a distinct nonlinear dependence on the magnetic field in both PM and FM phases. In these cases, we estimate the carrier densities through the two-band model fits for their PM states (see Supplemental Material, Sec. S2 [48]). For their FM phases which are characterized by the coexistence

of electron and hole pockets as discussed below, we extract the difference between the hole and electron density, $n_h - n_e$, from the slope of ρ_{yx} in the high field limit (approximately 40 T), where $\rho_{yx} \approx [B/(n_h - n_e)e]$ according to the two-band model (see below). We include these data as well as the carrier densities of the FM phases of other samples in Table I. All the carrier densities mentioned below refer to the Hall carrier density of the PM phases (Fig. 3) unless specified. Figure 3 shows that the $x \sim 0.26$ samples have a distinctly lower carrier density at their PM phases compared to other compositions; the minimum is approximately $2 \times 10^{18} \text{ cm}^{-3}$, about 2 orders of magnitude smaller than that of MnBi_2Te_4 [44,50]. Furthermore, we find that the samples with lower carrier density have higher carrier transport mobility μ_H . The maximum value of μ_H is approximately $1.1 \times 10^3 \text{ cm}^2/\text{Vs}$ (see Sec. III), about 2 orders of magnitude larger than in MnBi_2Te_4 (approximately $58 \text{ cm}^2/\text{Vs}$) and MnSb_2Te_4 ($9.7 \text{ cm}^2/\text{Vs}$) (Fig. 3).

The access to the lower carrier density in $\text{Mn}(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_4$ enables the observation of the transport signatures of the predicted ideal FM WSM. As noted above, it has been theoretically predicted that the AFM-to-FM transition driven by magnetic fields along the c axis should lead to a type-II Weyl state for which Weyl nodes are at the touching points of electron and hole pockets [36,37]. As such, for lightly hole-doped samples, we expect a transition from an electronic state with only a hole pocket in the AFM phase to the one with the coexistence of electron and hole pockets in the FM phase. In the ρ_{yx} measurements of samples S3B and S3C, we do observe evidence for such a transition. As shown in Figs. 1(c) and 1(d), the field dependences of ρ_{yx} of these samples exhibit signatures of hole doping and are nearly temperature independent in their PM states above 75 K. However, when the temperature is lowered below 75 K, their ρ_{yx} values reduce significantly, and the values of ρ_{yx} of the FM phase at 2 K are much smaller than those of the PM states above 75 K in a wide field range above H_{c2} . We plot the variation of ρ_{yx} with the temperature at 20 T in Fig. 1(h), where a drastic decrease of ρ_{yx} from the PM to the FM state is seen clearly in samples S3B and S3C, in stark contrast with the very small or negligible variation of ρ_{yx} with temperature seen in moderately and heavily doped samples (e.g., S2A [Fig. 1(a)] and S5A [Fig. 1(f)]). Moreover, the slopes of ρ_{yx} of samples S3B and S3C exhibit striking decreases from the PM to FM states but small increases from the PM to AFM states, sharply contrasted with heavily doped samples for which $d\rho_{yx}/d(\mu_0 H)$ hardly changes across the PM-FM transition, as shown in Fig. 1(g), which plots $d\rho_{yx}/d(\mu_0 H)$ at 10 and 1 T as a function of the temperature for samples S3B, S3C, S2A, and S5A. The drastic decreases of ρ_{yx} as well as the corresponding variation of $d\rho_{yx}/d(\mu_0 H)$ from the PM to the FM state in samples S3B and S3C can be attributed to the presence of

an electron pocket in the FM state, since electrons lead to negative Hall resistivity. That is, the lightly hole-doped samples should feature the coexistence of electron and hole pockets in their FM phases. Note that, while the T_N of the AFM phase is approximately 25 K, the ordering temperature of the field-driven FM phase should be beyond 25 K, since strong magnetic fields can force a PM state to an FM state at temperatures not far away from T_N . This result explains why ρ_{yx} decrease starting from 75 K for samples *S3B* and *S3C*. From Figs. 1(c) and 1(d), we can also see that the discrepancy of ρ_{yx} between the PM and FM states tends to diminish in the high field limit. This tendency is because ρ_{yx} in the high field limit resumes a single-band behavior, i.e., showing linear field dependence [i.e., $\rho_{yx} \propto B/(n_h - n_e)$] for noncompensated systems; this result can be derived from the two-band model for Hall resistivity, $\rho_{yx} = \{[(n_h\mu_h^2 - n_e\mu_e^2) + \mu_h^2\mu_e^2(n_h - n_e)B^2]/[(n_e\mu_e + n_h\mu_h)^2 + \mu_h^2\mu_e^2(n_h - n_e)^2B^2]\} \cdot (B/e)$.

The electronic structure transition concurrent with the spin-flop transition is also manifested by the anomalous peak of ρ_{yx} present in the CAFM state [see the red curves in Figs. 1(c) and 1(d)], which indicates that the electron pockets start to emerge in the CAFM state. The in-plane

transverse magnetoresistivity $MR = [\rho_{xx}(H) - \rho_{xx}(0)]/\rho_{xx}(0)$ measured on samples *S3B* and *S3C* provides further support for the electronic structure transition driven by the spin-flop transition. As shown in Fig. 1(i), the spin-flop transitions in these two samples lead to positive MR (e.g., approximately 110% at 9 T and 2 K for sample *S3B*), sharply contrasted with the heavily electron- and hole-doped samples *S1A* and *S5A*, whose spin-flop transitions result in the negative MR due to the suppression of magnetic scattering by magnetic fields. This contrast suggests that the spin-flop transition in the lightly hole-doped samples results in an additional large positive term in MR, which can be attributed to the sharp increase of carrier mobility due to the presence of Weyl nodes near the Fermi level, as discussed below. Furthermore, the electronic structure transition driven by the spin-flop transition is also evidenced by the temperature dependence of quantum oscillation frequency (see below).

If this implicit electronic structure transition leads to an FM WSM as the theory predicts [36,37], transport measurements should reveal the characteristic transport signatures such as a large intrinsic AHE and a pronounced negative MR due to the chiral anomaly. In our experiments, we indeed observe magnetotransport signatures that are

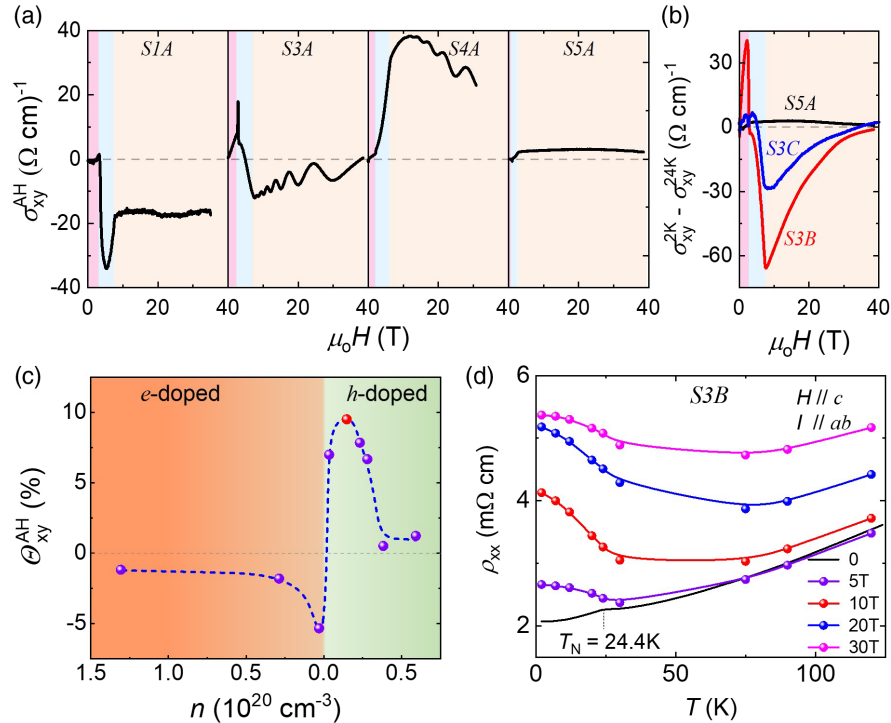


FIG. 4. Anomalous Hall effect in $\text{Mn}(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_4$. (a) Field dependence of anomalous Hall conductivity σ_{xy}^{AH} for samples *S1A*, *S3A*, *S4A*, and *S5A* at 2 and 0.7 K. σ_{xy}^{AH} is derived using $\sigma_{xy}^{AH} = \rho_{yx}^{AH}/[\rho_{xx}^2 + \rho_{yy}^2]$, where ρ_{yx}^{AH} is the anomalous Hall resistivity, obtained after subtracting the normal Hall contribution ($\rho_{yx}^N = R_H H$) (see Sec. III). (b) Field dependence of $\sigma_{xy}^{2K} - \sigma_{xy}^{24K}$ for *S3B*, *S3C*, and *S5A*. σ_{xy}^{2K} and σ_{xy}^{24K} are extracted using $\sigma_{xy} = \rho_{yx}/(\rho_{xx}^2 + \rho_{yy}^2)$ at 2 and 24 K, respectively. (c) The maximal anomalous Hall angle $\Theta_{xy}^{AH} = \sigma_{xy}^{AH}/\sigma_{xx}$ in the FM phase as a function of the carrier density, obtained from the field dependence of Θ_{xy}^{AH} shown in Supplemental Material, Fig. S3 [48]. All data points are obtained at 2 K, except for *S4A* (0.7 K, red circle). (d) Temperature dependence of longitudinal resistivity ρ_{xx} for *S3B* at various fields (applied along the *c* axis).

consistent with these effects in the lightly and moderately hole-doped samples. As indicated above, a large intrinsic AHE of an FM WSM arises from the diverging Berry curvature of the Weyl nodes, which is manifested by large intrinsic anomalous Hall conductivity σ_{xy}^{AH} and anomalous Hall angle ($\Theta_{xy}^{\text{AH}} = \sigma_{xy}^{\text{AH}}/\sigma_{xx}$). Figure 4(a) presents σ_{xy}^{AH} at $T = 2$ or 0.7 K for the heavily and lightly electron-doped samples (*S1A* and *S3A*) and moderately and heavily hole-doped samples (*S4A* and *S5A*). These data are derived from ρ_{yx} and longitudinal resistivity ρ_{xx} (Supplemental Material, Fig. S2 [48]) via tensor conversion, i.e., $\sigma_{xy}^{\text{AH}} = \rho_{yx}^{\text{AH}}/[\rho_{xx}^2 + \rho_{yx}^2]$, where ρ_{yx}^{AH} represents the anomalous Hall resistivity, obtained through subtracting the normal Hall resistivity $\rho_{yx}^{\text{N}} (= R_{\text{H}}H)$ from the measured ρ_{yx} . From Fig. 4(a), it can be seen that σ_{xy}^{AH} strongly depends on carrier type and density. Sample *S1A*, which is heavily electron doped, shows negative σ_{xy}^{AH} , and its absolute value $|\sigma_{xy}^{\text{AH}}|$ rises to a maximum of approximately $35 \Omega^{-1} \text{cm}^{-1}$ within the CAFM phase; we attribute these observations to the presence of an intrinsic AHE due to noncollinear spin structure [44]. However, in the FM phase, $|\sigma_{xy}^{\text{AH}}|$ drops to approximately $18 \Omega^{-1} \text{cm}^{-1}$ and becomes nearly independent of the magnetic field. For the lightly electron-doped sample *S3A*, its σ_{xy}^{AH} shows not only a sign reversal from being positive to negative in the CAFM phase, but also quantum oscillations in the FM phase, with its nonoscillatory component decreasing with the increase of the magnetic field. In contrast, for heavily hole-doped samples, σ_{xy}^{AH} shows very small positive values (e.g., $\sigma_{xy}^{\text{AH}} \sim 3 \Omega^{-1} \text{cm}^{-1}$ for sample *S5A*). However, when the hole carrier density is decreased to a moderate level, σ_{xy}^{AH} of the FM phase increases significantly. For instance, the maximal σ_{xy}^{AH} is approximately $38 \Omega^{-1} \text{cm}^{-1}$ for sample *S4A*, which is comparable to the theoretical value of σ_{xy}^{AH} estimated for the FM Weyl state using $\sigma_{xy}^{\text{AH}} = (e^2/2\pi\hbar)\Delta k$ [51], where Δk represents the Weyl node separation [$\sigma_{xy}^{\text{AH}} \sim 35 \Omega^{-1} \text{cm}^{-1}$ (or $55 \Omega^{-1} \text{cm}^{-1}$) for the Weyl node separation of approximately $0.06 \text{\AA}^{-1}$ [36] (or $0.095 \text{\AA}^{-1}$ [37]) along the k_z axis]. This result suggests that the relatively large σ_{xy}^{AH} observed in the moderately hole-doped samples should be of an intrinsic origin (see more discussions in Supplemental Material, Sec. S3 [48]). Note that sample *S4A* also exhibits quantum oscillations in σ_{xy}^{AH} with its background showing a nearly linear decrease in the FM phase. This result can be attributed to the normal Hall contribution of a small electron pocket, which is not considered in the normal Hall subtraction. We consider only a single hole band contribution in the normal Hall subtraction for this sample. The inclusion of a negative normal Hall contribution of a small electron pocket in the σ_{xy}^{AH} extracted using $\sigma_{xy}^{\text{AH}} = \rho_{yx}^{\text{AH}}/[\rho_{xx}^2 + \rho_{yx}^2]$ leads to an underestimated value of σ_{xy}^{AH} . The intrinsic σ_{xy}^{AH} of sample

S4A should be greater than $38 \Omega^{-1} \text{cm}^{-1}$. Similarly, the slight background increase of σ_{xy}^{AH} above H_{c2} for the lightly electron-doped sample *S3A* should also be attributed to the coexistence of a large electron pocket with a small hole pocket, and the small positive normal Hall contribution from the small hole pocket is not considered in the normal Hall subtraction.

The coexistence of electron and hole pockets in the FM phase is more remarkably manifested by the temperature dependence of Hall conductivity in the lightly hole-doped samples *S3B* and *S3C* [Figs. 1(c) and 1(d)]. For these samples, it is difficult to separate ρ_{yx}^{AH} from ρ_{yx}^{N} , since ρ_{yx}^{N} does not follow a linear field dependence at all due to the coexistence of electron and hole pockets [Figs. 1(c) and 1(d)]. For this reason, we attempt a different approach to evaluate σ_{xy}^{AH} for these samples. We compute the Hall conductivity at 2 and 24 K using $\sigma_{xy} = \rho_{yx}/(\rho_{xx}^2 + \rho_{yx}^2)$. Since the magnetic transition temperatures of these samples are approximately 24.4 K, we define the σ_{xy} at 24 K as the normal Hall conductivity σ_{xy}^{N} . If the PM-to-FM transition does not drive an electronic structure transition, the anomalous Hall conductivity σ_{xy}^{AH} at 2 K can be obtained by subtracting σ_{xy}^{N} from σ_{xy} at 2 K; this method is often used in analyzing the intrinsic anomalous Hall conductivity of magnetic topological semimetals (e.g., see Ref. [8]). As shown in Fig. 4(b), $\sigma_{xy}(2 \text{ K}) - \sigma_{xy}(24 \text{ K})$ shows a strong nonlinear field dependence for samples *S3B* and *S3C*; this result is inconsistent with the expected constant σ_{xy}^{AH} for the polarized FM phase but implies that the normal Hall contribution is not completely removed by subtracting $\sigma_{xy}(24 \text{ K})$ from $\sigma_{xy}(2 \text{ K})$ and undergoes a substantial change below the magnetic transition. This result not only provides further support for the argument that there is an electronic structure transition occurring concomitantly with the PM-to-FM magnetic transition near 24 K for the lightly hole-doped samples, but also suggests that the electronic structure varies with the temperature below 24 K, which is indeed verified by the temperature dependence of the quantum oscillation frequency as presented below.

By contrast, for the heavily hole-doped sample *S5A*, which is not expected to exhibit an electronic structure transition driven by the magnetic transition, the value of $[\sigma_{xy}(2 \text{ K}) - \sigma_{xy}(24 \text{ K})]$ indeed shows very weak field dependence and is comparable with the σ_{xy}^{AH} value obtained using $\sigma_{xy}^{\text{AH}} = \rho_{yx}^{\text{AH}}/[\rho_{xx}^2 + \rho_{yx}^2]$, as shown in Fig. 4(b). Such a sharp contrast between the lightly and heavily hole-doped samples provides further support for the field-driven electronic structure reconstruction in the lightly hole-doped samples. Although it is difficult to separate σ_{xy}^{AH} from the normal Hall contribution for the lightly hole-doped samples, the fact that the experimentally extracted value of σ_{xy}^{AH} for the moderately hole-doped sample *S4A* is comparable to the theoretical value expected for the ideal Weyl state as

indicated above suggests that a large intrinsic AHE should also occur to the lightly hole-doped samples, since their Weyl nodes are closer to the Fermi level. Furthermore, the positive-to-negative sign reversal of $[\sigma_{xy}(2\text{ K}) - \sigma_{xy}(24\text{ K})]$ in the CAFM phases of samples *S3B* and *S3C* suggests that the field-driven electronic reconstruction should start to occur in the CAFM phases. The above interpretations for the field-dependent Hall conductivity shown in Figs. 4(a) and 4(b) are consistent with the evolution of Fermi surface predicted by theory for $\text{Mn}(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_4$ [52]—the coexistence of electron and hole pockets is present only when the chemical potential is close to the Weyl nodes, and the relative electron or hole pocket size is sensitively dependent on the chemical potential which is tuned by Sb concentration. Our DFT calculations reproduce similar results (see Supplemental Material, Sec. S4 and Fig. S11 [48]).

To further examine the evolution of the AHE with carrier density, we convert σ_{xy}^{AH} to anomalous Hall angle Θ_{xy}^{AH} (see Sec. III and Supplemental Material, Fig. S3 [48]) and plot the maximal Θ_{xy}^{AH} or $-\Theta_{xy}^{\text{AH}}$ of the FM phase at $T = 2\text{ K}$ as a function of the carrier density in Fig. 4(c). We do not include samples *S3B* and *S3C* in this plot, since their σ_{xy}^{AH} cannot be separated from their normal Hall contribution. Figure 4(c) shows that $|\Theta_{xy}^{\text{AH}}|$ increases with decreasing carrier density for both electron- and hole-doping regimes. For the hole-doping side, Θ_{xy}^{AH} increases up to approximately 9% for moderately hole-doped samples. Since σ_{xy}^{AH} is slightly underestimated due to the residual normal Hall contribution of a small electron pocket for these samples as noted above, their intrinsic Θ_{xy}^{AH} should be greater than 9%.

The temperature dependences of ρ_{xx} measured under magnetic fields for the lightly hole-doped samples also agree well for the expected field-induced FM WSM. As shown in Fig. 4(d), when the magnetic field is strong enough to drive the AFM state to the CAFM or FM state, $\rho_{xx}(T)$ exhibits an insulatinglike behavior, in contrast with the metallic behavior under zero magnetic field. Given that the magnetic-field-induced insulatinglike behavior is commonly observed in topological semimetals due to their large magnetoresistance at low temperatures [15], such behavior is consistent with the predicted field-induced FM WSM. We note that prior theoretical studies by Zhang *et al.* [36] suggest that this FM WSM can be either type I or type II depending on the lattice parameters. However, recent DFT calculations by Wang [53] suggest that in the $\text{Mn}(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_4$ alloy system, as the Sb content x is increased from $x = 0$ to 0.25, a type-II Weyl state with the Weyl nodes being close to the Fermi level can be realized without the requirement of artificial lattice expansion, consistent with our experimental observations of the field-driven electronic structure transition in samples with x being close to 0.25. Our anomalous Hall conductivity data also point to a type-II WSM. If it were a type-I WSM,

we would expect the sign of σ_{xy}^{AH} to always remains positive even when the system is tuned from hole doping to electron doping [51]. But, for a type-II WSM, theoretical studies [54] show that the sign of σ_{xy}^{AH} could vary from negative to positive when the chemical potential is tuned across the charge neutral point. Therefore, our observation of the sign reversal of anomalous Hall conductivity tuned by chemical potential [Fig. 4(a)] supports the conjecture that the FM WSM driven by the spin-flop transition in $\text{Mn}(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_4$ is a type-II WSM.

Another distinct magnetotransport property of a WSM is the chiral anomaly, which is manifested by negative longitudinal magnetoresistivity (LMR) [6,20,55,56]. Since the predicted FM WSM state for MnBi_2Te_4 has one pair of Weyl nodes aligned along the k_z direction [36,37], its chiral anomaly is expected to be remarkable for the c -axis LMR. To probe such an effect, we perform c -axis magnetoresistivity $\text{MR} = [\rho_{zz}(H) - \rho_{zz}(0)]/\rho_{zz}(0)$ measurements on both lightly and heavily hole-doped samples. Figure 5(a) presents the typical c -axis MR data of a lightly hole-doped sample *S3F*, which is cleaved from a piece with the measured carrier density of $n_h = 0.097 \times 10^{20}\text{ cm}^{-3}$. The spin-flop transition in this sample leads to a large negative LMR, with its magnitude rising to approximately 45% at 9 T and 2 K. When the field is rotated away from the c axis [inset, Fig. 5(b)], the magnitude of the MR gradually decreases and the sign of MR changes to positive when the field tilt angle is above 60° . In contrast, in a heavily hole-doped sample *S5B* ($n_h = 0.59 \times 10^{20}\text{ cm}^{-3}$), we find that the spin-flop transition leads to a relatively small c -axis negative LMR ($\text{MR} \sim -20\%$ at 2 K and 9 T), and its MR hardly shows angular dependence in the polarized FM state above 4 T [see Fig. 5(b)], indicating that the negative MR of this sample originates from suppressed magnetic scattering, similar to the scenario seen in heavily electron-doped MnBi_2Te_4 [44]. The sharp contrast in the c -axis MR between the lightly hole-doped and heavily hole- and electron-doped samples suggests that the large negative LMR of sample *S3F* should be dominated by the chiral anomaly. Of course, the negative MR component arising from suppressed magnetic scattering may not be completely excluded in this sample, but its contribution should be relatively small, as evidenced by the striking angular dependence of its MR [Fig. 5(a)]. The reason why only lightly hole-doped samples such as *S3F* show signatures of chiral anomaly is that their Weyl nodes are close to the Fermi level, thus making the chiral anomaly effect significant. We also perform the in-plane LMR measurements on lightly hole-doped samples as well as the heavily electron- and hole-doped samples, as presented in Fig. S4. The lightly hole-doped samples exhibit positive in-plane LMR, contrasted with the negative LMR due to the suppression of magnetic scattering probed in the heavily doped samples. The absence of in-plane LMR associated with the chiral anomaly in the lightly hole-doped samples can be attributed

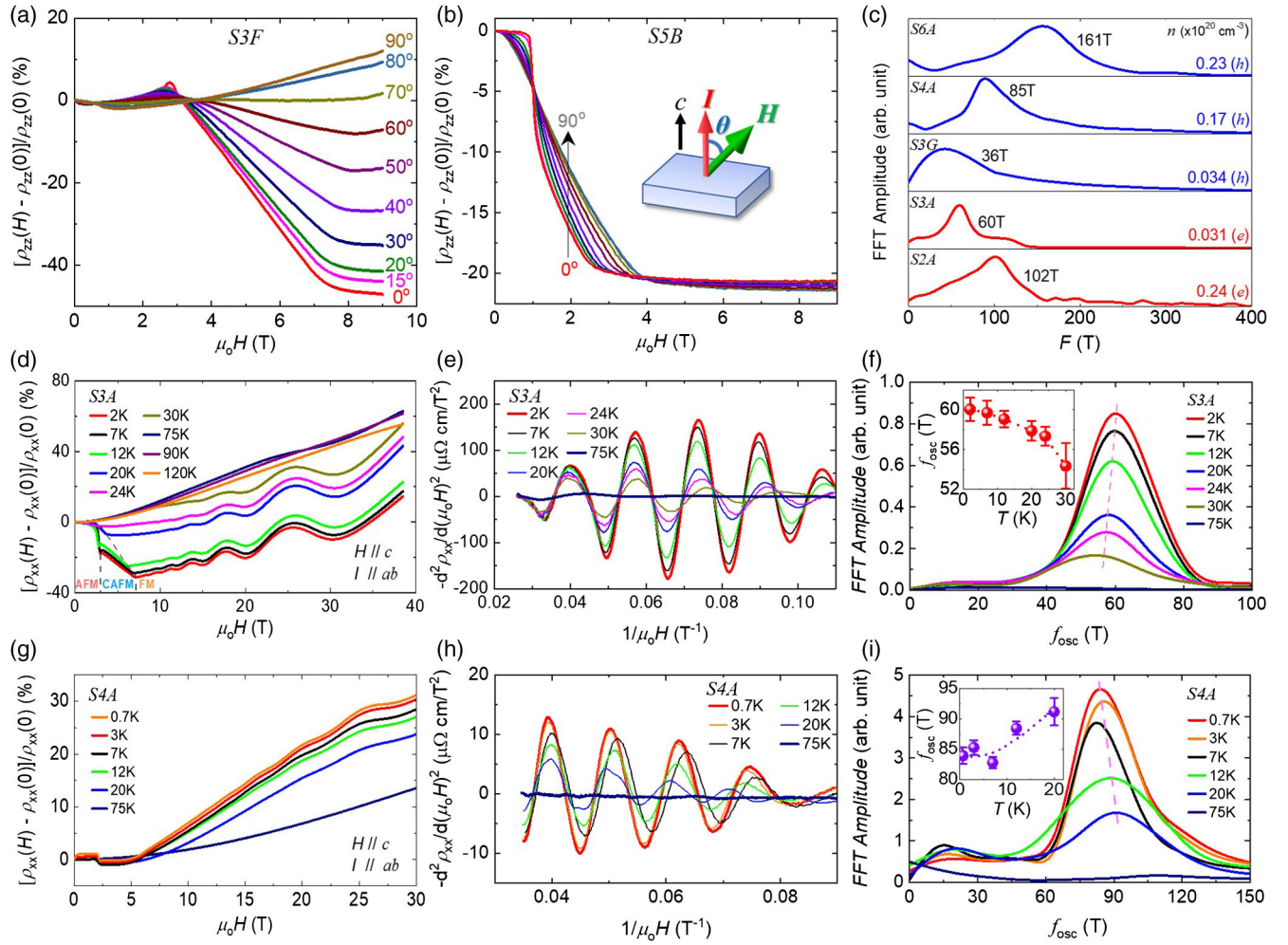


FIG. 5. Magnetoresistivity and SdH oscillations in $\text{Mn}(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_4$. (a),(b) The c -axis magnetoresistivity $\text{MR} = [\rho_{zz}(H) - \rho_{zz}(0)]/\rho_{zz}(0)$ under various field orientations for (a) the lightly ($S3F$) and (b) heavily hole-doped ($S5B$) samples. The schematic in (b) illustrates the experimental setup for the c -axis MR angular dependence measurements. (c) FFT spectra of $-d^2\rho_{xx}/d(\mu_0 H)^2$ for various moderately and lightly electron- and hole-doped samples [see Supplemental Material, Fig. S5(a) [48], for the SdH oscillations]. (d),(g) In-plane transverse MR of samples (d) $S3A$ and (g) $S4A$ at various temperatures. (e),(h) The second derivative of the in-plane resistivity $\rho_{xx}(H)$ with respect to field H (perpendicular to the plane) for samples (e) $S3A$ and (h) $S4A$ as a function of the inverse of the field at various temperatures. (f),(i) FFT spectra of the second derivative of $\rho_{xx}(H)$ for samples (f) $S3A$ and (i) $S4A$ at various temperatures. The insets in both (f) and (i) show the oscillation frequency (f_{osc}) as a function of the temperature for samples $S3A$ and $S4A$, respectively. The error bar shown in the insets in (f) and (i) is defined as the difference of the oscillation frequencies obtained from the FFT spectra of $-d^2\rho_{xx}/d(\mu_0 H)^2$ and $\Delta\rho_{xx}(H)$ [the oscillatory component of $\rho_{xx}(H)$], respectively. All the measurements shown in (c), (d), and (g) are conducted with the currents applied along the ab plane and the magnetic fields perpendicular to the plane.

to the fact that the chiral anomaly of a type-II Weyl state is expected to be anisotropic, and the striking chiral anomaly is observed only when current is applied to specific directions [1,27].

The electronic structure evolution of $\text{Mn}(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_4$ with the magnetic transition is also corroborated by Shubnikov-de Haas (SdH) oscillations (see Supplemental Material, Fig. S5 [48]). SdH oscillation frequency decreases with carrier density for both electron and hole-doping sides [Fig. 5(c)]. However, the lightly hole-doped samples ($S3B$ and $S3C$) do not exhibit SdH oscillations. This result is consistent with the picture established above, i.e., that the

Weyl nodes in the lightly hole-doped samples are close to the Fermi level. As shown in Figs. 5(d) and 5(g), the SdH oscillations of samples $S3A$ and $S4A$ appear following the spin-flop transitions, and they do not originate from topological surface states but stem from the bulk bands of the FM state (see more discussions in Supplemental Material, Sec. S5 [48]). The data in Fig. 5(d) also show that the SdH oscillations in sample $S3A$ extend to the PM state below $T = 75$ K, where the applied magnetic field can drive the system to a forced FM state as noted above. From the second derivative of $\rho_{xx}(H)$ [Figs. 5(e) and 5(h)], it can be seen that the SdH oscillation pattern varies with the temperature in

both samples. The oscillation frequency f_{osc} derived from fast Fourier transform (FFT) analyses [Figs. 5(f) and 5(i)] increases by approximately 9.5% from 30 to 2 K for S3A [inset, Fig. 5(f)] but decreases by approximately 8.8% from 20 to 0.7 K for S4A [inset, Fig. 5(i)]. Such significant variations of f_{osc} with the temperature suggest a strong coupling between the electronic structure and magnetism. As discussed above, the PM-to-FM transition in the lightly hole-doped samples drives an electronic structure transition with the Fermi surface evolving from a hole pocket in the PM state to the one consisting of both electron and hole pockets. Such a Fermi surface reconstruction across the magnetic transition arises from the band splitting and crossing caused by the FM exchange interaction. While the FM exchange interactions sets in at the magnetic ordering temperature, the further decrease of temperature suppresses thermal fluctuations, thus enhancing the ordered moments, which should increase the band splitting. Therefore, both the electron and hole pocket sizes should vary with the temperature, which accounts for the temperature dependence of f_{osc} . The variation of the Fermi surface with the temperature for the FM phase of the lightly hole-doped samples is also manifested by the strong nonlinear field dependence of $\sigma_{xy}^{2K} - \sigma_{xy}^{24K}$ [Fig. 4(b)], as discussed above. For those samples showing SdH oscillations, we also estimate their carrier densities from f_{osc} using $n_{\text{SdH}} = n_{2D}/c$, where $n_{2D} = 2ef_{\text{osc}}/h$, e is the elemental charge, h is the Planck constant, and c is the lattice parameter. As shown in Table I, n_{SdH} is comparable to the transport carrier density n_{Hall} of the FM phase extracted from the Hall coefficient for the heavily electron- and hole-doped samples, but we cannot make this comparison for the lightly hole-doped samples S3B and S3C, since they do not show SdH oscillations. It is worth noting that for sample S4A, while its anomalous Hall conductivity data [Fig. 4(a)] suggest its Fermi surface of the FM phase involves the coexistence of a large hole pocket and a small electron pocket, only the hole pocket is probed in the SdH oscillations. The fact that the electron pocket is not manifested by the SdH oscillations can be attributed to the low electron mobility.

We also estimate the effective cyclotron mass m_c^* through the fits of temperature dependences of the oscillation amplitude to the thermal damping factor of the Lifshitz-Kosevich (LK) formula (see Supplemental Material, Fig. S5 [48]); m_c^* is approximately $0.13m_0$ (m_0 is the free electron mass) for sample S3A and approximately $0.27m_0$ for sample S4A. The hole effective mass m^* extracted from the band structure calculated using experimental lattice parameters at the density functional theory level (see Supplemental Material, Sec. S4 [48]) is $m^* = 0.02m_0$ for the FM Weyl phase of MnBi_2Te_4 , much smaller than that of the AFM phase, $m^* = 0.33m_0$. For a calculated alloy structure of $\text{Mn}(\text{Bi}_{0.75}\text{Sb}_{0.25})_2\text{Te}_4$ close to the experimental critical composition ($x = 0.26$), the hole effective mass extracted from the FM Weyl state is also about $0.02m_0$, increasing toward $0.1m_0$ at the boundaries of the

plot, where the Fermi level is away from the Weyl point by approximately 60 meV (see Supplemental Material, Sec. S4 [48]). This value is smaller than the experimentally measured $m^* = 0.27m_0$ for sample S4A, which can be attributed to the fact that the Weyl nodes of this sample are farther from E_F than those of the samples with the composition of $\text{Mn}(\text{Bi}_{0.75}\text{Sb}_{0.25})_2\text{Te}_4$.

We note that the layered compound EuCd_2As_2 with an A-type AFM order has also recently been reported to host a WSM state with one pair of Weyl nodes in either its PM state [57] or field-driven FM phase [58,59]. However, the nature of the Weyl state in MnBi_2Te_4 is quite distinct from that in EuCd_2As_2 : First, the Weyl cones in MnBi_2Te_4 are much more strongly tilted, forming a type-II WSM state; second, the tunability of the chemical potential by substituting Sb for Bi enables the observation of remarkable exotic quantum transport properties of Weyl fermions, including an intrinsic AHE and the chiral anomaly.

III. METHODS

A. Sample preparation and measurements

Single crystals of $\text{Mn}(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_4$ with x from 0 to 1 are grown by a self-flux method as previously reported [50]. The mixtures of high-purity manganese powder (99.95%), antimony shot (99.9999%), bismuth shot (99.999%), and tellurium ingot (99.9999 + %) with the molar ratio of $\text{Mn}:\text{Sb}:\text{Bi}:\text{Te} = 1:5x:5(1-x):16$ are loaded into an aluminum crucible and sealed in evacuated quartz tubes. The mixtures are heated up to 900°C for 12 h to promote homogeneous melting and slowly cooled down (1.5°C/h) to a temperature within the 590°C–630°C range (depending on x), followed by centrifugation to remove excess flux. Higher temperatures are used for the samples with higher Sb content to avoid the formation of Sb_2Te_3 (melting point approximately 620°C). Soft, platelike single crystals up to $10 \times 5 \text{ mm}^2$ can be obtained.

The phase purity of these single crystals is checked by x-ray diffraction. As shown in Supplemental Material, Fig. S6 [48], the sharp (00L) x-ray diffraction peaks, which show only the small high-angle shift from MnBi_2Te_4 to MnSb_2Te_4 , demonstrate the excellent crystallinity and the formation of the desired crystal structure with the stacking of $\text{Te}(\text{Bi,Sb})\text{-Te-Mn-Te}(\text{Bi,Sb})\text{-Te}$ septuple layers in our single-crystal samples. The composition analyses by energy-dispersive x-ray spectroscopy (EDS) show the actual Sb content x deviating from the nominal composition. In this article, we use the measured Sb content x .

The low magnetic field magnetotransport and magnetization measurements are performed using a commercial physical property measurement system (Quantum Design). The high magnetic field transport and magnetization measurements are carried out using the 31, 35, and 41.5 T resistive magnets at the NHMFL in Tallahassee.

The standard four-probe method is used for magnetotransport measurements. Samples used for in-plane resistivity ρ_{xx} and Hall resistivity ρ_{yx} measurements have six attached leads, with one pair of current leads and two pairs of Hall voltage leads. The applied current is not aligned to any specific crystallographic axis in the ab plane. Field sweeps of ρ_{xx} and ρ_{yx} are conducted for both positive and negative fields. The field dependences of ρ_{xx} and ρ_{yx} are obtained, respectively, using $\rho_{xx} = [\rho_{xx}(+\mu_0 H) + \rho_{xx}(-\mu_0 H)]/2$ and $\rho_{yx} = [\rho_{yx}(+\mu_0 H) - \rho_{yx}(-\mu_0 H)]/2$. The transport mobility μ_H is inferred from the Hall coefficient $R_H \{= [d\rho_{yx}/d(\mu_0 H)]\}$ via $\mu_H = R_H/\rho_{xx}(0)$, where $\rho_{xx}(0)$ is the zero-field longitudinal resistivity (shown in Supplemental Material, Fig. S2 [48]). The carrier density for the samples exhibiting linear field dependence in ρ_{yx} is estimated via $n = (1/eR_H)$, where e is the electric charge. For those samples showing nonlinear $\rho_{yx}(H)$, two-band model fitting is used to estimate the carrier density and its mobility (see Supplemental Material, Sec. S2 [48]). The VSM method is used for high field magnetization measurements. The ARPES measurements on sample *S3D* are carried out using the 2DCC-MIP's ARPES facility at Penn State. The photoelectrons are excited by an unpolarized He-I $_{\alpha}$ light (21.218 eV), and two analyzers (Scientia R3000 and DA30L) are used for these measurements. The ARPES spectra of sample *S3E* are acquired at the Advanced Light Source Maestro Beam line (BL 7.0.2) using the photon energy of 81 eV and the microARPES end station. The energy resolution is approximately 19 meV, and the momentum resolution is $0.01 \text{ \AA}^{-1}$.

B. Analyses of anomalous Hall conductivity σ_{xy}^{AH} and anomalous Hall angle Θ_{xy}^{AH}

We extract anomalous Hall conductivity σ_{xy}^{AH} from measured Hall resistivity ρ_{yx} and longitudinal resistivity ρ_{xx} via resistivity-to-conductivity tensor conversion. For the samples showing linear field dependence in ρ_{yx} in their FM states (i.e., *S1A*, *S2A*, *S3A*, *S4A*, and *S5A*; see Fig. 1), their σ_{xy}^{AH} is derived using $\sigma_{xy}^{\text{AH}} = \rho_{yx}^{\text{AH}}/[\rho_{xx}^2 + \rho_{yx}^2]$, where ρ_{yx}^{AH} is anomalous Hall resistivity, obtained after subtracting the normal Hall contribution ($\rho_{yx}^{\text{N}} = R_H H$) from the measured ρ_{yx} . The ρ_{xx} data of the samples shown in Fig. 1 are presented in Supplemental Material, Fig. S2 [48], except for the $x = 0$ sample whose ρ_{xx} data are reported in our previous work [44]. For those lightly doped samples, since their ρ_{yx} of FM states do not show linear field dependence due to the coexistence of electron and hole pockets in the FM phase, we attempt a different approach to extract their anomalous Hall conductivity, i.e., $\sigma_{xy}^{\text{AH}} = \sigma_{xy} - \sigma_{xy}^{\text{N}}$ and $\sigma_{xy} = \rho_{yx}/(\rho_{xx}^2 + \rho_{yx}^2)$. Here, σ_{xy}^{N} is the normal Hall conductivity, which can approximately be defined as the σ_{xy} value at 24 K, slightly below $T_N (= 24.4 \text{ K})$. In Fig. S7 [48], we show the calculated σ_{xy} of the *S3B* sample as an

example. As discussed in the main text, the anomalous Hall angle Θ_{xy}^{AH} shown in Fig. 4(c) and Supplemental Material, Fig. S3 [48], is defined as $\sigma_{xy}^{\text{AH}}/\sigma_{xx}$; σ_{xx} is also obtained via tensor conversion, i.e., $\sigma_{xx} = \rho_{xx}/(\rho_{xx}^2 + \rho_{yx}^2)$.

C. Band structure calculations

All density functional theory calculations are performed using the Vienna *ab initio* simulation package (VASP) [60], using the generalized gradient approximation exchange-correlation functional as parametrized by Perdew, Burke, and Ernzerhof [61,62]. Structural relaxations are performed with cutoff energies for plane-wave expansions at 400 eV, until forces are below $0.01 \text{ eV/\AA}$. Strong on-site Coulomb interactions of localized *3d* electrons are treated under the GGA + U scheme, where the effective U parameters $U_{\text{eff}} = U - J$ (in the approach of Ref. [63]) are 4 eV for *3d* electrons in Mn. Noncollinear magnetism and spin-orbit coupling are included following the VASP implementation [64]. Interlayer dispersion forces are described by the DFT-D3 van der Waals correction scheme due to Grimme [65]. Following Ref. [43], the experimental lattice parameters we use are $a = 4.334 \text{ \AA}$ and $c = 40.931 \text{ \AA}$ for MnBi_2Te_4 and $a = 4.310 \text{ \AA}$ and $c = 40.939 \text{ \AA}$ for $\text{Mn}(\text{Bi}_{0.75}\text{Sb}_{0.25})_2\text{Te}_4$.

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Note added.—Recently, we became aware of a related work about SdH studies on the same material system [52].

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