A magnetic topological semimetal $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$ ($y, z < 0.10$)

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Weyl semimetals (WSM) evolve from Dirac semimetals (DSMs) in the presence of broken time reversal symmetry (TRS) or space inversion symmetry. The WSM phases in TaAs class materials and photonic crystals are due to the loss of space inversion symmetry. For TRS breaking WSMs, despite numerous theoretical and experimental efforts, few examples have been reported. In this article, we report a new type of magnetic semimetal $\text{Sr}_{1-y}\text{Mn}_1$. 
$\text{Sr}_{1-y}\text{Mn}_1\text{Sb}_2$ exhibits nearly massless relativistic fermion behavior ($m^* = 0.04-0.05 m_0$ where $m_0$ is the free electron mass). This material exhibits a ferromagnetic (FM) order for $304 \text{ K} < T < 565 \text{ K}$, but a canted antiferromagnetic order with a FM component for $T < 304 \text{ K}$.

The combination of relativistic fermion behavior and ferromagnetism in $\text{Sr}_{1-y}\text{Mn}_1\text{Sb}_2$ offers a rare opportunity to investigate the interplay between relativistic fermions and spontaneous TRS breaking.
3D Dirac semimetals (DSMs) can be viewed as 3D analogues of graphene and are characterized by linear energy-momentum dispersions near the Fermi level along all three momentum directions\(^1\)\(^{-6}\). The linear band crossing point, \(i.e.\) the Dirac point, is protected against gap formation by crystal symmetry. Such unique band structures of 3D DSMs result in peculiar, exotic properties such as high bulk carrier mobility\(^7\) and large linear magnetoresistance\(^7\),\(^8\). 3D DSMs have been experimentally realized in many material systems such as Na\(_3\)Bi\(^2\), Cd\(_3\)As\(_2\)\(^4\)-\(^6\), ZrTe\(_5\)\(^9\) \textit{etc.} Moreover, a new form of DSM state featuring Dirac band crossing along a line/loop has also been established in PbTaSe\(_2\)\(^10\), PtSn\(_4\)\(^11\), and ZrSiS\(^12\). DSMs can be regarded as parent materials of Weyl semimetals (WSMs). When either time-reversal symmetry (TRS) or space inversion symmetry is broken, DSMs evolve into WSMs\(^13\),\(^14\) due to lifted spin degeneracy. Remarkable characteristics of Weyl state include surface Fermi arcs connecting Weyl points\(^15\) and the chiral anomaly, which originates from charge pumping between Weyl points with opposite chirality and is manifested as negative longitudinal magnetoresistance (LMR)\(^16\)-\(^18\). WSMs generated by the broken space inversion symmetry were first experimentally realized in TaAs-class materials\(^19\)-\(^23\) and photonic crystals\(^24\). A Weyl state due to spontaneous TRS breaking was recently reported in YbMnBi\(_2\)\(^25\) and its TRS breaking is suggested to be caused by a net ferromagnetic (FM) component of a canted antiferromagnetic state (CAFM)\(^25\).

In this letter, we report the first observation of relativistic fermion behavior in a material showing ferromagnetic properties, \(i.e.\) \(\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2\) \((y, z < 0.1)\). We observed strong Shubnikov-de Haas (SdH) and de Haas-van Alphen (dHvA) oscillations in this material. The analyses of the SdH and dHvA oscillations demonstrate this material harbors nearly massless relativistic fermions \((m^* = 0.04\text{-}0.05m_0)\) along with a nontrivial Berry phase. Neutron scattering
measurements reveal a FM transition at 565 K, followed by a transition at 304 K to a CAFM with a net ferromagnetic (FM) component, similar to the predicted CAFM state for YbMnBi$_2$ $^{25}$. These findings make Sr$_{1-y}$Mn$_{1-z}$Sb$_2$ a promising candidate for investigating the effect of TRS breaking on the electronic band structure.

SrMnSb$_2$ is closely related to AMnBi$_2$ (A=Sr, Ca, Eu or Yb), as these materials exhibit fascinating properties which have origins in their structural building blocks, namely, the 2D Bi square net which harbors relativistic fermions. SrMnBi$_2$ and CaMnBi$_2$ possess anisotropic Dirac cone states $^{26,27}$; EuMnBi$_2$ was recently found to show a quantum Hall effect due to the magnetically confined 2D Dirac fermions $^{28}$, and YbMnBi$_2$ displays a TRS breaking Weyl state due to the lifted spin degeneracy caused by the FM component of a canted AFM state $^{25}$. Given that the 2D Bi square net can host relativistic fermions, one natural expectation is that 2D Sb network in SrMnSb$_2$ may also host relativistic fermions. One advantage of SrMnSb$_2$ is that the Sb 2D network should have weaker spin-orbital coupling (SOC) than the Bi square net, which might reduce or close the SOC-induced gap near the Dirac point, making it easy to observe exotic phenomena related to massless Dirac fermions $^{29-30}$. This idea serves as the motivating principle for our work.

We synthesized plate-like single crystals of Sr$_{1-y}$Mn$_{1-z}$Sb$_2$ ($y, z < 0.1$) (see the inset to Fig. 1a) using self-flux method (see the Method section). Single-crystal neutron scattering measurements reveal that the synthesized material crystalizes in an orthorhombic structure with space group $Pnma$, similar to the previously-reported structure of SrMnSb$_2$ $^{31}$. No structural transition was observed down to 5 K. The lattice and other structural parameters obtained from
The neutron diffraction refinement are presented in Note 1 in the Supplementary Information (SI).

The compositions of our single crystals were analyzed using an energy dispersive X-ray spectrometer, which shows that the actual composition involves Mn and Sr deficiency as described by Sr$_{1-y}$Mn$_{1-z}$Sb$_2$ ($y, z < 0.1$). Interestingly, the Mn and Sr nonstoichiometry were found to have a strong effect on the magnetic properties of Sr$_{1-y}$Mn$_{1-z}$Sb$_2$. Samples with larger Sr deficiency ($y \sim 0.08, z \sim 0.02$) display stronger FM behavior, while weaker FM behavior occurs in samples with enhanced Mn deficiency ($y \sim 0.01-0.04, z \sim 0.04-0.1$). According to the magnitude of the FM saturated moment $M_s$, we categorize our samples into three types; $M_s \sim 0.1-0.6 \mu_B$/Mn for type A, 0.04-0.06 $\mu_B$/Mn for type B and 0.004-0.006 $\mu_B$/Mn for type C. Detailed comparisons of magnetic properties between type A, B and C samples will be given below. All samples used in this study were first screened through magnetization measurements and labelled with different numbers (e.g., A#1, A#2). Then we cleaved each screened crystal to small pieces for various measurements and label each small piece by adding a lower case letter (e.g., A#1a, A#1b).

We will first present the electronic transport properties of Type A samples and compare them with those of type B and C samples. All the transport data presented in Fig. 1a-1c and 2a-2e were collected on type A samples. As seen in Fig. 1a, both in-plane ($\rho_{xx}$) and out-of-plane ($\rho_{out}$) resistivity exhibit metallic temperature dependences, with the in-plane residual resistivity ratio $\rho_{xx}(300K)/\rho_{xx}(2K)$ being $\sim 29$. The $\rho_{out}/\rho_{xx}$ ratio increases markedly with decreasing temperature and reaches 609 at 2K, suggesting quasi-2D electronic band structure. We conducted both in-plane and out-of-plane ($i.e.$ [100]) magnetotransport measurements for a type A sample. The magnetoresistivity MR ($= [\rho(B)-\rho(0)]/\rho(0)$) along both directions exhibit strong SdH oscillations for $T < 30$ K. We present the out-of-plane MR data in Fig. 1b and the in-plane MR data is given
in Note 2 in SI. We find that the relative oscillation amplitude $\Delta \rho_{\text{out}} / \rho_{\text{avg}}$ is considerably large, reaching 100% near 23T at 1.6 K. A large SdH oscillation amplitude is consistent with a quasi-2D electronic structure. Furthermore, we also performed pulsed field measurements for fields up to 65T for $\rho_{\text{out}}$ and observed signatures of Zeeman splitting, as shown in Fig. S3, from which the Landé factor is estimated to 6.7 (see Note 3 in SI). In addition to SdH oscillations, we also observed dHvA oscillations in $\text{Sr}_{1-y} \text{Mn}_{1-z} \text{Sb}_2$ in magnetization measurements by a SQUID magnetometer. Type C samples exhibit the largest oscillation amplitude (up to 5 emu/mol near 6.5T) due to their relatively weak FM background, as shown in Fig. 1d and 1e.

The Fast Fourier transformation (FFT) analyses of the oscillatory resistivity $\Delta \rho_{\text{out}}$ (Fig. 1c) and $\Delta \rho_{\alpha\alpha}$ (Fig. S2b) show that the oscillation frequencies are $\sim 69.6$ T and 70.1 T, respectively. The field was about 20° misaligned relative to the [100] axis for both $\rho_{\text{out}}(B)$ and $\rho_{\alpha\alpha}(B)$ measurements. When the field was exactly aligned along [100], the frequency drops to 67T (see the inset to Fig. 1b). While the SdH oscillations show only a single frequency, the FFT spectra of the dHvA oscillations (Fig. 1f) reveal two frequencies, $F_{\alpha'} = 66.7$ T and $F_{\parallel} = 53.3$T. The inconsistency between SdH and dHvA oscillations is often seen in low dimensional materials such as layered organic conductors and originates from the different mechanisms of SdH and dHvA oscillations. SdH oscillations originate from the oscillating scattering rate and can thus be complicated by the detailed scattering mechanisms. This in some cases leads the observed resistivity oscillations to substantially deviate from the prediction by the Lifshitz-Kosevich (LK) theory, which is the case for $\text{Sr}_{1-y} \text{Mn}_{1-z} \text{Sb}_2$ as discussed below. By contrast, the dHvA effect is caused directly by the oscillations of electrons’ free energy and can be well described by the LK model for both 3D and 2D cases. As a result, the dHvA effects can
provide more direct information on Fermi surface (FS) for 2D-like materials such as Sr$_{1-y}$Mn$_{1-z}$Sb$_2$.

We evaluated the extremal cross-sectional area $A_F$ of the Fermi surface (FS) using the Onsager relation $F=(\Phi_0/2\pi^2)A$. The frequency of 67 T corresponds to $A_F = 0.64(0)$ nm$^2$, about one half of $A_F (=1.45$nm$^2$) probed in the same field configuration for SrMnBi$_2$.$^{26}$ Such a small value of $A_F$ indicates a small FS. Moreover, we also examined the dependence of $F$ on the magnetic field orientation angle $\theta$ via measuring SdH oscillations under various field orientations (Fig. S4). As shown in the inset to Fig. 1b, $F(\theta)$ can be fitted with a $F_0/cos\theta$ function, suggesting that the FS responsible for the SdH oscillations in Sr$_{1-y}$Mn$_{1-z}$Sb$_2$ is quasi-2D. A remarkable signature of quantum oscillations of a low-dimensional system is the large oscillation amplitude as noted above. The second frequency component $F_\beta$ probed in the dHvA oscillations suggests that the Fermi surface of Sr$_{1-y}$Mn$_{1-z}$Sb$_2$ is somewhat warped.

In general, the effective mass $m^*$ of quasi-particles on the Fermi pocket can be obtained from the fit of the temperature dependence of the FFT amplitude of the SdH/dHvA oscillations to the temperature damping factor of the LK equation, i.e. $R_T=\alpha T m^*/[m_0 B \sinh(\alpha T m^*/m_0 B)]$ where $\alpha = (2\pi^2 k_B m_0)/(he)$. However, as seen in the inset of Fig. 1c and Fig. S2c, the LK formula barely fits the SdH FFT amplitude data due to the steep increase of the oscillation amplitude below 7 K (see the black fitted curve), which is not surprising since the LK theory cannot precisely describe SdH oscillations in low-dimensional systems in many cases as indicated above. If the data points below 7K are not included in the fit, a better fit can be obtained (the red
fitted curve in the inset to Fig. 1c), which yields $m^* = 0.14m_0$. The effective mass obtained in such a way is rough since the fit does not cover all the data points. In contrast, accurate $m^*$ can be found from the fit of the temperature dependence of the dHvA FFT amplitude to the LK formula. As seen in the inset to Fig. 1f, all the dHvA oscillation FFT amplitude data points can be best fitted, which yields $m^* = 0.050m_0$ for $F_{\alpha'}$ and $0.043m_0$ for $F_{\beta}$, comparable with that of the gapless Dirac semimetal Cd$_3$As$_2$ ($m^* = 0.023$-0.044$m_0$), implying Sr$_{1-y}$Mn$_{1-z}$Sb$_2$ likely harbors relativistic fermions.

To seek further evidence for relativistic fermions in Sr$_{1-y}$Mn$_{1-z}$Sb$_2$, we examined the Berry phase $\phi_B$ accumulated along cyclotron orbits. For a Dirac/Weyl system, pseudo-spin rotation under a magnetic field should result in a non-trivial Berry phase, which can be accessed from the Landau level (LL) index fan diagram or the direct fit of the SdH/dHvA oscillation pattern to the LK formula. For a 2D or quasi-2D system with relativistic fermions, the intercept $n_0$ on the $n$-axis of the LL fan diagram is expected to be $1/2$, for which the corresponding Berry phase is $2\pi n_0 = \pi$. In Fig. 2d, we present the LL fan diagram established using the oscillatory conductivity $\Delta \sigma_{xx}$ data (Fig. 2c), which is obtained by subtracting the background from the conductivity $\sigma_{xx}$ (Fig. 2b). $\sigma_{xx}$ is converted from the longitudinal resistivity $\rho_{xx}$ and the transverse (Hall) resistivity $\rho_{xy}$ data collected on an identical type sample A#4 (Fig. 2a) using $\sigma_{xx} = \rho_{xx}/(\rho_{xx}^2+\rho_{xy}^2)$. Following the customary practice of defining LL index, we assigned integer LL indices to the minima of $\Delta \sigma_{xx}$ as illustrated in Fig. 2c and 2d. As seen in Fig. 2d, the intercept $n_0$ on the $n$-axis we obtained from the extrapolation of the best linear fit in the fan diagram is $0.55\pm0.02$, close to the expected value of 0.5 for a 2D or quasi-2D system with relativistic fermions. The frequency obtained from the fit is $69.8T$, only about 1% higher than
the frequency (68.9 T) obtained from the FFT analyses of $\sigma_{xx}$ (Fig. 2e), which should be considered as reliable metric for the linear fit in the fan diagram (note that the field is $\sim 5^\circ$ misaligned along [100] for $\rho_{xx}$ and $\rho_{xy}$ measurements shown in Fig. 2a). Therefore, our result of the intercept $n_0 = 0.55 \pm 0.02$ obtained on A#4 sample can be considered to be strong evidence of the $\pi$ Berry phase of relativistic fermions in $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$. This is further verified by the LL fan diagram analyses on another two samples (see Note 5 in SI) as well as by our direct fit of the dHvA oscillation pattern to the LK formula which takes Berry phase into account for a Dirac system (see the Method section). As seen in Fig. 2f, the Berry phases extracted from the dHvA oscillation pattern fit are $(0.45 \pm 0.06)\pi$ and $(0.8 \pm 0.07)\pi$, respectively, for the $F_{\alpha}'$ and $F_{\beta}$ components, which are clearly non-trivial. The sample difference in non-stoichiometry composition may lead to slight difference in Fermi surface morphology, which may explain the discrepancy in $\phi_B$ probed in these two different approaches. Additionally, the positive sign of $R_H$ (Fig. 2a) indicates the dominant charge carries responsible for quantum oscillations are hole-like.

Besides relativistic fermion behavior, $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$ exhibits FM properties as mentioned above, in contrast with the AMnBi$_2$ materials with Dirac/Weyl fermions$^{25,26,28,38}$, for which no ferromagnetism has been reported. To examine if the relativistic fermion transport is coupled with ferromagnetism in $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$, we have measured the magnetotransport properties of several groups of samples with different magnitudes of magnetic moment. Before we compare the quantum transport properties among those samples, we will first discuss the origin of ferromagnetism in $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$. In the top panel of Fig. 3a, we show the isothermal magnetization data at 5 K for three typical type A samples which exhibit relatively strong FM behavior with $M_s \sim 0.14 \text{-} 0.6 \mu_B$/Mn for the magnetic field along the [100] direction. We also
measured the temperature dependence of the magnetization and found the FM polarization occurs even at 400 K (Fig. S6 in SI). In order to understand the nature of such FM behavior, we performed single-crystal neutron diffraction measurements using a type A sample with $M_s \sim 0.2 \mu_B$ at 5 K. We found $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$ exhibits complex magnetic states. First, a long-range FM order with Mn moments along the $b$-axis occurs below $T_C=565$ K (Fig. 4b). Fig. 4a shows the temperature dependence of the FM scattering intensity that overlaps with the nuclear scattering intensity at the (200) reflection, from which a clear FM transition around 565 K can be seen, consistent with the FM polarization behavior up to 400K in the magnetization measurements (Fig. S6). The calculated FM ordered Mn moment along the $b$-axis at 350K is $\sim 0.28(6) \mu_B$/Mn. Second, the FM order parameter shows an unusual decrease below 300K, coinciding with the presence of a strong AFM order below 304 K, as seen in the temperature dependence of the (001) magnetic peak intensity in Fig.4c. Both the magnetic and nuclear structures are determined from the refinement of the neutron diffraction data collected at 5 K. A C-type AFM structure was found to best fit the data. In consideration of the existence of ferromagnetism below $T_N$ as observed from the magnetization and the (200) nuclear intensity being higher than that above 565 K, we conclude that the ground state in type-A sample should be a canted AFM state (Fig. 4d). The nearest-neighbor Mn spins with moments of $\sim 3.77(9) \mu_B$ are aligned antiparallel within the $bc$ plane and aligned parallel along the $a$-axis. The canting leads to a FM component along the $b$-axis, with the size of moment of $\sim 0.2 \mu_B$, as seen from the magnetization measurements. Details of the magnetic structure refinement are given in Note 7 in SI.

It is worth pointing out that the canted C-type AFM state probed in $\text{Sr}_{1,y}\text{Mn}_{1,z}\text{Sb}_2$ is analogous to the canted AFM state expected for $\text{YbMnBi}_2$ whose FM component is believed to
be responsible for the TRS breaking Weyl state\textsuperscript{25}, though recent neutron scattering experiments did not resolve such a canted AFM state in YbMnBi\textsubscript{2}\textsuperscript{39,40}. Given that Sr\textsubscript{1-y}Mn\textsubscript{1-z}Sb\textsubscript{2} shares a similar layered structure with YbMnBi\textsubscript{2} and exhibits coexistence of relativistic fermion and ferromagnetism as discussed above, there is a possibility that the FM component in this material might result in a TRS breaking Weyl state, which is yet to be clarified by further experimental and theoretical studies. As indicated above, one remarkable signature of a Weyl state is a chiral anomaly, which is manifested as negative LMR. For Sr\textsubscript{1-y}Mn\textsubscript{1-z}Sb\textsubscript{2}, we have indeed observed negative LMR in some type A samples with the current along the [100] direction (see Note 8 in SI). As shown in Fig. S8, the field and field-orientation dependences of the LMR, as well as the $B^2$ dependence of magnetoconductance, appear to be in line with the chiral anomaly effect. However, these phenomena have not been observed in in-plane magnetotransport measurements. Therefore, it is unclear whether the observed negative LMR is associated with chiral anomaly. Although Sr\textsubscript{1-y}Mn\textsubscript{1-z}Sb\textsubscript{2} shows FM properties, the negative LMR seen in our experiments should not be due to magnetic scattering, since the magnetoconductance continue to follows $B^2$ dependence even when the magnetization becomes saturated above 2T (Fig. S8c).

Next we compare the quantum transport properties of the samples with different FM components (Fig. 3b). As summarized in the top panel of Fig. 3c, the relative SdH oscillation amplitude exhibits a remarkable enhancement with the increase of the FM saturated moment $M_S$ for both $\rho_{xx}$ and $\rho_{out}$, from ~10\% for type C to ~25-45\% for type B, and ~20-60\% for type A samples. Coincidentally, the SdH oscillation frequency $F$ statistically decreases with increasing $M_S$, except for a large deviation of one data point collected on a type C sample which is likely caused by sample inhomogeneity (Fig. 3c, bottom panel). In general, disorder is expected to
suppress quantum oscillations, but the suppression of SdH oscillations in type B or C samples cannot be understood in terms of increased disorder. This is most clearly seen by examining the lack of correlations between the residual resistivity ratio RRR (which characterizes the disorder level) and the relative SdH oscillation amplitude as demonstrated in Fig. S9. In Sr_{1-y}Mn_{1-z}Sb_{2} the disorder induced by Sr and Mn vacancies are separated from the 2D Sb conduction layers and are expected to have only a weak effect on the in-plane cyclotron motion. This idea also accounts for the insensitivity of quantum oscillations to disorder in this material system.

Our observations of enhanced SdH oscillations in the samples with stronger ferromagnetism suggest an enhanced carrier mobility in those samples. The correlation of ferromagnetism and mobility in Sr_{1-y}Mn_{1-z}Sb_{2} might be understood in terms of the change in band structure driven by ferromagnetism. Indeed, this has been shown to be the case for YbMnBi_{2} by first principle calculations^{25}. Here, the spin degeneracy can be lifted by a FM component arising from the formation of a canted AFM state. The relative shift of the bands in the magnetically ordered state closes the SOC-induced gap leading to the formation of Weyl nodes. In Sr_{1-y}Mn_{1-z}Sb_{2}, our observation of a decrease in the quantum oscillation frequency \( F \) caused by enhanced ferromagnetism appears to support the gap-closing scenario due to TRS breaking. As seen in Fig. 3c, \( F \) decreases from \( \sim 72 \) T to \( \sim 67 \) T when \( M_s \) increases by two orders of magnitude from type C to A samples, which corresponds to a \( \sim 7\% \) shrinking of the cross-section area of FS. Such a decrease in the quantum oscillation frequency accompanied by the simultaneous increase of quantum mobility with increasing \( M_s \) implies the TRS-breaking plays a role in modifying band structure.
Finally, we discuss the inconsistency between our experimental observations and the early DFT calculation results\textsuperscript{29}. These calculations predict that the orthorhombic structural distortion in SrMnSb\textsubscript{2} causes the absence of a Dirac band crossing near Fermi level. Instead, the distortion is thought to cause a rather large gap (\(~1\text{eV}\)) to form at momentum points where Dirac crossing is expected. However, this gap is expected to close if the unit cell volume shrinks by 10\% as would under external pressure\textsuperscript{29}. We note that the structural parameters used in the DFT calculations\textsuperscript{29} were taken from a previous report\textsuperscript{31} and these lattice parameters differ slightly from our structural parameters probed by neutron scattering (see Table S1 in SI). In fact, the unit cell volume of our sample is 2\% less than that of the previously reported sample\textsuperscript{31}, with the difference most likely induced by the Sr and Mn deficiencies. The chemical pressure due to Sr and Mn vacancies, while not large enough to close the \(~1\text{eV}\) gap, can possibly lead to the closing of a smaller gap (\(~0.2\text{eV}\)) near Y points at the BZ boundary. Since the band dispersion near Y is quasi-linear\textsuperscript{29}, a Dirac band crossing at Y may occur when the gap closes due to chemical pressure and TRS breaking. Indeed, Dirac band crossing near Y was observed in ARPES measurements on YbMnBi\textsubscript{2}\textsuperscript{25}. Given the structural similarity between YbMnBi\textsubscript{2} and Sr\textsubscript{1-y}Mn\textsubscript{1-z}Sb\textsubscript{2}, it is reasonable to expect similar Dirac crossings near Y in Sr\textsubscript{1-y}Mn\textsubscript{1-z}Sb\textsubscript{2}. Although the DFT calculations did not give a result consistent with our experimental observations for SrMnSb\textsubscript{2}, its prediction of Dirac fermion behavior in tetragonal BaMnSb\textsubscript{2} at ambient pressure has been demonstrated in recent experiments\textsuperscript{30,41}. The Dirac fermions in BaMnSb\textsubscript{2} are also nearly massless (\(m^* \sim 0.050-0.052m_0\)). However, BaMnSb\textsubscript{2} does not display FM order above room temperature, instead ordering in a G-type AFM structure below 283K. This AFM order is in contrast with the striking FM properties seen in type A samples of Sr\textsubscript{1-y}Mn\textsubscript{1-z}Sb\textsubscript{2}. The coexistence of nearly massless Dirac fermions and ferromagnetism in Sr\textsubscript{1-y}Mn\textsubscript{1-...
\( _2 \text{Sb}_2 \) offers a unique platform for exploring the effect of TRS breaking on Dirac bands and seeking a possible magnetic Weyl state.

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Methods

Single Crystal Preparation

The $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$ ($y$ or $z < 0.1$) single crystals were synthesized using a self-flux method with a stoichiometric mixture of Sr, Mn and Sb elements. The starting materials were put into a small alumina crucible and sealed in a quartz tube in an Argon gas atmosphere. The tube was then heated to 1050 °C for 2 days, followed by a subsequently cooling to 400 °C at a rate of 3 °C/h. Plate-like single crystals as large as a few millimeters were obtained (Fig. 1a, inset). The composition and structure of these single crystals was examined using Energy-dispersive X-ray spectroscopy and X-ray diffraction measurements.

Magnetotransport and Magnetization Measurements

The magnetotransport measurements were performed with a four-probe method using 9T/14T Physics Property Measurement Systems and the 31 T resistive magnets at National High Magnetic Field Laboratory (NHMFL) in Tallahassee. The magnetoresistivity measurements up to 65 T were performed using a four-probe method in a capacitor-bank-driven pulsed magnet at the NHMFL in Los Alamos. The experimental data collected under pulse fields and at constant temperatures between 0.6K and 150K were recorded on a digitizer using a custom-designed high-resolution low-noise synchronous lockin technique running at 296 kHz. In order to minimize the Eddy-current heating caused by the pulsed magnetic fields, a small crystal with dimensions of 0.8 mm × 0.7 mm × 0.4 mm was used for measurement. Magnetic fields were applied along the [100] axis. AC current used were between 1 and 10 mA, depending on the sample temperature. The magnetization measurements were carried out using a SQUID magnetometer (Quantum Design).
Berry Phase determination by the fit of dHvA oscillation pattern to the LK formula

In addition to using the LL index fan diagram to determine Berry phase $\phi_B$, $\phi_B$ can also be obtained through the direct fit of the dHvA oscillation pattern to the multiband LK formula, in which the observed dHvA oscillations are treated as the linear superposition of two frequency ($F_\alpha'$ and $F_\beta$) oscillations. Each frequency oscillation can be described by the LK formula, which takes Berry phase into account for a Dirac system:

$$\Delta M \propto -B^{1/2} R_T R_D R_S \sin[2\pi (\frac{F}{B} - \gamma - \delta)],$$

where $R_T$ is the temperature damping factor mentioned in the text, $R_D = \exp(-\alpha T_{D} m^*/m_0 B)$ and $R_S = \cos(\pi g m^*/2m_0)$ are Dingle and spin reduction factors respectively. $T_D$ is Dingle temperature. The thermal and Dingle damping are both due to LL broadening, whereas the spin reduction factor is associated with Zeeman splitting. The oscillation of $\Delta M$ is described by the sine term with a phase factor $-\gamma - \delta$, in which $\gamma = \frac{1}{2} - \frac{\phi_B}{2\pi}$. The phase shift $\delta$, which is determined by the dimensionality of Fermi surface, is 0 and $\pm 1/8$, respectively for 2D and 3D cases. In our fit shown in Fig. 2d, $\delta$ is taken as zero, since the electronic band structure of Sr$_1$-$y$Mn$_{1-z}$Sb$_2$ is quasi-2D as discussed in the text. As shown in Fig. 2d, we fitted the dHvA oscillation pattern at 2K to the LK formula with two frequency components. To reduce the number of fitting parameters, we took the values of $m^*$, $F_\alpha'$ and $F_\beta$ obtained in Fig. 1f as fixed parameters.

To achieve the best fit, the FM background has to be precisely subtracted from the dHvA oscillation spectrum and this has to be done manually. Note that in the analyses of SdH
oscillations, “Igor” software was used to subtract the background and derive the FFT spectra (Fig. 1c). We also tried using “Igor” to analyze the dHvA data and found the background cannot be fully removed, so we chose to do manual background subtraction, which is quite successful as shown in Fig. 2e.

**Neutron scattering measurements**

Two relatively large $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$ ($y$ and $z < 0.1$) crystals with the mass of ~ 17 and 100 mg were investigated by neutron diffraction on HB-3A four-circle diffractometer (FCD) at the High Flux Isotope Reactor (HFIR), Oak Ridge National Laboratory, USA. One crystal was measured at 315 K above $T_N$ with a wavelength of 1.003 Å from the bent Si-331 to study the crystal structure in the absence of $\lambda/2$ contamination, whereas the other crystal was measured with a wavelength of 1.542 Å which contains ~1.4% $\lambda/2$ contamination (Si-220 monochromator in high resolution mode (bending 150)). The crystal and magnetic structure were investigated at 5 K and the order parameter of nuclear/FM (200) and AFM (001) Bragg peaks was measured. Data were recorded over a temperature range of $5 < T < 760$ K using a Closed Cycle Refrigerators (CCR) available at HB-3A. The SARAH representational analysis program was used to derive the symmetry allowed magnetic structures. All the neutron diffraction data were analyzed using Rietveld refinement program Fullprof suite.

**Data availability**

The authors declare that the main data supporting the findings of this study are available within this article and its Supplementary Information files. Extra data are available from the corresponding author upon reasonable request. See author contributions for specific data sets.
Reference for methods

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Author contributions

J.Y.L., J.H. and Q.Z equally contributed to this work. The single crystals used in this study were synthesized by J.Y.L. The magnetotransport measurements in 14 T PPMS was carried out by J.Y.L., D.J.A., Z.Q.M. and L.S. The high field measurements at NHMFL were conducted by J.H., D.G., S.M.A.R., I.C., L.S. and Z.Q.M. G.F.C., X.L., J.W. and W. A. P. contributed to x-ray structure characterization and crystal quality examination. J.H., J.Y. L. and Y.L.Z. performed magnetization measurements. Q.Z., H.B.C. J.F.D. and D.A.T. conducted neutron scattering experiments and analyses. M.J. and F.B. did pulse magnetic field measurements. J.Y.L., J.H., Y.L.Z and Z.Q.M conducted transport data analyses. All authors contributed to scientific discussions and read and commented on the manuscript. This project was supervised by Z.Q.M.
Figure captions

Figure 1 | Quantum oscillations in Sr$_{1-y}$Mn$_{1-z}$Sb$_2$. a, in-plane ($\rho_{xx}$) and out-of-plane ($\rho_{out}$) resistivity as a function of temperature, with $\rho_{xx}(300 K)/\rho_{xx}(2 K) \sim 29$ and $\rho_{out}/\rho_{xx} \sim 609$ at 2K; inset, an optical image of a large single crystal. b, the out-of-plane magnetoresistivity, $\Delta \rho_{out}/\rho_{out} = [\rho_{out}(B)-\rho_{out}(0)]/\rho_{out}(0)$, as a function of magnetic field at various temperatures (1.6, 3.0, 5.0, 7.0, 10.0, 12.0, 15.0, 20.0, 30.0, 40.0 and 50.0 K), which shows strong SdH oscillations. Inset: the angular dependence of the SdH oscillation frequency $F(\theta)$ with $\theta = 0^\circ$ corresponding to the field aligned along the out-of-plane [100] direction; $F(\theta)$ can be fitted to $F_0/cos\theta$, suggesting a quasi-2D Fermi surface. c, the FFT spectra of $\Delta \rho_{out}(B)$ at various temperatures (1.6, 3.0, 5.0, 7.0, 10.0, 12.0, 15.0, 20.0, 30.0, 40.0 and 50K). $F_{2a}$, $F_{3a}$, $F_{4a}$ and $F_{5a}$ represent harmonic peaks. The inset shows the temperature dependence of the FFT amplitude normalized by $\rho_{out}(B=0)$. The solid black and red curves represent the fits to the LK formula in the 2-50 K and 7-50 K temperature ranges respectively, which do not appear to be satisfactory (see text). d, isothermal magnetization at various temperatures (2.0, 3.5, 5.0, 7.5, 10.0, 12.5 and 15.0 K), showing strong dHvA oscillations. e, oscillatory component of magnetization $\Delta M$ at various temperatures (2.0, 3.5, 5.0, 7.5, 10.0, 12.5 and 15.0 K). f, FFT spectra of $\Delta M$ at various temperatures (2.0, 3.5, 5.0, 7.5, 10.0, 12.5 and 15.0 K). The inset shows the fits of the temperature dependence of the dHvA FFT amplitudes to the LK formula from which the quasi-particles are found to be nearly massless.

Figure 2 | Non-trivial Berry phase of realistic fermions in Sr$_{1-y}$Mn$_{1-z}$Sb$_2$. a, the in-plane longitudinal ($\rho_{xx}$) and the transverse (Hall) ($\rho_{xy}$) resistivity as a function of magnetic field measured at 2K on an identical sample A#4; both $\rho_{xx}$ and $\rho_{xy}$ show strong SdH oscillations. b, the in-plane conductivity $\sigma_{xx}$ converted from the $\rho_{xx}$ and $\rho_{xy}$ data shown in a using $\sigma_{xx} =$
\[ \rho_{xx}/(\rho_{xx}^2 + \rho_{yx}^2) \] for sample A#4. c, the in-plane oscillatory conductivity \( \Delta \sigma_{xx} \) vs. \( 1/B \) at 2K. d, the Landau Level (LL) index fan diagram derived from the oscillatory conductivity at 2K of sample A#4. The integer LL indices \( n \) are assigned to the minima of \( \Delta \sigma_{xx} \). The intercept \( n_0 \) on the \( n \)-axis obtained from the extrapolation of the best linear fit in the fan diagram is 0.55±0.02, suggesting the \( \pi \) Berry phase accumulated in the cyclotron orbits. e, FFT spectra of the in-plane conductivity \( \sigma_{xx} \) at 2K for sample A#4. f, the fit of the dHvA oscillation pattern at 2 K to the LK formula (see the method section) for sample C#1, from which non-trivial Berry phases are also obtained.

Figure 3 | Coupling between magnetism and quantum transport properties in Sr\(_1-y\)Mn\(_1-z\)Sb\(_2\). a, field sweeps of magnetization at 5K for type A samples (showing significant FM behavior, see the upper panel), type B samples (showing relatively weak FM behavior, see the middle panel), and type C samples (displaying very weak FM behavior, see the bottom panel). All these samples exhibit dHvA oscillations. dHvA oscillations of type A and B samples are observable only when the data are zoomed in (see Fig. S5d). b, SdH oscillations of \( \rho_{xx} \) (upper panel) and \( \rho_{out} \) (lower panel) of type A, B, and C samples. All these data were collected in the 14T PPMS system and the fields were roughly aligned along [100] axis, with the misalignment angle < 10°; the relative shift of the oscillation pattern between samples is due to the field misalignment. c, the relative SdH oscillation amplitude near 12.5T (upper panel) and the SdH oscillation frequency (lower panel) as a function of the FM saturation moment \( M_s \) for type A, B, and C samples. The error bars of the frequencies are obtained from the linear fits of the LL fan diagrams, representing the uncertainties of the fitted slopes. The error bars of the SdH oscillation amplitudes represent the differences between the peak and valley amplitudes near 12.5T, about
10% of the averaged amplitudes. A general trend of increased oscillation amplitude and decreased oscillation frequency with increasing $M_s$ can be observed, as denoted by the dashed lines.

**Figure 4** | **Magnetism of Sr$_{1-y}$Mn$_{1-z}$Sb$_2$.**

- **a**, the temperature dependence of the neutron diffraction intensity at (200) obtained with a counting time of 10 minutes per point. The error bar uses the standard deviation, *i.e.*, square root of the total counts. Inset, the FM magnetic structure of the Mn sub-lattice, viewed from the (101) direction. The Mn moments are aligned along the $b$-axis.

- **b**, the magnetic structure of the FM state for $304 \text{ K} < T < 565 \text{ K}$.  

- **c**, temperature dependence of the AFM order parameter (*i.e.* the (001) magnetic peak intensity). The error bars are smaller than the symbols so that they cannot be viewed on this scale. Inset, the canted AFM magnetic structure of the Mn sub-lattice, viewed from the (101) direction. A net FM component is due to the canting of magnetic moment along the $b$-axis.

- **d**, the C-type AFM magnetic structure in the AFM state for $T < 304 \text{ K}$. 

Figure 1
Figure 2
Figure 3
Figure 4