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Influence of magnetism on Dirac semimetallic behavior in nonstoichiometric $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$ ($y \sim 0.07, z \sim 0.02$)

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Quasi-2D magnetism and origin of the Dirac semimetallic behavior in nonstoichiometric $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$ ($y, z < 0.1$)

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Nonstoichiometric $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$ ($y, z < 0.1$) is known to exhibit a coexistence of magnetic order and the nontrivial semimetallic behavior related to Dirac or Weyl fermions. Here, we report inelastic neutron scattering analyses of the spin dynamics and density functional theory studies on the electronic properties of $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$. We observe a relatively large spin excitation gap ≈ 8.5 meV at 5 K, and the interlayer magnetic exchange constant only 2.8 % of the dominant intralayer magnetic interaction, providing evidence that $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$ exhibits a quasi-2D magnetism. Using density functional theory, we find a strong influence of magnetic orders on the electronic band structure and the Dirac dispersions near the Fermi level along the Y-S direction in the presence of a ferromagnetic ordering. Our study unveils novel interplay between the magnetic order, magnetic transition, and electronic property in $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$, and opens new pathways to control the relativistic band structure through magnetism in ternary compounds.

Topological semimetals [1, 2] are a newly emerged frontier in condensed matter physics and have stimulated tremendous research interest because they give access to new quantum phenomena and are very attractive for both fundamental research and technological application. Particular attention has focused on Weyl or Dirac semimetals that exhibit a coexistence with magnetism as a promising route to modify and control Weyl/Dirac fermions, electronic transport properties and band topology, among which SrMnSb_2 has attracted much interest recently. Liu *et al.* [3] reported a nontrivial semimetallic behavior related to Dirac or Weyl fermions including nearly massless quasiparticles with a π Berry phase coupled to ferromagnetism in nonstoichiometric $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$. For the magnetic behavior, it displays ferromagnetic (FM) order below $T_C \sim 565$ K, followed by a transition to canted antiferromagnetic (AFM) order with a net FM component below $T_{FM-AFM} \approx 304$ K [3]. The nontrivial topological semimetal behavior was further supported by optical conductivity and ultrafast optical pump-probe measurements [4]. Nevertheless, Ramankutty *et al.* [5] reported zero Berry phase indicative of trivial topology in nearly stoichiometric SrMnSb_2 . Previous density functional theory (DFT) calculations [5–7] showed that the lattice distortion in the orthorhombic structure prevents the formation of the Dirac points near the Fermi level by opening a gap. Both results cannot account for the observed topological semimetallic behavior reported in Ref. [3]. It is therefore challenging to explore whether there are indeed Dirac/Weyl points in proximity to the Fermi level and what drives their formation in $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$.

Furthermore, SrMnSb_2 offers a wonderful opportunity to address an important question whether there is a close correlation between magnetic order and band topology in 3D Dirac compounds.

In addition to SrMnSb_2 , other Alkaline earth ternary AMnC_2 “112” compounds ($A = \text{Sr, Ca, Ba}$, $C = \text{Bi or Sb}$) [8–12] were reported to be Dirac semimetal candidates with the coexistence of AFM order. An interplay between magnetic order and electronic transport properties was found in CaMnBi_2 [13] due to coupling of the interlayer ferromagnetic component to the planar Bi electrons. The Dirac carriers in Bi layers were reported to enhance the interlayer exchange coupling J_\perp significantly between magnetic layers in AMnBi_2 ($A = \text{Ca, Sr}$) by the Raman spectroscopy [14]. However, Rahnet *et al.* [8] argued that the neglect of single-ion anisotropy D in the Raman analysis may significantly exaggerate the obtained interlayer magnetic coupling since D and J_\perp are correlated. All of these facts emphasize the importance of an accurate determination of the interlayer magnetic coupling and the magnetic dimensionality in “112” compounds.

Here, by a combination of inelastic neutron scattering and linear spin wave theory, we report the magnetic excitation spectra and determination of the accurate magnetic exchange couplings and the single-ion anisotropy, which evidences a quasi-2D magnetism in $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$. More interestingly, we found a strong coupling between various magnetic orders and the relativistic band structures near the Fermi level by density functional theory (DFT), which reveals that FM order with the moment along b axis induces the Dirac

points near the Fermi level in the band structure whereas various AFM orders have a disfavoring effect on it in $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$.

$\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$ crystals were grown using a flux technique [3]. Several single crystals with a total mass of approximately 600 mg were co-aligned at the (0 *K* *L*) horizontal scattering plane within ~ 3 degrees mosaicity. Inelastic neutron measurements were performed using the Spallation Neutron Source's SEQUOIA spectrometer with its high-flux mode at Oak Ridge National Laboratory. The data were collected at 5 K and 350 K using a few different incident energies of 35, 70, 100, 160, 200 meV. The wave vectors \mathbf{Q} reported here are defined in reciprocal lattice unit (rlu). The constant-energy (*E*) cuts were fitted using a Lorentz function to obtain both the spin wave dispersion and intensity. The fits to spin wave dispersion and intensity using the SpinW package [15] yield the magnetic exchange constants and single-ion anisotropy. DFT was performed using the generalized gradient approximation and projector augmented wave approach [16] as implemented in the Vienna *ab initio* simulation package (VASP) [17, 18].

Figure 1(a) shows the crystal and magnetic structures in $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$. It crystallizes in the orthorhombic structure with space group *Pnma* (No. 62), consisting of a MnSb layer with edge-sharing MnSb(2)₄ tetrahedral and flat Sb(1) layer sandwiched between two staggered Sr planes. Note that all the Sb(1) square net, Sr and MnSb(2)₄ are distorted, different from the tetragonal structure in Bi-based “112” compounds. The two magnetic structures previously determined [3] for $T < T_{\text{FM-AFM}}$ and $T_{\text{FM-AFM}} < T < T_C$ are illustrated in Fig. 1 (a). Fits to the order parameter of the magnetic Bragg peak (001) in Fig. 1 (b) to a power law of the form $I \propto (T_N - T)^{2\beta}$ yields a critical exponent $\beta \approx 0.25$. Such a β value falls in the region $0.1 < \beta < 0.25$ expected for quasi-2D systems [19] (compared to the 0.36 in the 3D Heisenberg model) and is similar to that in typical quasi-2D pnictide LnMnSbO (Ln=La or Ce) [20]. This implies that $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$ may be magnetically quasi two-dimensional. The field dependence of the magnetization in the inset of Fig. 1 (b) confirms a clear ferromagnetism below $T < T_C$ down to 5 K.

To investigate the spin dynamics of $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$, we performed inelastic neutron scattering measurements. Figure 1 (c,e) and (d,f) compares the magnetic excitations near the AFM zone center (0 0 1) at 5 K ($T < T_{\text{FM-AFM}}$) and 350 K ($T_{\text{FM-AFM}} < T < T_C$). At 5 K within the AFM ordered state, a clear spin gap $E_g \approx 8.5$ meV is observed indicative of the existence of a single-ion anisotropy. The spin gap closes at 350 K when the long-range AFM order disappears. Another difference between these two temperatures is that, whereas the spin wave dispersion exists at 5 K, the dispersion disappears, and evolves into a spin fluctuation spectra near the AFM zone centers at 350 K. The magnetic excita-

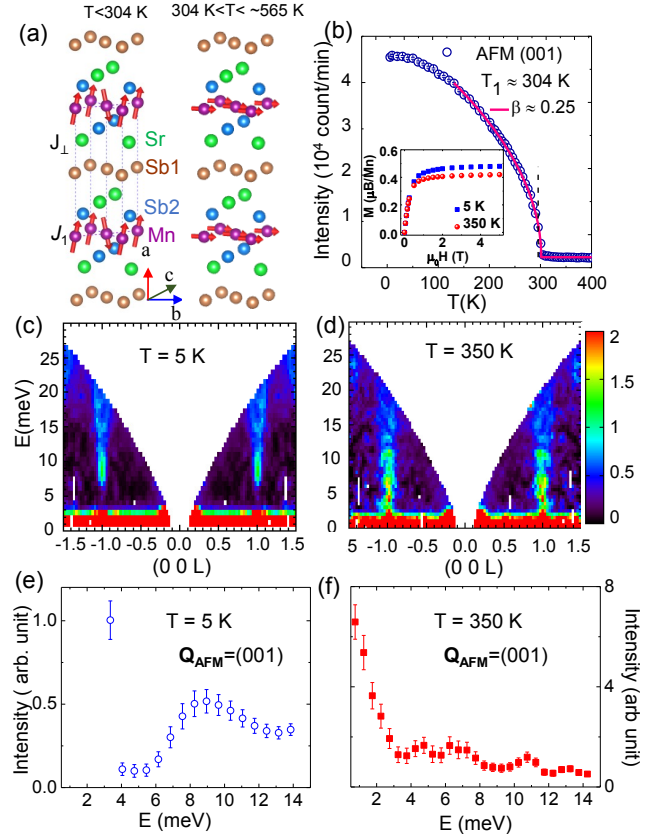


FIG. 1: (color online) (a). Magnetic structures of $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$: canted AFM order in $T < 304$ K and FM order with moment along *b* axis in $304 \text{ K} < T < \sim 565$ K. (b). Temperature dependence of the peak intensity for the pure AFM Bragg peak (001) taken from Ref. [3]. The solid line is the fit to the powder law. Inset shows the field dependence of the magnetization at different temperatures. Magnetic excitations near AFM zone center (0 0 ± 1) at (c) 5 K and (d) 350 K. The corresponding constant-*Q* cuts at 5 K and 350 K are shown in (e) and (f), respectively.

tions along out-of-plane *H*, in-plane *K*, *L* and diagonal [0 *K* *K*] directions at 5 K are displayed in Fig. 2 (a-d) (the high-symmetric brillouin symbols are illustrated in the inset of Fig. 4 (a)). There is a steep dispersion along in-plane directions extending to ≈ 70 meV at the AFM zone boundary Z and T points, but the dispersion along out-of-plane *H* direction is much weaker, with $E \approx 18$ meV at the zone-boundary X point. This indicates that the out-of-plane magnetic interaction is much weaker than the in-plane one suggesting a quasi-2D magnetism.

Figure 3 compares constant-energy slices in the (0 *K* *L*) scattering plane at energy transfers around 5, 17 and 30 meV at 5 and 350 K. At 5 K, no magnetic excitations are apparent around 5 meV in Fig. 3(a) owing to the existence of higher spin gap of $E_g \approx 8.5$ meV. As the transferred *E* increases, a ring of scattering emerges at AFM zone center positions such as (0 0 ± 1) and (0 ± 1 0). The diameter of the rings increases with increas-

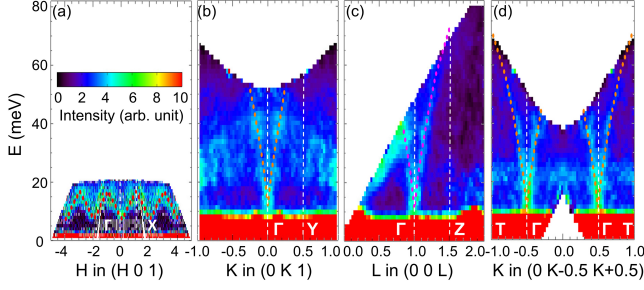


FIG. 2: (color online) Magnetic excitations near AFM zone center (0 0 1) at 5 K, along high-symmetry directions: (a) out-of-plane H , in-plane (b) K , (c) L and (d) diagonal $[0 K K]$ directions. The dashed lines are fits to the experimental spin wave dispersions. The vertical lines indicate the AFM zone center or boundaries, with the brillouin symbols marked in the figures.

ing the E transfer (see Fig. 3(b) and (c)), indicative of dispersive spin waves. In sharp contrast, the magnetic excitations exhibit different features at 350 K. A diffuse magnetic excitations can be seen in Fig. 3(d) at around 5 meV due to the closure of the spin-gap. As E transfer increases, the magnetic excitations become more diffuse and spread out without the ring-like feature, as shown in Fig. 3 (e) and eventually evolve to be hardly visible at high E transfer region in Fig. 3 (f). This indicates the existence of the low- E AFM spin fluctuations at 350 K. It is worthwhile pointing out that we do not observe clear spin-wave branches associated with FM ordered phase at 350 K, which is understood due to the low FM moment $\approx 0.41 \mu_B$ (see inset of Fig. 1 (b) and small mass of the coaligned crystals (≈ 600 mg)).

To analyze the observed spin waves and quantitatively determine the magnetic interactions in $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$, we have performed linear spin wave calculations using the SpinW package [15] for the following spin Hamiltonian:

$$H = \sum_{i,j} S_i J_{ij} S_j + \sum_{\alpha,\beta,i} S_i^{\alpha} A_i^{\alpha,\beta} S_i^{\beta} \quad (1)$$

where S_i are spin vector operators, J_{ij} are pair coupling between spins and $A_i^{\alpha,\beta}$ are 3×3 anisotropy matrices. By fitting to the experimental SW dispersions and intensities, we obtain the AFM nearest-neighbor (NN) $J_1 \approx 9.3(3)$ meV, interlayer FM $J_{\perp} \approx -0.26(8)$ meV, and anisotropy parameter $A_i = \text{diag}(0 \ 0.12(5) \ 0.3(1))$. Here we enumerate the key results from the SW fitting: 1). The NN J_1 is dominant and the next-nearest-neighbor (NNN) J_2 is found to be negligible, i.e., $J_2/J_1 < 1/2$. Together with FM J_{\perp} , this could account for the formation of the overall C -type AFM order [8, 20, 21]. Furthermore, the AFM NN J_1 and FM out-of-plane J_{\perp} are consistent with the associated AFM and FM spin arrangements (see Fig. 1 (a)), respectively. All these results indicate there is no signature of strong spin frustration.

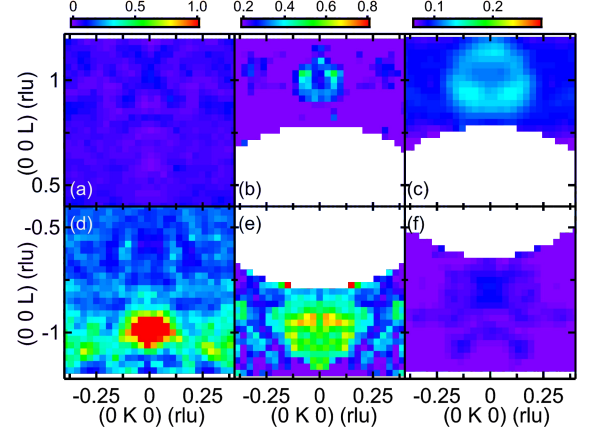


FIG. 3: (color online) Constant- E slices in (0 K L) scattering plane around E of (a) 5 meV, (b) 17 meV and (c) 30 meV at 5 K. Corresponding constant- E slices at 350 K are shown in (d-f).

2). The interlayer J_{\perp} , which characterizes the dispersion along the out-of-plane a axis, is less than 2.8 % of the in-plane J_1 , signaling the quasi-2D magnetism. The J_{\perp} is considerably weaker than that proposed in *Ref.* [14] on AMnBi_2 ($A=\text{Ca}, \text{Sr}$) (with similar interlayer Mn-Mn distances), which was claimed as enhancement due to the Dirac carrier Bi layers. 3). The emergence of the spin gap is ascribed to the uniaxial single-ion anisotropy matrices $A_i^{\alpha,\beta}$.

To gain insight into the experimental observations, we performed DFT calculations for stoichiometric SrMnSb_2 using the high-temperature structure at 315 K and the low-temperature one at 5 K [3]. For Sr, a potential, in which semi-core s and p states are treated as valence states, is used (Sr_{sv}), and for Mn and Sb, standard potentials were used (Mn and Sb, respectively, in the VASP distribution). In most cases, we use a $2 \times 8 \times 8$ \mathbf{k} -point grid and an E cutoff of 500 eV with spin-orbit coupling included. The $+U$ correction is not included because SrMnSb_2 is an itinerant magnetic system.

We first investigate the magnetic properties at high temperature. Using the 315 K structure, we compute the total E of several symmetry-allowed magnetic configurations [3], including C -type AFM (CAFM), G -type AFM, A -type AFM, and FM with spin orientation taken along the a , b or c axis (the spin orientation will be indicated as a subscript as for example CAFM_a). It turned out that CAFM_a is most stable, while the FM_b state is more stable than FM_a and FM_c . This discrepancy is resolved by constrained magnetization calculations. As shown in Fig. 4 (a), the relative E between FM_b and CAFM_a is reversed when the ordered Mn moment M_s is suppressed at around $M_s = 1.6 \mu_B$. In the PM state ($M_s = 0$), the density of states (DOS) has a sharp peak near the Fermi level ($E = 0$) as shown in Fig. 4 (b). Thus, the leading

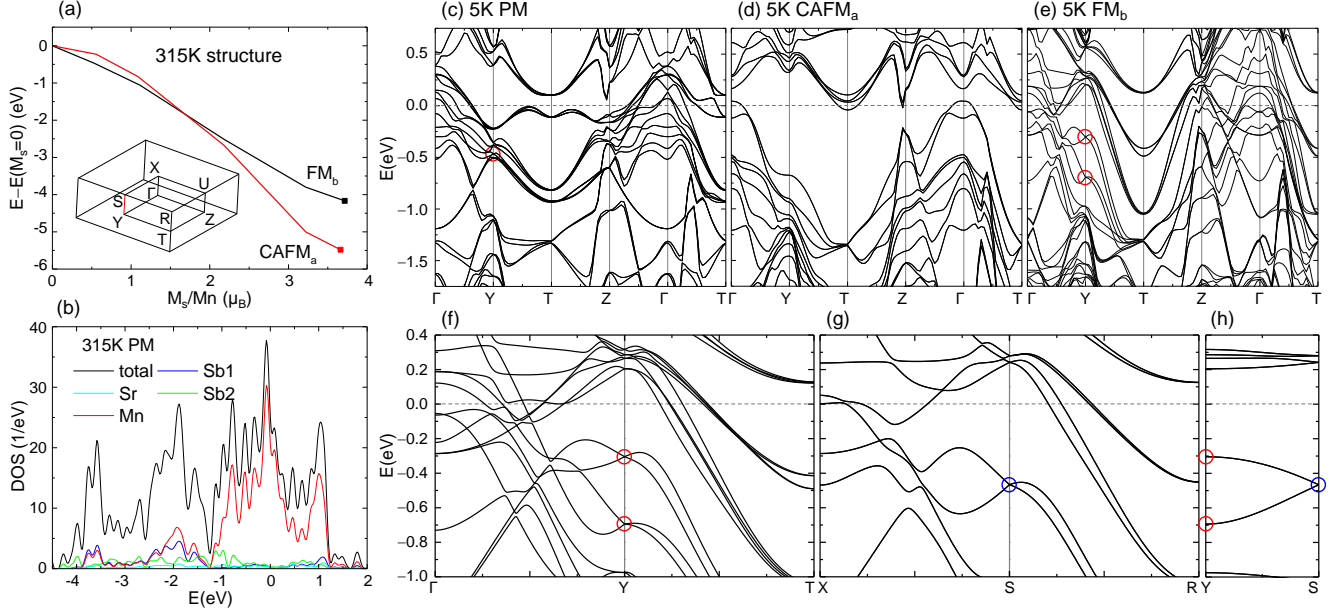


FIG. 4: Density functional theory results. (a) Total E as a function of ordered moment M_s on a Mn site for FM_b and CAFM_a . Squares indicate equilibrium positions. (b) Density of states in the PM phase. Dispersion relation along high-symmetry lines for (c) PM, (d) CAFM_a , and (e) FM_b . Dirac dispersions are indicated by circles in (c) and (e). (f)-(h) are the magnified view of the Dirac dispersions in the (e) FM_b state along different directions, showing the evolution of two non-degenerate Dirac cones (red circles) at the Y point merge to a twofold degenerate Dirac cone at the S point (blue circle). The results in (a) and (b) are obtained using the high-temperature structure at 315 K, while (c-h) using the low-temperature structure at 5 K. The inset of (a) shows the 1st Brillouine zone, with the Dirac line node along the Y-S direction indicated by a red line. Energy in (b)-(h) is measured from the Fermi level.

instability in the high-temperature PM phase is toward FM ordering due to the Stoner instability, followed by the first-order transition from FM_b to CAFM_a at lower temperature with much larger M_s ($\approx 3.6\mu_B$ at 5 K [3]). This provides a natural explanation for the sequence of magnetic transitions reported from experiments [3].

Now we turn to the low-temperature electronic property using the structural information at 5 K. We computed the total energy of several magnetic configurations using the low-temperature structure at 5 K. We found it is robust that the in-plane (out-of-plane) exchange is AFM (FM) and the single ion anisotropy is uniaxial [22], which indicates that the CAFM_a state is most stable, and the FM_b is metastable at 5 K. It is insightful to examine the electronic band structures of the PM, the CAFM_a and the FM_b states, as shown in Figure 4(c), (d), and (e), respectively. Because of the non-symmorphic symmetry, Dirac cones are expected at the Y and the T points [5, 23] (X and M points in the notation of Ref. [5]). In the PM state, the Dirac cones are observed at $E \sim -0.5$ eV at the Y point, but those at the T point are not clearly resolved in this E window. The band structure is influenced by magnetic order. In the CAFM_a state, the Dirac cone at the T point is clearly seen at $E \sim -1.3$ eV. This dispersion relation is consistent with the one reported in Refs. [5, 6] except for the current semimetallic behavior

due to the difference in the exchange correlation potential or local U . As discussed in Ref. [5], the location of the Dirac cone is too far from the Fermi level to account for the π Berry phase reported in Ref. [3]. Interestingly, when the metastable FM_b state is considered, the Dirac cone appearing in the PM state at the Y point splits due to the spin polarization, and one of them becomes closer to the Fermi level ($E \sim -0.3$ eV) [Fig. 4 (e)]. As shown in Fig. 4 (f)-(h), these Dirac cones form a line node along the Y-S direction, and two Dirac cones merge at the S point. This is a consequence of two units of MnSb_2 and Sb(1) layers, each of which supports two-dimensional Dirac cones, and the mixing between the two units is suppressed at $k_x = \pi$.

Once the Fermi level is tuned near the Dirac cones, the π Berry phase should be manifested in the quantum oscillations of magnetoresistance or magnetization. A natural question is how one can realize this condition in SrMnSb_2 . In reality, the π Berry phase is observed only in non-stoichiometric $\text{Sr}_{1-y}\text{Mn}_{1-z}\text{Sb}_2$ samples in Ref. [3]. Thus, the current study suggests the following scenario. The relative energy between the CAFM_a and FM_b in non-stoichiometric samples could be much smaller than that in stoichiometric samples. As a result, the FM_b state could remain in place more easily at low temperatures, resulting in either canted AFM order [3] or possi-

ble phase separation between collinear AFM_a and FM_b phases. The FM state induces the splitting of Dirac cone at Y point with one of them being closer to the Fermi level. Moreover, the Fermi level is expected to be lowered by Sr and/or Mn off-stoichiometries to locate near Dirac cones. In order to verify this scenario, it is necessary to control the defect density and magnetism, and isolate FM regimes from others.

In summary, we have examined the magnetic excitations, and investigated the origin of the Dirac semimetallic behavior in nonstoichiometric Sr_{1-y}Mn_{1-z}Sb₂. The magnetic exchange constants are determined, indicative of a quasi-2D magnetism, with no significant enhancement of J_{\perp} by the Dirac carrier Sb layers. The constrained magnetization calculations by our DFT interpreted the occurrence of the successive PM-FM-AFM transition. We further demonstrated that while the AFM order does not favor the formation of the Dirac cones near the Fermi level, the FM order/component plays a key role in inducing the Dirac points in proximity to the Fermi level to drive the system to be Dirac semimetal in Sr_{1-y}Mn_{1-z}Sb₂. Our study provides a new clue to the understanding of the origin of Dirac semimetals and to seek for novel Dirac semimetals by adjusting the magnetic order.

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