Competing magnetic energy scales in the topological flat-band ferrimagnet TbMn$_6$Sn$_6$

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TbMn$_6$Sn$_6$ is a metallic ferrimagnet displaying signatures of Weyl-semimetallic behavior and topological magnons arising from flat-band magnetism and spin-orbit coupling within its Mn kagome layers. Inelastic neutron scattering measurements find strong ferromagnetic (FM) interactions within the Mn kagome layer and reveal a magnetic bandwidth of \(\sim 230\) meV. The low-energy magnetic excitations are characterized by strong FM Mn-Mn and antiferromagnetic (AF) Mn-Tb interlayer magnetic couplings. We observe weaker, competing long-range AF and AF Mn-Mn interlayer interactions similar to those driving helical magnetism in the YMn$_6$Sn$_6$ system. Combined with density-functional theory calculations, we find that competing Mn-Mn interlayer magnetic interactions occur in all RMn$_6$Sn$_6$ compounds with \(R = \text{Y, Gd–Lu}\), resulting in magnetic instabilities and tunability when Mn-R interactions are weak. In the case of TbMn$_6$Sn$_6$, strong AF Mn-Tb coupling ensures a highly stable three-dimensional ferrimagnetic network.

I. INTRODUCTION

The potential technological applications of magnetic topological insulators and Weyl semimetals has generated new research directions aimed at understanding the coupling between magnetism and topological fermions. This has brought renewed interest in magnetic kagome materials, such as Mn$_3$Ge [1, 2], Fe$_2$Sn$_2$ [3, 4], Co$_3$Sn$_2$S$_2$[5–7] and FeSn [8], where both magnetism and topological electronic band crossings are hosted in the kagome layer. Interesting topological responses, such as large anomalous Hall conductivity, are tied to the underlying magnetic order that can be impacted by both geometrical frustration and Dzyaloshinskii-Moriya (DM) interactions. In principle, these materials may host topological magnons in the presence of DM interactions [9], opening up even more interesting avenues for the study of topological phenomena in metallic kagome systems.

The hexagonal RMn$_6$Sn$_6$ (R166) compounds (\(R = \text{rare-earth}\)) consist of alternating Mn kagome and \(R\) triangular layers. These materials display a variety of magnetic structures, including antiferromagnetic (AF), ferrimagnetic, and complex helical ordering, that are dependent on the nature of the host \(R\) ion [10–18]. While magnetic complexity is often encountered in geometrically frustrated kagome and triangular layers with AF interactions, in R166 the intralayer Mn-Mn interactions are strongly ferromagnetic (FM). Magnetic instabilities to temperature and applied magnetic fields in R166 compounds [19–22] arise from a combination of competing Mn and \(R\) magnetic anisotropies (for moment-bearing \(R\)-ions) and competing interlayer magnetic interactions [23–26]. The latter case is more apparent for non-moment-bearing \(R = \text{Y and Lu}\) ions.

Contemporary studies of R166 compounds have focused on the interplay between magnetism and topological electronic band crossings that are hosted in the kagome layer. For example, Y166 is an easy-plane antiferromagnet where competition between FM and AF interlayer couplings drives complex helical magnetic phases possessing net chirality and a topological Hall response [12, 14, 21, 22]. In Tb166, collinear ferrimagnetism with uniaxial out-of-plane anisotropy has realized Chern-gapped topological fermions with a quantized magneto-transport response [27]. These discoveries demonstrate great potential for novel topological phenomena to be discovered by exploring other R166 materials via rare-earth engineering [28] or the application of symmetry-breaking external fields.

There are still several fundamental open questions regarding the magnetism within R166 compounds. For example, is the Mn magnetism of an itinerant or local-moment nature and are the Mn-Mn interactions transferrable across the R166 materials? What is the variability of \(R\)-Mn interactions and \(R\) anisotropy across the series? Answering these questions will allow us to understand the potential tunability of R166 compounds for accessing different magnetic and topological states.

Here, we address the magnetic interactions in Tb166 in detail using inelastic neutron scattering (INS) and density-functional theory (DFT) calculations. We observe a hierarchy of competing interlayer Mn-Mn interactions in Tb166 similar to those used to explain the complex temperature- and field-driven helimagnetism observed in Y166 [12, 14, 21, 22]. We find that strong uniaxial Tb magnetic anisotropy and AF coupling between Mn and Tb layers generates a rigid three-dimensional ferrimagnetic lattice. Results of our DFT calculations largely agree with the sign, magnitude and overall hierarchy of interlayer couplings found experimentally after the introduction of on-site Coulomb repulsion (DFT+U).

The INS data also show that FM intralayer Mn-Mn interactions in both Tb166 and Y166 (Ref. [29]) are comparably strong and push the overall magnon bandwidth up to \(\sim 230\) meV. However, increasingly broad lineshapes for Tb166 do not allow the observation of magnetic exci-
tions above ~ 125 meV. Unlike reports of a gap appearing in the magnon spectrum at K-point in Y166 caused by DM interactions [29], this severe line broadening in Tb166 obscures any evidence of gapping. This suggests that, despite our quantitative modeling of the spin-wave spectrum presented here, there is still much to be learned about the itinerant character of Mn magnetism and the role of spin-orbit interactions in R166 materials.

II. EXPERIMENTAL DETAILS

Single crystals of Tb166 were grown from excess Sn using the flux method. A nominal (TbMn$_6$)$_5$Sn$_{25}$ molar ratio of elemental Tb (Ames Laboratory 99.9%), Mn (Research Organic/Inorganic Chemical Corp, 99.995%), and Sn (Alfa Aesar, 99.99%) was weighed and loaded into the crucible side of a 5 mL fritted alumina crucible (Research Organic/Inorganic Chemical Corp, 99.995%), and then heated to 1180°C in 12 hours. After dwelling at 1180°C for 3 hours, the furnace was slowly cooled over 300 hours to 575°C. Upon reaching the final temperature, the tube was rapidly removed from the furnace, inverted into a metal centrifuge, and the excess flux decanted. The crucibles were opened to reveal the furnace, inverted into a metal centrifuge, and the excess flux decanted. The crucibles were flame sealed under vacuum into the growth side of a 5 mL fritted alumina crucible (Research Organic/Inorganic Chemical Corp, 99.995%), and then heated to 1180°C in 12 hours. After dwelling at 1180°C for 3 hours, the furnace was quickly cooled in 3 hours to 775°C and then slowly cooled over 300 hours to 575°C. Upon reaching the final temperature, the tube was rapidly removed from the furnace, inverted into a metal centrifuge, and the excess flux decanted. The crucibles were opened to reveal large (up to 300 mg), shiny, hexagonal crystal plates (see Fig. 1(a)).

Low temperature magnetization was measured using a Quantum Design Magnetic Property Measurement System (MPMS 3), SQUID magnetometer (T = 1.8 – 300 K, $H_{\text{max}}$ = 70 kOe). A Tb166 single crystal sample was mounted on a plastic disc and the field was applied along c. Prior to measuring the sample, the blank disc was measured and used for a background subtraction. Figure 1(a) shows low temperature magnetization measured at 2 K with H || c that accurately reproduce the previously reported hysteresis loop displaying a saturated magnetization of $\approx 4 \mu_B/\text{f.u.}$ [31].

Tb166 crystallizes in the HfFe$_6$Ge$_6$-type structure with hexagonal space group $P6/mmm$ (No. 191) and Mn, Sn1, Sn2, Sn3 and Tb ions, respectively, sitting at the 6i, 2e, 2d, 2c and 1b Wyckoff positions [32], see Fig. 1(b-d). From a Rietveld analysis of XRD data collected at 300 K (see Fig. 1(b)), we obtain refined values of 5.53317(6) and 9.0233(1) Å for lattice parameters a and c, as well as atomic coordinates $z_{\text{Mn}} = 0.2539(2)$ and $z_{\text{Sn1}} = 0.1624(2)$, in close agreement with previous reports [13]. Below 423 K, both the Mn and Tb layers simultaneously develop FM order, but couple antiferromagnetically, resulting in an overall ferrimagnetic order. All magnetic moments initially lie in the basal plane but remarkably, a spin re-orientation takes place as the system is further cooled between ~330 and ~250 K, resulting in the ground state collinear ferrimagnetic arrangement of Mn and Tb moments along the c-axis [13, 15] shown in Fig. 1(c) and 1(d).

INS measurements were performed on the Wide Angular-Range Chopper Spectrometer (ARCS) located at the Spallation Neutron Source at Oak Ridge National Laboratory [33]. An array of five crystals with a total mass of 495.6 mg was co-aligned with the $(H, 0, L)$ scattering plane set horizontally, and attached to the cold head of a closed-cycle-refrigerator. Data were collected at the base temperature of 7 K using incident energies of $E_i = 30, 75, 160$ and 250 meV (elastic resolutions are listed in Table I in the SM). For $E_i = 30, 160$ and 250 meV, the sample was rotated around 180 degrees in one degree increments to full cover the q, E space coverage, where E is the energy transfer. For $E_i = 75$ meV, the rotation increment was reduced to half a degree.

The INS data were reduced to q, and E, symmetrized to improve statistics, and cuts made for further analysis using Mantid [34]. The neutron scattering data are described using the momentum transfer in hexagonal reciprocal lattice units, $q(H,K,L) = \frac{2\pi}{a}\left(\frac{H}{2},\frac{K}{2},L\right) + \frac{2\pi}{a}L$. The INS data are presented in terms of the orthogonal vectors $(1,0,0)$, $(-1,2,0)$, and $(0,0,1)$, as shown in Fig. 1(e). Special K- and M-points in the Brillouin zone are found at $(H,K,L) = (\frac{1}{2},\frac{1}{2},0)$ and $(\frac{1}{2},\frac{1}{2},0)$ and symmetry-related points, respectively. The INS data are displayed as intensities that are proportional to the spin-spin correlation function $S(q,E)$. We first examined the elastic scattering from our co-aligned crystals [shown in Fig. 1(g)] and compared the data to simulations of the nuclear and magnetic scattering [shown in Fig. 1(f)]. Using the Bilbao crystallographic server, we find that below 250 K the magnetic structure adopts the high-symmetry magnetic space group $P6/mmm$ (No. 191) where both magnetic sublattices are restricted to have their ordered moments lying along the c-axis [35]. The ordered magnetic moment at 4.5 K are reported as 2.17 and 9.0 $\mu_B$ for Mn and Tb, respectively [19]. Using these values and the $P6/mmm$ symmetry, we simulated the corresponding nuclear and magnetic neutron diffraction patterns for the $(0,K,L)$ plane using MAG2POL [36]. The good agreement obtained between simulated and experimentally measured patterns confirms the high quality of our samples as well as the previously reported low temperature ferrimagnetic ground state in Tb166.

III. MINIMAL HEISENBERG MODEL FOR THE SPIN EXCITATIONS

Before describing the INS data, we first discuss a minimal description of the magnetic interactions in Tb166 and the key features of the resultant spin excitations. Kagome layers are known for unusual magnetic behavior due to geometric frustration and the role of spin-orbit coupling via the DM interaction. All known hexagonal R166 compounds possess FM kagome layers with an easy-plane Mn magnetic anisotropy which minimizes the
and $2.17$ (Mn) components for Tb$_{166}$ below 250 K. The reciprocal space is here set in the conventional way. Antiparallel magnetic moments of $9.0$ (Tb)

Magnetic interactions within a single Mn-Sn kagome layer. (e) 2D hexagonal Brillouin zone showing conventional reciprocal lattice vectors $a^*$ and $b^*$ and special points, $\Gamma$ (black), $K$ (blue) and $M$ (red). Inelastic neutron scattering data are discussed in terms of the orthogonal vectors $(1,0)$ and $(-1,2)$. (f) Simulated $(0,K)$ plane at 7 K.

FIG. 1: (a) Single-crystal magnetization data for Tb$_{166}$ recorded at 2 K with $H$ applied along $c$. The inset shows a typical single-crystal sample of Tb$_{166}$. (b) Powder x-ray diffraction measurements of Tb$_{166}$ collected at room temperature and fitted using Rietveld refinement analysis. (c) Ferrimagnetic ground state structure of TbMn$_3$Sn$_6$. Key interlayer interactions are shown with heavy black arrows. (d) Magnetic interactions in a single Mn-Sn kagome layer. (e) 2D hexagonal Brillouin zone showing conventional reciprocal lattice vectors $a^*$ and $b^*$ and special points, $\Gamma$ (black), $K$ (blue) and $M$ (red). Inelastic neutron scattering data are discussed in terms of the orthogonal vectors $(1,0)$ and $(-1,2)$. (f) Simulated $(0,K)$ plane at 7 K.

For $R166$ compounds with magnetic rare-earth ions, two additional factors control the magnetic behavior. The first is strong AF coupling between the $R$ and Mn sublattices that can result in tightly bound Mn-$R$-Mn collinear ferrimagnetic trilayers. The second factor is the single-ion anisotropy of the rare-earth ion. For ferrimagnetic Gd$_{166}$, the weak anisotropy of the spin-only Gd$^{3+}$ ion combined with easy-plane Mn anisotropy and Gd–Mn coupling results in antiparallel ordered Gd and Mn moments lying in the basal layer [13]. On the other hand, $R = \text{Tb–Ho}$ ions possess uniaxial anisotropy that competes with the Mn easy-plane anisotropy. This competition, along with higher-order contributions to the $R$ anisotropy [23, 25], drives spin reorientation transitions where the ordered Mn and $R$ moments rotate in unison [13]. As mentioned above, Tb$_{166}$ adopts an out-of-plane uniaxial ferrimagnetic ground state [see Fig. 1(c)], with Mn and Tb moments collectively rotating to fully lie in the basal plane above $T_c = 330$ K. $R = \text{Dy and Ho}$ are similar ferrimagnets with spin reorientation transitions, but the weaker $R$-ion anisotropy results in a ground state easy-axis that is tilted away from the $c$-axis [13]. Close to $T_c$, the competing $R$ and Mn single-ion anisotropies drive first-order magnetization processes in applied magnetic fields [19, 23].

Given the already interesting role of competing interlayer interactions in Y$_{166}$ and competing anisotropies in $R = \text{Tb–Ho}$, it remains to consider their combined role in $R166$ with magnetic rare-earths. We define a general Heisenberg model with the Hamiltonian $\mathcal{H} = \mathcal{H}_{\text{intra}} + \mathcal{H}_{\text{inter}} + \mathcal{H}_{\text{aniso}} + \mathcal{H}_{\text{DM}}$ that consists of intralayer and interlayer pairwise exchange, single-ion anisotropy, and DM interactions.

In our minimal description, each Mn kagome layer

role of intralayer geometric frustration [37]. However, the competition between Mn-Mn FM and AF interlayer magnetic interactions is known to cause magnetic instabilities in Y$_{166}$ that lead to complex helical phases [12, 14].

FIG. 2: (a) Monolayer kagome spin wave dispersion with energy in units of $sJ$ (orange dots) and Mn-Mn bilayer dispersion with $J_{2}^{MM} = 0.5J$ (blue lines). The latter shows the bilayer splitting of odd and even modes by $\omega_B = 2sJ_{2}^{MM}$. (b) Low-energy dispersion when Mn bilayers are coupled through Tb with $S = 3s$ and $J_{2}^{MT} = -0.04J$ (blue lines). The odd bilayer mode and Tb mode (dashed line) are shifted by the ferrimagnetic exchange field, $\omega_P = 2(6s - S)J_{2}^{MT}$, as shown. Red lines include uniaxial Tb single-ion anisotropy with $K_T = 0.07J$ and $K_M = 0$ that introduces a spin gap in the even mode ($\Delta$) and increases the Tb mode spin gap ($\Delta_{\text{Tb}}$). (c) Interlayer dispersion of low-energy branches with identical bilayer splitting, $J_{2}^{MM} + J_{2}^{MT} = 0.5J$ for cases where $J_{2}^{MM} = J_{2}^{MM}$ (blue lines), $J_{1}^{MM} = 0$ (red lines), and $J_{2}^{MM} = 0$ (gray lines). (d) Interlayer dispersion of low-energy branches when $J_{2}^{MM} = J_{2}^{MM}$ and the coupling between Mn layers in adjacent unit cells, $J_{2}^{MM}$, is either ferromagnetic (red lines), antiferromagnetic (blue lines) or zero (gray dashed lines).
possesses strong nearest-neighbor (NN) FM exchange ($J < 0$) which determines the large overall magnon bandwidth.

$$H_{\text{intra}} = J \sum_{\langle i,j \rangle} s_i \cdot s_j$$  \hspace{1cm} (1)$$

Here, $s$ is the Mn spin operator with magnitude $s = 2$. Fig. 2(a) shows the dispersion for a single Mn kagome layer given by $H_{\text{intra}}$ within linear spin wave theory. The overall bandwidth is $6s|J|$ with a Dirac band crossing at the K-point with energy $3s|J|$. As described below, our data analysis does not benefit from the introduction of longer-ranged intralayer interactions, although we cannot exclude them.

To describe the interlayer interactions, Fig. 1(c) shows that nearly equidistant FM Mn kagome layers are stacked along the $c$-axis. Tb layers are inserted after every two Mn layers and with opposite magnetization, forming a Mn(↑)-Mn(↑)-Tb(↓)-Mn(↑)-Mn(↑)-Tb(↓) pattern. Several unique interlayer magnetic couplings between Mn layers and between Mn and Tb layers are possible, giving

$$H_{\text{inter}} = \sum_k \sum_{i<j} J_{k}^{MM} s_i \cdot s_{j+k} + J^{MT} \sum_{\langle i,j \rangle} s_i \cdot S_j.$$  \hspace{1cm} (2)$$

Here, $J^{MT} > 0$ is the AF coupling between neighboring Mn and Tb layers, with Tb having a total spin-plus-orbital angular momentum of $S = 6$. We label interactions between Mn layers by a layer index $k$ ($J_k^{MM}$). Due to the Tb layer, adjacent Mn layers above and below a given Mn layer are inequivalent. Our data indicate that the FM coupling between next-nearest neighbor (NNN) Mn-Mn layers separated by a Sn$_4$ block ($J_2^{MM}$) is stronger than the coupling between NN Mn-Mn layers separated by a TbSn$_2$ block ($J_1^{MM}$), in agreement with analysis of neutron diffraction data [12, 26].

By itself, $J_2^{MM}$ forms strongly coupled FM Mn-Mn bilayers and generates a bilayer splitting $\omega_B = 2s|J_2^{MM}|$ of the single-layer dispersion into odd and even modes, as shown in Fig. 2(a). The K-point splits into two (odd and even) topological magnon crossings that remain ungapped in the absence of DM interactions.

The strong AF interaction $J^{MT}$ generates a ferrimagnetic exchange field with energy scale $\omega_F = 2(6s - S)J^{MT}$. $\omega_F$ increases the odd-even splitting and gives rise to a new branch of Tb character with a spin gap of $\Delta_{\text{Tb}} = \omega_F$ at the $\Gamma$-point, as shown in Fig. 2(b).

The introduction of uniaxial single-ion anisotropy for both Tb and Mn ($K^T$ and $K^M$) is given by

$$H_{\text{aniso}} = K^M \sum_i (s_i^z)^2 + K^T \sum_i (S_i^z)^2$$  \hspace{1cm} (3)$$

where the sums are over each sublattice. Whereas Mn is expected to have a weak easy-plane anisotropy ($K^M \gtrsim 0$), Tb has a large uniaxial anisotropy at low temperatures ($K^T < 0$). With $K^M = 0$, $K^T$ generates a spin gap $\Delta = \sqrt{2sSK^T J^{MT}}$ for the even branch and increases $\Delta_{\text{Tb}}$ such that $\Delta_{\text{Tb}} - \Delta = 2SK^T + \omega_F$, as shown in Fig. 2(b).

We now consider the effect of $J_3^{MM}$. When $J_3^{MM} = 0$, the interlayer dispersion of the low-energy branches is mainly controlled by $J^{MT}$. As $J^{MM}$ is increased, models indicate that the bilayer splitting becomes $\omega_B = 2s|J_2^{MM} + J_3^{MM}|$ and the interlayer bandwidth of odd and even modes sharply increases and reaches a maximum when $J_2^{MM} = J_3^{MM}$, as shown in Fig. 2(c). The limit where $J_2^{MM} = 0$ corresponds to isolated trilayer Mn-Tb-Mn blocks where the interlayer bandwidth is zero.

To better describe the experimental data, an interaction between like Mn layers in adjacent unit cells, $J_3^{MM}$, is introduced as well. As shown in Fig. 2(d), $J_3^{MM}$ opposes the interlayer odd and even bandwidths while preserving the A-point gap at $q = (0,0,1/2)$. For example, when $J_3^{MM}$ is AF, the bandwidth of the odd mode increases and the even mode decreases.

Finally, the presence of DM interactions is principally associated with gapping at the Dirac points at $K$ and has recently been reported in Tb166 [29]. However, as described below, we find no clear evidence for a K-point gap in Tb166, due to the presence of strong damping. Therefore, it is not necessary to introduce DM interactions to model our data ($H_{\text{DM}} = 0$).

IV. INTERLAYER DISPERSIONS

Having outlined the various expectations for the spin wave dispersion in Tb166, we now describe the features of the INS data. Figure 3(a) shows a slice through the $E_i = 30$ meV data along the $(H,0,0)$ and $(0,0,L)$ directions through the $(0,0,2)$ $\Gamma$-point. The lowest energy mode is the even branch, which displays a clean spin gap of $\Delta = 6.5$ meV as shown by the resolution-limited peak in the interlayer dispersion in Tb166, we now describe the features of the INS data. Figure 3(a) shows a slice through the $E_i = 30$ meV data along the $(H,0,0)$ and $(0,0,L)$ directions through the $(0,0,2)$ $\Gamma$-point. The lowest energy mode is the even branch, which displays a clean spin gap of $\Delta = 6.5$ meV as shown by the resolution-limited peak in the energy cut through the $\Gamma$-point at $(0,0,2)$ [Fig. 3(b)]. Along $(0,0,L)$, the even branch has limited interlayer dispersion, reaching only 14 meV at the $A$-point, whereas the intralayer dispersion of the even branch along $(H,0,0)$ extends to much higher energies.

We also glimpse a narrow band of excitations near $\sim 25$ meV in Fig. 3(a) that corresponds to the Tb mode. Figure 4 shows the Tb mode dispersion along $(H,0,0)$ and $(0,0,L)$ more clearly using $E_i = 75$ meV and focusing on Brillouin zones where the structure factor of the even branch is close to zero ($L = \text{odd}$ or $H = \text{odd}$).

The odd branch is observed in slices of the data taken with higher incident energies of 75 and 160 meV, as shown in Fig. 5. The even and odd branches have structure factors that are maximized in Brillouin zones with $L = \text{even}$ and $L = \text{odd}$, respectively. Fig. 5(a) and the constant energy cuts in Fig. 5(b) show that the interlayer odd branch disperses from roughly 60 meV at the $\Gamma$-point down to 40 meV at the $A$-point. Constant-$q$ energy cuts at $(0,0,3)$ and $(0,0,4)$ in Fig. 5(c) also demonstrate a $\Gamma$-point energy of $\sim 60$ meV for the odd branch. Considering
the spin gap, this allows for an estimate of an odd-even splitting of $\omega_B + \omega_F \approx 55$ meV. Figs. 5(a)–(c) show that the odd branch is significantly weaker and broader than the resolution-limited low-energy even and Tb branches, but has a much larger bandwidth.

Various data cuts similar to those shown in Figs. 3–5 were used to produce a list of dispersion points, $\omega_i(\mathbf{q})$, for even, odd, and Tb interlayer branches in various Brillouin zones. In this list, we also include the energies of the intralayer Tb modes along ($0,0,0$) [Fig. 4(a)] and ($-K,2K$) whose dispersions are sensitive to $J_{11}^{MM}$ and $K^T$. We used this list of 100 observables to fit the experimental dispersion to the reduced Heisenberg model $\mathcal{H} = \mathcal{H}_{\text{inter}} + \mathcal{H}_{\text{aniso}}$ using SpinW [1]. The Mn and Tb spin values are fixed to $S = 2$ and $S = 6$, respectively.

For $\mathcal{H}_{\text{aniso}}$, the spin reorientation transition of Tb166 and the general magnetic structures of other R166 compounds suggest that Mn has weak easy-plane anisotropy ($K^M \gtrsim 0$). However, fixing $K^M = 0$ results in a fitted spin gap that is much lower than experimental values. We assume that this discrepancy is caused by additional contributions to the magnetic anisotropy, such as exchange anisotropy, that are not included in our model. The introduction of $K^M < 0$ to our fitting (as an effective uniaxial Mn anisotropy) dramatically improves the fitted spin gap.

For $\mathcal{H}_{\text{inter}}$, the observed odd-even splitting of $\sim 55$ meV is determined primarily by $|J_1^{MM} + J_2^{MM}|$ and the A-point gap of $\sim 25$ meV by $|J_1^{MM} - J_2^{MM}|$. However, the determination of the signs and relative strength of $J_1^{MM}$ and $J_2^{MM}$ requires careful fitting of the interlayer dispersions. We ran 41 different fitting iterations starting with equal values of $J_1^{MM}$ and $J_2^{MM}$. All fitting sessions find $J_1^{MM} + J_2^{MM} \approx 12$ meV with two local minima where $J_1^{MM}/J_2^{MM} \approx 4$ or $1/3$. Both interactions are FM. The case where $J_2^{MM}/J_1^{MM} \approx 4$ turns out to be the global minimum with a reduced $\chi^2 = 0.8$ which is lower than $\chi^2 = 1.0$ for the other case. The fits find that $J_2^{MM}$ is the dominant interlayer interaction, confirming the expectation based on neutron diffraction studies of the double-flat spiral AF structure of Y166 [12, 14].

In the overall fits to $\mathcal{H}_{\text{inter}}$, we find that an AF $J_3^{MM}$ must be introduced to account for the different bandwidths of even ($\sim 10$ meV) and odd ($\sim 20$ meV) interlayer dispersions, as shown in Figs. 2(d) and 5(a). An AF $J_3^{MM}$ will compete with FM $J_1^{MM}$ and could lead to a destabilization of the ferrimagnetic stacking sequence. However, calculations of the classical stability of the ferrimagnetic state described below suggest that $J_3^{MM}$ is not strong enough to create such an instability in Tb166. Similar competing interactions have been proposed for Y166, but with AF $J_3^{MM}$ and FM $J_1^{MM}$ [14, 21]. This cannot be the case for Tb166, since the odd branch would have a minimum in the dispersion at $\Gamma$, which is not observed experimentally.

Fitting the spin wave dispersions produced the set of interlayer exchange parameters in Table 1 where error bars correspond to the variances obtained over all fitting iterations. Further details of the fitting procedure are described in the SM. Within our model, the fit parameters predict an additional four modes (two odd and two even)
TABLE I: Heisenberg parameters for TbMn$_6$Sn$_6$ as obtained from fits to the neutron data.

| Coupling | Energy (meV) | description |
|----------|--------------|-------------|
| $J$      | -14.4 (1)    | intralayer FM |
| $J_{MT}$ | 0.71 (3)     | interlayer AF |
| $J_{1}^{MM}$ | -2.21 (19) | interlayer FM |
| $J_{2}^{MM}$ | -9.58 (10) | interlayer FM |
| $J_{3}^{MM}$ | 0.90 (10)  | interlayer AF |
| $K^{M}$  | -0.65 (3)    | uniaxial anisotropy |
| $K^{T}$  | -0.85 (6)    | uniaxial anisotropy |
| $\omega_B$ | ~47         | bilayer splitting |
| $\omega_F$ | ~8          | ferrimagnetic exchange |

at higher energies. These modes are not clearly observed in the current experiment, as discussed below.

V. INTRALAYER DISPERSIONS

The intralayer dispersions are steeper than the interlayer modes and can extend well beyond 100 meV. The odd and even modes can be isolated in the INS data based on their structure factors which are maximized in Brillouin zones with $L = odd$ and $L = even$, respectively. Slices from the $E_1 = 75$ and 160 meV data corresponding to even modes with $L = 4$ and odd modes with $L = 3$ are shown in Figs. 6(a) and 6(b), respectively. To gain better statistics, the data are averaged over $\Delta L = \pm 0.5$ rlu which broadens features by effectively averaging over the interlayer bandwidth. For $L = 4$, the even mode has a M-point energy of $\approx 70$ meV. For $L = 3$, the odd mode is more strongly broadened by interlayer interactions than the even mode, but we clearly observe the even-odd mode splitting of $\approx 55$ meV.

We obtained the intralayer exchange parameters defined in $\mathcal{H}_{\text{intra}}$ by fitting various cuts of the lowest odd and even branches similar to those shown in Figs. 3 and 6. During the fit, all parameters of $\mathcal{H}_{\text{inter}}$ and $\mathcal{H}_{\text{aniso}}$ were fixed to the values in Table I. Ultimately, we achieved satisfactory agreement with the data with only one parameter corresponding to the nearest-neighbor Mn-Mn intralayer FM interaction with $J = -14.4$ (1) meV. The main reason for this simple result is that the dispersive features quickly deteriorate at higher energies by becoming very broad and weak.

Figure 7(a)-(c) shows intralayer dispersion data after summing over a large range of $\Delta L = \pm 7$ rlu. This improves statistics and allows higher-energy features to be observed, but it mixes odd and even modes and averages over the intralayer dispersions. Excitations are observed up to $\sim 125$ meV which includes evidence for the top of the odd branch near the M-point at $\sim 115$ meV [Fig. 7(b)] and the bottom of the fourth branch (even) at the M-point near 70 meV [Fig. 7(c)]. These data are compared to model calculations in Figs. 7(d)-(f) that average over the same reciprocal space ranges. From the model, the K-point Dirac crossing of the even mode is predicted to occur near 90 meV. However, we are not able to resolve any K-point gapping in the INS data.
The DFT results, we define \( \tilde{J}_{ij} \) which is similar to Eqns. (1) and (2). However, for the J\(_{1}\) and experimental values for the exchange are related by
\[
J_{1} = \tilde{J}_{ij} \cdot S_i \cdot S_j
\]
which is similar to Eqns. (1) and (2). However, for the DFT results, we define \( \tilde{J}_{ij} = m_i^s / 2 \) where \( m_i^s \) is the spin magnetic moment on site \( i \). Thus the DFT and experimental values for the exchange are related by
\[
J_{1} = \tilde{J}_{ij} \cdot S_i \cdot S_j
\]

VI. FIRST-PRINCIPLES CALCULATIONS OF THE MAGNETIC EXCHANGE INTERACTIONS

DFT calculations were carried out to investigate the intrinsic magnetic properties in Tb166, which includes exchange couplings and magnetocrystalline anisotropy. The strongly correlated Tb-4f states were treated in both the DFT+U method and the so-called open-core approach. We also explored the effects on the exchange couplings of additional electron repulsion for Mn-\( d \) orbitals. The around-the-mean-field double-counting scheme [39] was used in DFT+U. For Mn, various U values of 0–2 eV were employed to calculate exchange couplings and compare them with experimental results. Details of these calculations can be found in the SM.

The calculated total magnetic moments of Tb and Mn, \( m_{Mn} = 9.24 \) \( \mu_B \) and \( m_{Mn} = 2.42 \) \( \mu_B \), respectively, agree well with experimental results [19]. Four interlayer isotropic exchange couplings, including three Mn-Mn couplings \( J_{i}^{MM} (i = 1, 2, 3) \) and one Tb-Mn coupling \( J_{MT} \), were calculated by mapping the total energies of five collinear spin configurations (see SM) into the Heisenberg model defined as
\[
\mathcal{H} = \sum_{i<j} J_{ij} S_i \cdot S_j
\]

The calculated spin moments and interlayer exchange parameters with various \( U \) are summarized in Table II

\[
U \ (eV) \quad m_{Mn}^Tb \quad m_{Mn}^{Mn} \quad J_{1}^{MM} \quad J_{2}^{MM} \quad J_{3}^{MM} \quad J_{MT}^{MM}
\]
\[
0 \quad 6.34 \quad -2.43 \quad 4.52 \quad -27.69 \quad -1.27 \quad 2.39
\]
\[
1.0 \quad 6.34 \quad -2.40 \quad 3.57 \quad -27.99 \quad 0.12 \quad 2.82
\]
\[
1.5 \quad 6.34 \quad -2.38 \quad -0.04 \quad -24.09 \quad 2.86 \quad 3.17
\]
\[
2.0 \quad 6.34 \quad -2.36 \quad -5.97 \quad -15.73 \quad 7.54 \quad 3.76
\]

In contrast to experiments, calculations with \( U_{Mn} = 0 \) give an AF \( J_{2}^{MM} \) and FM \( J_{3}^{MM} \), although the relative sign of these interactions does not modify the overall ferrimagnetic structure which is stabilized by \( J_{2}^{MM} \) and \( J_{MT}^{MM} \). The application of additional electron repulsion on Mn-3d orbitals further separates the occupied and unoccupied Mn-5d bands. However, the Mn electron occupancies in the two spin channels nearly remain the same (1.39 and 3.80 at \( U = 0 \)). Thus, as shown in Table II, the Mn spin magnetic moment only slightly decreases by 3% and the Tb spin magnetic moment remains the same when \( U \) changes from 0 to 2 eV. In contrast, the variation of the exchange parameters is much more pronounced. Remarkably, both \( J_{1}^{MM} \) and \( J_{3}^{MM} \) can change their signs with increasing \( U \) and become more consistent with experimental observations. It is worth noting that additional electron repulsion on Mn-\( d \) orbitals is also needed to correctly describe the magnetic interactions in materials such as MnBi\(_2\)Te\(_4\) [40] and MnSb\(_2\)Te\(_4\) [41, 42].

The magnetic anisotropy energy (MAE) is also investigated by calculating the total energies of the ferrimagnetic state as a function of spin quantization direction. Spin-orbit coupling is included through the second variation approach. Fig. 8(b) shows the calculated total MAE energy as a function of spin quantization direction. Spin-orbit coupling is included through the second variation approach. Fig. 8(b) shows the non-monotonic dependence of \( E \) on \( \theta \) that is consistent with substantial higher-order MAE constants that drive the spin reorientation transition [23].

The MAE calculations were fit to the expression
\[
E(\theta) = K_1 \sin^2 \theta + K_2 \sin^4 \theta
\]
for different ranges of \( \theta \) with the fitted parameters shown

FIG. 7: Slices of the \( E_1 = 250 \) meV data after averaging over \( \Delta L = \pm 7 \) showing the dispersion along the (a) (H,0), (b) (−K,2K,0) and (c) (2K,1/2 − K) directions. For all panels, the data are additionally averaged over either \( \Delta H = \pm 0.1 \) or \( \Delta K = \pm 0.058 \). (d)-(f) Model calculations of the neutron intensities with the same reciprocal space averaging of the data as in (a)-(c) and convolved with a Gaussian energy FWHM of 12 meV. In all panels, pink lines correspond to model dispersions with \( L = 0 \) (solid lines) and \( L = 0.5 \) (dashed lines).
FIG. 8: (a) Interlayer exchange parameters as functions of $U$ applied on Mn $d$-states and compared to the experimental values. All exchange values are shown with experimental exchange scaling. (b) Variation of magnetic anisotropy energy (MAE) as a function of spin quantization axis rotation in TbMn$_x$Sn$_{6-x}$. $\theta = 0^\circ$ corresponds to the out-of-plane spin orientation parallel to the c-axis. Calculations are performed with experimental ferrimagnetic collinear spin ordering.

TABLE III: Different fits to the angular dependence of the MAE calculated from DFT. See Fig. 8(b) for details.

| Fit | $K_1$ (meV) | $K_2$ (meV) |
|-----|-------------|-------------|
| 1   | 61.6        | -49.0       |
| 2   | 48.7        | -22.2       |
| 3   | 45.7        | -9.0        |

in Table III. The large ratio of $K_2/K_1$ from fit 1 is sufficient to drive the spin reorientation. However, fit 3 best corresponds to the DFT estimate of the ground state single-ion anisotropy which can be compared to the experimental value [see Eqn.(3)] according to the scaling $K^T + K^M \approx -K_1/s^2 = -1.3$ meV. This is in reasonable agreement with the experimental result from Table I.

VII. DISCUSSION

The INS data for Tb166 provide a minimal set of exchange and anisotropy parameters that are largely consistent with our DFT results and indirect estimations of these energy scales from magnetization and neutron diffraction data (see e.g. Refs. [23] and [26]). The key conclusions are: (1) large intralayer FM interactions between Mn ions, (2) interlayer interactions that are dominated by FM coupling between Mn layers spaced by Sn layers ($J^{MM}_3$) and AF coupling between Mn and Tb layers, (3) the presence of competing, weaker AF and FM Mn-Mn interlayer couplings, and (4) a net uniaxial magnetic anisotropy.

With respect to (2) and (3), we consider the overall stability of the ferrimagnetic structure of Tb166 by examining the classical magnetic energies of collinear layer stackings given by

$$E = 6J^{MT}_M[(s_1+s_2)\cdot s_3 + (s_3+s_4)\cdot s_1] + 3J^{MM}_1(s_1\cdot s_2+s_3\cdot s_4) + 3J^{MM}_2(s_1\cdot s_4 + s_2\cdot s_3) + 6J^{MM}_3(s_1\cdot s_3 + s_2\cdot s_4). \quad (6)$$

Here, the numbers label successive Mn layers and letters label Tb layers for a six-layer stack. The ground state ferrimagnetic structure has an energy of

$$E_{\text{ferr}} = -24s^2(J^{MM}_1 - 6s^2(J^{MM}_2 + 2J^{MM}_3)). \quad (7)$$

The next higher-energy state corresponds to AF up-down-down-up (UDDU) Mn layer stacking. For uniaxial anisotropy, the classical UDDU state will decouple the Mn and the Tb layers and

$$E_{\text{UDDU}} = -6s^2(-J^{MM}_1 + J^{MM}_2 + J^{MM}_3). \quad (8)$$

The parameters in Table I provide $E_{\text{ferr}} = -440$ meV and $E_{\text{UDDU}} = -220$ meV, indicating that the high stability of the ferrimagnetic ground state arises from $J^{MM}_1$. In the absence of $J^{MM}_1$ (as for Y166), the collinear ferromagnetic, ferrimagnetic and UDDU states are nearly degenerate since $J^{MM}_1 \approx 2J^{MM}_3$. This suggests that similar competition between these interlayer interactions drives complex helical ordering observed in Y166.

Based on these comparisons, it is interesting to consider the transferability of exchange interactions in Tb166 with other R166 compounds. INS investigations of Y166 in Ref. [29] report a NN intralayer exchange that is nearly identical to Tb166. While the interlayer interactions in Y166 are not studied in detail in Ref. [29], the bilayer splitting energy is reported as $|J^{MM}_1 + J^{MM}_2| \approx 12$ meV, which is the same as Tb166. This suggests that $J^{MM}_1$ and $J^{MM}_2$ interactions are both FM and have similar strengths in Y166 and Tb166. One caveat is that additional intralayer and interlayer interactions are also fit in Ref. [29]. Interestingly, our DFT calculations support an AF $J^{MM}_1$ and FM $J^{MM}_3$ and vice versa, with the result depending on the correlation parameter $U$. Overall, these comparisons give some confidence that the Mn-Mn magnetic interactions in these and other R166 are transferable, although slight changes in structure may affect the overall balance of $J^{MM}_1$ and $J^{MM}_3$.

There is little data reporting the magnitude of the Mn-R coupling in other R166 compounds. For Gd166, the energy scale for the Gd mode is reported to be $\sim 24$ meV from powder INS data [43], very similar to the Tb mode energy observed here. However, given the absence of Gd single-ion anisotropy, this should correspond to only to $\omega_F$ and we estimate that $J^{Gd}_{\text{MnGd}} \approx \omega_F/2(6s - S_{\text{Gd}}) = 1.4$ meV where $S_{\text{Gd}} = 7/2 - 2$. The energy of Tb mode is lifted appreciably by anisotropy, $2SK^T = 11$ meV. Thus, our reported $J^{MT}$ is only half as large as $J^{\text{MnGd}}$, a result that is roughly consistent with deGennes scaling. Extrapolating the deGennes scaling to Ho166 and Er166 should result in weaker ferrimagnetism. For Er166, this weakening results in a decoupling of the Mn and Er sublattice magnetic ordering at high temperatures [11].
The magnetic anisotropies of Tb166 determined from INS may present some inconsistencies with our understanding of R166 compounds. At low temperatures, Tb166 is dominated by the large uniaxial anisotropy of the Tb ion, a result that is consistent with our INS data and DFT results. However, the INS data cannot be modeled with an easy-plane Mn anisotropy parameter since the spin gap becomes too small. Instead, we obtain the best fitting results by assuming that Mn also has uniaxial single-ion anisotropy. This is inconsistent with INS data from Y166 that finds a rather large value for the Mn easy-plane single-ion anisotropy parameter (2.5 meV in our units), although the spin gap itself is not reported [29]. It is very possible that Mn-Mn and Mn-Tb exchange anisotropy contributes to the spin gap as well. First-principles calculations find significant exchange anisotropy of the intralayer coupling in Y166 [14]. In Tb166, the Tb magnetic anisotropy is temperature dependent, and our MAE calculations in the ground state are consistent with the expected conditions for the spin reorientation transition that occurs at 330K. It will be interesting to study the spin excitations in this temperature regime to learn more about the unusual magnetic anisotropy of R166 compounds.

VIII. SUMMARY

INS data for Tb166 provide a minimal set of exchange and anisotropy parameters that are largely consistent with indirect estimations of these energy scales provided by magnetization data and neutron diffraction. The key conclusions are: (1) large intralayer FM interactions between Mn ions, (2) interlayer interactions that are dominated by FM coupling between Mn layers spaced by Sn layers ($J_{1}^{MM}$) and AF coupling between Mn and Tb layers, (3) the presence of weaker FM and AF Mn-Mn interlayer couplings, and (4) an overall uniaxial magnetic anisotropy. These results suggest that the magnetism of R166 compounds, with a variety of magnetic ground states and high-temperature or high-field instabilities, may be understood with transferable set of magnetic interactions.

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Competing magnetic energy scales in the topological flat-band ferrimagnet TbMn₆Sn₆
(Supplementary Information)

Supplementary Table I: Configurations of the ARCS spectrometer.

| $E_i$ (meV) | Chopper (Hz) | Elastic FWHM (meV) |
|------------|-------------|-------------------|
| 30         | 120         | 1.4               |
| 75         | 180         | 3.9               |
| 160        | 240         | 7.6               |
| 250        | 300         | 11.9              |

I. INELASTIC NEUTRON SCATTERING MEASUREMENTS AND DATA ANALYSIS

Inelastic neutron scattering measurements were performed on the ARCS spectrometer at four incident energies; $E_i = 30, 75, 160,$ and $250$ meV. Table I shows the chopper configurations and their elastic energy resolution full-width-at-half-maximum (FWHM).

The procedure for fitting the Heisenberg parameters is described as follows. A series of constant-energy and constant-$q$ cuts is made through various spin wave branches. The cuts are fit to Gaussian lineshapes, providing points on the dispersion curve of the spin wave branches. The cuts are fit to Gaussian lineshapes, providing points on the dispersion curve of the spin wave branches. The cuts are fit to Gaussian lineshapes, providing points on the dispersion curve of the spin wave branches.

We fit the dispersion points to the Heisenberg Hamiltonian described Eqs. (1)-(3) in the main text using SpinW [1]. The fitting was performed on two separate sets of dispersion points. The first set is a collection of 100 dispersion points, mainly the interlayer dispersions, that determined the parameters of the interlayer ($H_{\text{inter}}$) and anisotropy terms ($H_{\text{aniso}}$). These are fit while fixing the intralayer exchange $J$. The second is a collection of 110 intralayer dispersion points where $J$ is fit and parameters from $H_{\text{inter}}$ and $H_{\text{aniso}}$ are fixed. Fits of the two sets were performed iteratively to optimize all parameters of the Hamiltonian.

The final fitting iteration for the interlayer parameters was performed while keeping the intralayer interaction $J$ fixed at a value of -14.4 meV. Figure 1(c) shows one such fitting. The dispersion points (in red) are shown at $q$-points indicated on the $x$-axis and the circles are the fitted dispersion values. This fit was repeated 41 times and the set of parameters with the lowest $\chi^2$ value are reported in Table I of the main text. Errors in the Heisenberg parameters are reported as the variance of the parameters over all 41 fitting iterations. A similar procedure was used to fit the intralayer dispersion and a typical result is shown in Fig. 1(d).

II. DENSITY-FUNCTIONAL THEORY CALCULATIONS

Density-functional theory (DFT) calculations are performed using a full-potential linear augmented plane wave (FP-LAPW) method, as implemented in Wien2k [2]. The generalized gradient approximation of Perdew, Burke, and Ernzerhof [3] is used for the correlation and exchange potentials. To generate the self-consistent potential and charge, we employed $R_{\text{MT}} \cdot K_{\text{max}} = 8.0$ with muffin-tin radii $R_{\text{MT}} = 2.7, 2.4,$ and $2.5$ a.u., for Tb, Mn, and Sn, respectively. The calculations are performed with 74 $k$-points in the irreducible Brillouin zone. They are iterated until charge differences between consecutive iterations are smaller than $10^{-3}$, and the total energy difference is lower than 0.01 mRy. The experimental lattice parameters [4] are adopted in all calculations.

The Tb atom in Tb166 has a configuration of [Xe]6s²5d³4f⁸. Due to the large spin-orbit coupling of the Tb-4f shell, in general, it is expected that 4f electron configuration follows the Hunds’ rules, giving spin magnetic moment $m_{s}^{\text{Tb-4f}} = 6 \mu_{B}$ and orbital magnetic moment $m_{l}^{\text{Tb-4f}} = 3 \mu_{B}$ and a total Tb-4f moment $m_{\text{tot}}^{\text{Tb-4f}} = 9 \mu_{B}$. Within DFT+$U$ at $U = 0.5$ mRy, the calculated spin magnetic moment and orbital magnetic moments on the Tb site are $m_{s}^{\text{Tb}} = 6.26 \mu_{B}$ and $m_{l}^{\text{Tb}} = 2.98 \mu_{B}$, satisfying expectations from the Hunds’ rules well. The Tb-5d electrons are spin-polarized by the neighboring 12 magnetic Mn atoms through hybridization. The resulting 5d spin moment aligns parallel with the 4f spin moment, which further spin-polarizes the 5d electrons, and has a value of $m_{5d} \approx 0.31 \mu_{B}$.

Exchange parameters are extracted by mapping the total energies of various supercell spin configurations, as shown in Fig. 2. A $1 \times 1 \times 3$ supercell of three formula units (f.u.) is constructed to accommodate various collinear spin configurations. There are six Mn layers and three Tb layers in the constructed supercell.

Additional $U$ potentials are applied on Mn-3d orbitals. The exchange couplings between the Tb and Mn atoms are dominated by Tb-5d and Mn-3d orbitals. For simplicity, we treat Tb 4f states as core states and constrain their spin magnetic moment to be $6 \mu_{B}$. We also calculate $J$ values by treating Tb-4f electrons within DFT+$U$ and obtain similar results, validating the $f$-core treatment of Tb states for the exchange coupling calculations. In all calculations with $U = 0-2$ eV on Mn-3d, the experimental ferrimagnetic states are found to have the lowest energy among all spin configurations.
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Supplementary Figure 1: (a) Fits to the even branch along (0, 0, L) with $E_i = 30$ meV. (b) Fits to the Tb branch along (1, 0, L) with $E_i = 75$ meV. (c) Typical results of SpinW fits to $H_{\text{inter}}$ and $H_{\text{aniso}}$ for the 100 $q$-points listed on the $x$-axis. Red symbols are data points with error bars. Open circles are fitted model results. (d) Typical results of SpinW fits to $H_{\text{intra}}$ for the 110 $q$-points listed on the $x$-axis.
Supplementary Figure 2: Supercell spin configurations used to calculate the exchange parameters. Tb atoms are indicated with spheres in blue color. Sn atoms are in grey. Magneta and green spheres are Mn atoms with up and down spin orientations, respectively.