Local symmetry breaking with correlation to ferromagnetism and the Weyl state in \( \text{Co}_3\text{Sn}_2\text{S}_2 \)

Qiang Zhang\(^1,\ast\), Yuanpeng Zhang\(^1,\ast\), Masaaki Matsuda\(^1\), Vasile O Garlea\(^1\), Jiaqiang Yan\(^2\) Michael A. McGuire\(^2\), D. Alan Tennant\(^{1,3,4,\dagger}\), & Satoshi Okamoto\(^2\)

\(^1\)Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA

\(^2\)Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA

\(^3\)Quantum Science Center, Oak Ridge, Tennessee 37831, USA

\(^4\)Shull Wollan Center, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA

\(^\dagger\)current address: Dept. of Physics and Astronomy, University of Tennessee Knoxville, TN 37996-1200

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Magnetic semimets have attracted intense interest recently due to the exotic quantum phenomena for potential application and exploring new fundamental physics. Currently, theoretical studies and the interpretation of the experimental results on them are based on utilizing the average crystal structure and/or magnetic order. Here, we report the discovery of local symmetry breaking and its significant effect on the local ferromagnetism and band topology in a kagome-lattice Weyl semimetal \( \text{Co}_3\text{Sn}_2\text{S}_2 \), by a combined use of half polarized neutron diffraction, total scattering, and density function theory. We find a local symmetry breaking from rhombohedral \( R-3m \) to monoclinic \( Cm \) involving a local distortion of the
cobalt kagome lattice with an in-plane projection along the (100) direction emerges with the onset of ferromagnetic order below $T_C$, revealing a strong coupling between lattice instability and the ferromagnetism. Such local symmetry breaking has the tendency to drive a local ferromagnetic moment reorientation by around $19^\circ$ to the monoclinic $a_m$ axis, and plays a detrimental role in the formation of the Weyl points by breaking mirror symmetries. Our results highlight the important role that local symmetry breaking plays in $\text{Co}_3\text{Sn}_2\text{S}_2$, which may open a new avenue for further theoretical and experimental studies on a wide variation of topological Dirac or Weyl semimetals and kagome magnets.\textsuperscript{*}

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Magnetic Dirac or Weyl semimetals\textsuperscript{1–7}, as a newly emerged class of quantum materials, have stimulated tremendous research interest very recently since they give access to new quantum phenomena with the coexistence to the magnetism. Current understandings of their topological properties for instance anomalous Hall effect or electronic band topology, are based on the globally averaged crystal symmetry\textsuperscript{8} and/or long-range magnetic order\textsuperscript{9,10}. Nevertheless, in a doped alloy or even an ordered, non-alloyed crystal, a hidden local symmetry breaking such as local atomic displacement or polyhedral distortion may be different from the average crystal structure.\textsuperscript{11,12} Such structural complexity was found in some doped nonmagnetic\textsuperscript{13} or magnetic\textsuperscript{14,15} alloys, the origin of which is related to the different local configurations due to site disorder arising from the chemical doping or nonstoichiometry. Relatively few ordered and non-alloyed magnetic materials\textsuperscript{12,16–19} exhibit a locally lower symmetry than the average structure due to the lacking of the site disorder. Recent investigations revealed that the local symmetry breaking plays a vital role in providing a valid insight to their physical properties and advancing our understanding on these magnetic materials. For example, a canted ferromagnetic order is preceded by a local cubic symmetry breaking at higher temperature in Ba\textsubscript{2}NaOsO\textsubscript{6}, which provides an experimental confirmation of the microscopic quantum models\textsuperscript{17}. Furthermore, from the theoretical perspective, it may not be a safe practice to proceed the electronic band structure calculations using the average crystal or magnetic structure without considering the distinct local structure.\textsuperscript{12,20} Unfortunately, such hidden local symmetry breaking remains undiscovered in magnetic Dirac or Weyl semimetals. More importantly, the effect of the local symmetry breaking on the local magnetic configurations and topological properties in the magnetic semimetals has not been explored.
The newly discovered Weyl semimetal Co$_3$Sn$_2$S$_2$ with a kagome lattice of cobalt ions has triggered intense interest due to the novel topological states coupled to the peculiar magnetism. Co$_3$Sn$_2$S$_2$ crystallizes in a rhombohedral structure with space group $R-3m$, which persists down to $\approx 6$ K. It exhibits two anomalies at $T_A \approx 135$ K and $T_C \approx 175$ K in the susceptibility. Theoretical calculations, magnetization and spin waves measurements show that the ground state of Co$_3$Sn$_2$S$_2$ is a very likely ferromagnetic order with easy axis along the out-of-plane $c_H$ axis. Interestingly, Guguchia et al. proposed a phase separation of the out-of-plane FM order and a possible $120^\circ$ in-plane antiferromagnetic (AFM) order in $T_A < T < T_C$, followed by a sole FM order in $T < T_A$ based on the local probe technique Muon spin rotation ($\mu$SR), as shown in Fig. 1(a). The FM volume fraction starts to decrease with increasing the temperature above $T_A$, with a compensation of the AFM volume fraction. Nevertheless, the observation of the spin glass phase and exchange bias indicate that it may not be a sole FM order in $T < T_A$. A coexistence of the spin glass phase and FM phase was proposed to interpret the exchange bias. The existence of the spin glass phase raised a question if the ferromagnetism is short-range or long-range ordered in $T < T_A$. While the magnetization shows a saturation magnetization $\approx 0.3 \, \mu_B/Co$ for the ferromagnetic order, the microscopic $\mu$SR reveals a much lower moment $\approx 0.15 \, \mu_B$ for both FM and AFM orders. In addition, ferromagnetic order is critical to break the time reversal symmetry to induce the Weyl state and in-plane ferromagnetic moment direction could modify the electronic band structures and induce the appearance of the nodal rings based on the first-principles calculations. Thus, it is of importance to provide unambiguous proof by the neutron technology on the ferromagnetic moment direction, size and correlation. However, given the ferromagnetic
moment is very low, unpolarized powder neutron diffraction$^{29,31}$ failed to detect any ferromagnetic signal. On the other hand, although there is an intrinsic geometric frustration to the cobalt kagome lattice and a weak spin frustration$^{29}$, the appearance of the spin glass phase in $T < T_A$ is surprising since there lacks the site disorder in the global rhombohedral structure that is generally needed for the formation of spin glass state. Furthermore, the separated FM and AFM phases in $T_A < T < T_C$ belong to two different irreducible representations$^{27}$, which implies an existence of a structural complexity or a symmetry breaking. Given that the global rhombohedral structure with space group $R-3m$ is maintained down to $\approx 6$ K$^{29}$ and there are successive magnetic transitions at $T_C$ and $T_A$, it is of great interest to explore if there is any local symmetry breaking associated with these two magnetic transitions, which may in turn influence the local moment configuration and electronic band topology significantly.

In this paper, we discovered a local symmetry breaking from rhombohedral $R-3m$ to monoclinic $Cm$ appearing with the onset of the long-range FM order with average moment along the $c_H$ axis below $T_C$ in $\text{Co}_3\text{Sn}_2\text{S}_2$. The in-plane projection of the local distortion for the cobalt kagome lattice points to the (100) direction. Such local symmetry breaking, indicative of lattice instability below $T_C$, leads to a local ferromagnetic instability and may have the tendency to drive a local FM moment reorientation by $\approx 19^\circ$ relative to the $c_H$ axis. Strikingly, Density function theory calculations further reveal that the local monoclinic distortion plays a detrimental role in the formation of the Weyl points by breaking mirror symmetries. Our findings established $\text{Co}_3\text{Sn}_2\text{S}_2$ as a rare example in two categories of magnetic semimetals and kagome magnets where a locally lower symmetry than average lattice symmetry exists and correlates to the magnetism and electronic
Half-polarized neutron diffraction was employed to study the ferromagnetic order which has a low magnetic moment in Co$_3$Sn$_2$S$_2$. In contrast to the unpolarized scattering from a weak ferromagnetic component that produces only a minor change in the intensity of nuclear peaks, the half polarized scattering allows to enhance the sensitivity to the magnetic signal by taking advantage of the interference term $P_0^*F_M^*F_N$, where $F_N$ and $F_N$ are the nuclear and magnetic structure factors, and $P_0$ is the incident neutron beam polarization. A vertical external magnetic field was applied to co-align the ferromagnetic domains and avoid the beam depolarization. For the setup with $Q \perp c_H$ in the hexagonal notation, the up-spin and down-spin diffraction intensities $I^+$ and $I^-$ for the $Q=(110)$ peak are described as $(F_N^2+2P_0 \times F_N F_M + F_M^2)$ and $(F_N^2-2P_0 \times F_N F_M + F_M^2)$, respectively. Thus, the flipping ratio $R=I^+/I^- \approx 1+4P_0 \times F_M/F_N$. On the other hand, the difference between $I^+$ and $I^-$ is $4P_0 \times F_N F_M$, which is proportional to $F_M$. Therefore, even if the FM moment is small, it can be detected by the flipping ratio $R$ or flipping difference $(I^+-I^-)$.

Figure 2 (a-c) shows the $\theta - 2\theta$ scan of the up-spin diffraction intensity $I^+$ and down-spin diffraction intensity $I^-$ of (110) peak at 210 K ($T > T_C$), 158 K ($T_A < T < T_C$) and 1.5 K ($T < T_A$) collected at the triple axis spectrometer HB1, with $P_0$ of $\approx 80\%$. At 210 K, $I^+$ and $I^-$ show no difference indicating there is no ferromagnetic signal. At 158 K, there is a difference in $I^+$ and $I^-$, which becomes larger at 1.5 K. The temperature dependence of the integrated intensity for $I^+$ and $I^-$ is displayed in Fig. 2 (d). The flipping ratio $R$ and difference of $I^+$ and $I^-$ are shown in Fig. 2(e). The flipping ratio derived from HYSPEC is over-plotted in Fig. 2(e), showing a good
consistency. Both the flipping ratios and difference of (110) peak exhibit a clear increase below around $T_C$ and become unchanged near $T_A$. This indicates that (110) is the ferromagnetic peak that appears below $T_C$ and becomes saturated below $T_A$. There is no broadening in the linewidths of the peaks for spin-up and spin-down channels going through $T_C$ and $T_A$, indicating that ferromagnetic order is of long range. At base temperature 4 K, the flipping ratio is 1.055. We obtained $F_M/F_N \approx (R-1)/(4 \times P_0) = 0.0172$ yielding $F_M^2/F_N^2 \approx 0.00030$. Therefore, the ordered moment of the long-range ferromagnetic order is determined to be $\approx 0.14(2) \mu_B$ at 1.5 K, with the moment along the $c_H$ axis. The long-range ordered FM moment size is consistent with the magnitude based on the microscopic $\mu SR$ measurements.

To explore the possible correlation between magnetic order and the crystal lattice, high resolution neutron powder diffraction experiments were employed to study the temperature dependence of the lattice constants, unit cell volume, bond distances and angles. The Rietveld analysis on the neutron diffraction patterns were shown in Fig. S1 (a-c) and the main results are displayed in Fig. 3(a-d). While the lattice constant $c$ exhibits monotonous decrease without any anomaly at $T_C$ and $T_A$, there is a clear anomaly observed near $T_C$ in lattice constant $a$, unveiling a strong spin-lattice coupling occurring in the $a_Hb_H$ plane. The cobalt atoms are octahedrally coordinated by two S atoms and two in-equivalent Sn(1) and Sn(2) atoms. Within the octahedral environment of cobalt, the Co-S and Co-Sn(1) bond lengths, and the Co-S-Co and Co-Sn(1)-Co angles do not exhibit any anomaly at $T_C$ or $T_A$. The appearance of the ferromagnetic order drives a clear anomaly in the Co-Sn(2) bond lengths. As reported previously, the ferromagnetic order is dominated by the third-neighbor “across-hexagon” $J_d$ model via Co-Sn(2)-Co exchange pathway. Thus, this
demonstrates an intimate connection between dominant $J_d$ and Co-Sn(2) bond lengths and the manifestation of the ferromagnetic order in $T < T_C$.

Going beyond the Bragg patterns, high quality neutron total scattering data at a series of temperatures were further inspected and modeled, to explore whether local structure distortion takes place. Representative pair distribution functions (PDFs) going across the magnetic transition temperatures $T_C$ and $T_A$ are presented in Fig. 4. Interestingly, one can observe the emergence of new PDF peaks indicative of a local symmetry breaking as $T$ is lowered. Representative PDF patterns in a typical region $\approx 5 \AA$ are depicted in the inset of Fig. 4(a). The detailed temperature dependence of the PDF patterns in the region of $4.2 \AA < r < 5.55 \AA$ shown in Fig. 2(f) indicates that strikingly, the new PDF peaks near 5.1 and 4.9 \AA co-emerge with the long-range FM order at $T_C$ and with the further decrease of $T$, they become stronger. The PDF peak intensity at 4.5 \AA also shows a rapid increase below $T_C$. The RMC approach\textsuperscript{33, 34} we used here is different from the conventional crystallographic study in that it models the total scattering data (in both $Q$ and $r$-space) along with the Bragg data simultaneously and can unveil the rich information on the local structure within the average structural framework. The fitting results on PDF patterns in real-space are presented in Fig. 4 (c-e), with the $Q$-space and Bragg results that can be found in Fig. S2 in the supplementary material. The partial PDFs corresponding to each pair of atoms were calculated and put in Fig. 4 (b). Comparing among the three $T$ points, one can notice more complicated local bonding environments as the temperature is lowered to the magnetic ordered region. For example, the Sn-S partial exhibits a gradual splitting as $T$ decreases. Meanwhile, multiple partial PDFs show shoulder peaks around 2.5 \AA in both 160 K and 6 K datasets, with a stronger signature at
Such features observed in multiple partial PDFs are consistent with the strengthening of the shoulder peak $\approx 2.5 \text{ Å}$ between the first two main peaks in the experimental PDF patterns in Fig. 4 (a). Note that for the dataset above $T_C$, the shoulder peak $\approx 2.5 \text{ Å}$ in PDF patterns can also be observed indicating that it comes from Fourier ripple, since no local distortion was found. Thus, the shoulder peak at 250 K in Fig. 4(a) can be treated as a baseline when considering the shoulder features $\approx 2.5 \text{ Å}$ below $T_C$.

However, since multiple partials (Co-Co, Co-Sn, etc.) exhibit shoulder features, it is still unclear in what form the local structure is distorted. Thus, the structural configurations obtained through RMC modeling were further analyzed. First, local bond angles were calculated and those Co and Sn(2) relevant angles in kagome lattice in the $a_Hb_H$ plane are presented in Fig. 5 (a-b). For the 250 K dataset, both Co-Co-Co and Co-Sn(2)-Co angles show Gaussian-like distributions around the nominal position, i.e., 60° and 120°. For the 160 K dataset, weak shoulder features emerge alongside both peaks for Co-Co-Co and Co-Sn(2)-Co and the signature becomes very clear for 6 K. Such features in both Co-Co-Co and Co-Sn(2)-Co angle distributions indicate that in $T < T_C$, the kagome lattice within the $a_Hb_H$ plane is distorted locally breaking the local 3-fold point symmetry. Second, to further reveal the distortion of the kagome lattice and local atomic displacements, all Co and Sn(2) coordinates in the collapsed unit cell were projected onto the $a_Hb_H$ plane, from which the atomic distribution density was estimated through kernel density estimation (KDE). Results at all the three temperatures are presented in Fig. 5 (c-e). The interesting finding here is an obvious skewed distribution of the cobalt kagome lattice and central Sn2 atoms along the (100) direction as indicated by the black dashed arrow in Fig. 5 (d) and (e). Similar signature
of skewed distribution can also be observed in 160 K dataset but not in 250 K dataset. Such a skewed distribution for 6 K dataset is consistent with the emergence of shoulder peaks in the angle distributions. Third, to determine the local symmetry, we employed the program FINDSYM \textsuperscript{35,36} to reconstruct the space group from the collapsed supercell configurations obtained via RMC modeling. Concerning the symmetry mining, a tolerance is set for the atomic displacement – atoms displaced by less than the tolerance will be regarded as displacement-free. A higher tolerance tends to construct higher symmetry, and vice versa. A series of tolerance values were tried for all analysis from which a heatmap could be built as presented in Fig. 1(c). For 6 K dataset, the $Cm$ symmetry (No. 8) is determined for the tolerance as high as 0.055 Å. The same low symmetry could also be observed at 160 K dataset with a much lower tolerance due to the weak distortion at 160 K.

Thus, upon cooling below $T_C$, although the global crystal symmetry is kept to be rhombohedral $R$-$3m$, there is a local symmetry breaking to monoclinic $Cm$. The obtained Wyckoff positions and atomic positions for the monoclinic structure are summarized in Table 1. The relationship between these two structure is illustrated in Fig. 1(b). As compared to the average rhombohedral structure, one cobalt and sulfur site split into two due to the local atomic displacements in monoclinic $Cm$. The local displacements of Co1 are confined in the $a_mc_m$ plane, whereas the local displacements of Co2 are in the 3-dimensional direction in the monoclinic cell. The in-plane projection of the local kagome lattice distortion along the (100) direction in the hexagonal notation is well consistent with the $Cm$ symmetry. For example, the local displacements of Co1 occur in the $a_mc_m$ plane of the monoclinic unit cell, and its projection in kagome lattice plane corresponds to (100) direction of the hexagonal notation. The distinct local symmetry from the average sym-
metry is usually driven by competing internal forces\textsuperscript{11} and geometric frustration\textsuperscript{16} was proposed to be responsible for such structural complexity in ordered and non-alloyed compounds. We believe that the intrinsic geometric frustration existing in the kagome lattice of cobalt in Co\textsubscript{3}Sn\textsubscript{2}S\textsubscript{2} and the ferromagnetic interactions may contribute to the occurrence of the local symmetry breaking below $T_C$. The discovery of the local symmetry breaking could help understand the previously irreconcilable magnetic properties in Co\textsubscript{3}Sn\textsubscript{2}S\textsubscript{2}. The local symmetry breaking below $T_C$ reconciles with the occurrence of the phase separation in $T_A < T < T_C$. Upon cooling below $T_A$, the stronger local atomic displacements/disorder of the two Co sites and the geometric/magnetic frustration may cooperatively contribute to the appearance of spin glass state.

Next we investigate the effect of monoclinic lattice distortion on the magnetism and electronic band structure using density functional theory (DFT). Because of the difficulty including local distortion in the global undistorted structure, we consider the global structure in which the local distortion is repeated in the entire lattice. We first considered a FM state without spin-orbit coupling (SOC). Then, we turn on SOC and consider symmetry-allowed four FM states with Co ordered moments pointing along the $a_m$ direction (FM\textsubscript{a}), the $b_m$ direction (FM\textsubscript{b}), the $c_m$ direction (FM\textsubscript{c}), and in-between $a_m$ and $c_m$ directions (FM\textsuperscript{*}) in the monoclinic notation. Note that the FM moment along the hexagonal $c_H$ axis is not allowed by the monoclinic symmetry $Cm$. Because the degree of lattice distortion is relatively small, the size of the ordered moment per Co is about 0.35 $\mu_B$ in all the cases: without SOC, FM\textsubscript{a}, FM\textsubscript{b}, and FM\textsuperscript{*}. This value is consistent with the theoretical moment along the ferromagnetic order with moment along the hexagonal $c_H$ axis in the undistorted rhombohedral structure\textsuperscript{29}. Among all these four symmetry allowed FM states, FM\textsubscript{a} is
found to be most stable, followed by FM$_c$ ($\approx 0.3$ meV/f.u. higher than FM$_a$) and then FM$_b$ ($\approx 0.6$ meV/f.u. higher than FM$_a$). The energy of FM* depends on the angle between the $a_m$ axis and the moment direction in the $a_m c_m$ plane. It continuously increases from the energy of FM$_a$ at zero angle to the one of FM$_c$ when the moment is along the $c_m$ axis. Thus, the long-range monoclinic distortion could induce a spin reorientation by $\approx 19^\circ$ from the hexagonal $c_H$ axis to the monoclinic $a_m$ axis in the most stable ground state FM$_a$. Given that there is only local monoclinic distortion in Co$_3$Sn$_2$S$_2$, this indicates that while the average long-range ferromagnetic moment points to the hexagonal $c_H$ axis, the local FM moment along the $c_H$ axis may become unstable since it is forbidden by the local symmetry $Cm$. Instead, the local FM moment may deviate away from the $c_H$ axis and has the tendency to reorientate to the local monoclinic $a_m$ axis below $T_C$, as illustrated in the right panel of Fig. 1(b). Thus, the structural instability indicated by the local symmetry breaking below $T_C$ is accompanied with the local changes in ferromagnetic moment direction and stability.

Figure 6(b) compares the electronic band structure of the most stable FM$_a$ order with moment along the monoclinic $a_m$ axis with and without SOC in the monoclinic distorted structure. Momentum is taken along high-symmetry lines as shown in Fig. 6(c). These band structures resemble those of undistorted Co$_3$Sn$_2$S$_2$ as shown in Fig. 6(e), where the momentum is also taken along the corresponding high-symmetry lines in Fig. 6(f). Other FM states such as FM$_b$, FM$_c$, and FM* have similar band structures to that of FM$_a$ (shown in Fig S3 in the SM). To determine the existence and the location of Dirac points or nodal lines or Weyl points, we use the Wannier90 code and derive the maximally-localized Wannier functions. Detailed analysis was carried out using effective tight binding models with the WannierTools package. Similar to the band structure in
the undistorted rhombohedral structure \(^2\text{9}\), there appear Dirac nodal lines without SOC. However, because of the monoclinic distortion, there is only one mirror plane (shown as a gray plane in Fig. 6(c)), resulting in one nodal ring shown by green lines in Fig. 6(c). SOC lifts the band degeneracy along this nodal line shown as green circles in Fig. 6(b) but keeps degeneracy at two points along the nodal ring. These two points form a pair of Weyl points. In the FM\(_a\) phase, there appear an additional 4 pairs of Weyl points that are not protected by the existing mirror symmetry. The existence and position of Weyl points are intimately related to the orientation of magnetic moment.

Because the ordered moment is parallel to the mirror plane, the FM\(_a\), FM\(_c\) and FM\(^*\) phases break the mirror symmetry but maintain mirror+time-reversal symmetry. Thus, these states have Weyl points along the nodal ring. On the other hand, since magnetic moment in the FM\(_b\) state is perpendicular to the mirror plane, this state maintains the mirror symmetry. Therefore, the original nodal ring is protected in the FM\(_b\) case (see Fig. S4 for details). In all cases, FM\(_a\), FM\(_b\), FM\(_c\), and FM\(^*\), there appear additional Weyl points away from the mirror plane. Results for FM\(_b\) and FM\(_c\) are presented in the Fig. S4 in the Supplemental Material.

The consequences of local monoclinic distortion could be twofold: 1). Because two mirror planes out of three are lacking depending on the monoclinic axes, two pairs of Weyl points associated with those mirror symmetries are eliminated inside monoclinic regions. This could, in turn, induce finite broadening at bulk Weyl points. 2). At the boundary between a bulk hexagonal region and a local monoclinic region, or between monoclinic regions with different crystallographic axes, Fermi arcs could be formed because Weyl points in monoclinic regions are gapped out. However, since the volume of each monoclinic region (or boundary area) is small, detecting such individual
Fermi arc would be beyond the resolution of current measurements. Instead, broad Fermi arc like feature may be formed in the bulk electronic structure. Such a feature could be detected by bulk sensitive probes, such as angle-resolved photoemission spectroscopy. An accurate description of the electronic structure by considering local symmetry $Cm$ within the global undistorted symmetry $R-3m$ is beyond the scope of the standard DFT capability since this requires a large supercell. Nevertheless, our DFT calculation demonstrates how the local monoclinic distortion could influence the electronic band structure and Weyl points by breaking mirror symmetries.

In summary, we discovered a hidden local symmetry breaking and demonstrated its impact on the local spin configuration and electronic band topology in the Weyl semimetal and kagome magnet $\text{Co}_3\text{Sn}_2\text{S}_2$. The intrinsic geometric frustration in the kagome lattice of cobalt and the ferromagnetic interactions may be responsible for the occurrence of the local symmetry breaking below $T_C$ whereas the global rhombohedral symmetry is unchanged. The local symmetry breaking provides some new insight to help understand the previously irreconcilable magnetic properties for instance the origins of the spin glass and phase separation. Although the average ferromagnetic moment points to the hexagonal $c_H$ axis, such local monoclinic symmetry has the tendency to induce a local ferromagnetic reorientation away from the original $c_H$ axis and to play a detrimental role in the formation of the Weyl points by breaking mirror symmetries. Our study offers an opportunity to pursue advanced theoretical calculations on the accurate electronic structures on the nonmagnetic/magnetic topological Dirac/Weyl semimetals involving the locally lower symmetry and to make a more careful interpretation on the experimental observation of their electronic and topological properties by the necessary consideration of the overlooked and distinct local symme-
Methods

Sample preparation and half polarized neutron diffraction. The preparation details of both polycrystalline and single crystalline samples were reported previously \(^{29}\). The crystal was aligned in the \((HK0)\) plane for the neutron experiments. The 1st half polarized neutron diffraction measurements were performed on the triple-axis neutron spectrometer HB-1 installed at the High Flux Isotope Reactor at Oak Ridge National Laboratory. For full polarization analysis, Heusler alloy (111) crystals were used as monochromator and analyzer. A flipping ratio of \(\approx 9\) was observed at nuclear reflections. For half polarization analysis, Heusler alloy (111) and Pyrolitic Graphite (002) crystals were used as monochromator and analyzer, respectively. A vertical magnetic field of 3 T cryomagnet was applied to point along the \(c_H\) axis to align the ferromagnetic domains and saturate the magnetization of the sample. The horizontal collimator sequence was 48’-80’-sample-60’-240’ with a fixed incident neutron energy of 13.5 meV. The contamination from higher-order beams was effectively eliminated using PG filters.

To double check the ferromagnetic order and moment size, another half polarized neutron diffraction experiment on a different piece of crystal was conducted at time-of-flight instrument HYSPEC at the Spallation Neutron Source located in Oak Ridge National Laboratory. The crystal aligned with \((HK0)\) reciprocal-lattice plane within the horizontal scattering plane, was loaded inside a permanent-magnet yoke that provided a vertical magnetic field of \(\sim 0.7\) T, oriented parallel to the \(c_H\) direction. This assembly was loaded in a bottom-loading closed-cycle refrigerator.
An incident beam with the incident energy $E_i = 20$ meV (wavelength $\lambda = 2.02$ Å) was polarized using a Heusler monochromator, and a Mezei flipper was used to flip the spin direction of the incident neutron beam. Measurements were carried out with the Fermi chopper operated at 60Hz frequency, providing an energy resolution with FWHM of about 3.4 meV at the elastic position. The effective polarization factor of the incident neutron beam was estimated using the scattering from a Heusler single-crystal probe to be about 80%. The data were collected using 3He linear position-sensitive tube detectors assembled into 20 sets of 8-packs that covered an angular range of 60° in the horizontal scattering plane and a vertical acceptance of +/- 7°. Flipping ratios were obtained from successive measurements with polarized beam oriented parallel and anti-parallel to the external magnetic field.

**Powder Neutron diffraction and pair distribution function (PDF) experiments.** High resolution neutron diffraction and pair distribution function (PDF) patterns were collected simultaneously using the neutron band with the center wavelength of 0.8 Å at powder diffractometer POWGEN, located at Spallation Neutron Source, Oak Ridge National Laboratory. Data collection was conducted at a series of temperatures from 6 K to 300 K. Rietveld analysis on the neutron diffraction data with a wide $Q$ coverage 1.3-16 Å was performed using GSAS-II package $^{39}$ to obtain the temperature dependence of the lattice constants, bond distances and angles.

**Reverse Monte Carlo (RMC) analysis on neutron total scattering data** 12×12×6 supercell configurations were built from the average structure corresponding to each temperature point and they were used as the starting configuration for RMC modeling. The RMC method relaxes structure configuration in a data-driven manner, following the Metropolis routine. For each temperature
point, total scattering data in both $Q$ and $r$-space together with the Bragg scattering data were used for RMC modeling to provide constraints to the model. Typically, PDF data in real-space focuses more on local structure while $Q$-space and Bragg data focuses more on average structure. Here, the Bragg data for all temperature points were first refined with a Rietveld approach using Topas software, for which the RMCProfile package has an interface to read in peak profiles and background. Concerning the RMC modeling, the $12 \times 12 \times 6$ supercell contains 18, 144 atoms and for 250 K and 160 K datasets, $\sim$300 moves per atom were tried among which $\sim$30 moves per atom were accepted on average. For the 6 K dataset, due to the complexity in local environment, the statistics is higher, given $\sim$900 moves per atom were tried among which $\sim$90 moves per atom were accepted. Given that there are multiple datasets involved in RMC modeling, an automatic weight assigning scheme was used-for details, one can refer to the appendix in Ref.34.

The structural distortion modes induced by irreducible representations of the parent rhombohedral $R-3m$ space group and the symmetry-allowed magnetic structures for the monoclinic $Cm$ space group were analyzed using ISODISTORT. The local symmetry $Cm$ in $T < T_c$ determined from modeling total scattering data is well consistent with the monoclinic distortion mode derived from the ISODISTORT.

**Density function theory (DFT) calculations.** Density function theory (DFT) calculations were carried out using the projector augmented wave method with the generalized gradient approximation in the parametrization of Perdew, Burke, and Enzerhof for exchange correlation as implemented in the Vienna ab initio simulation package (VASP). For Co and S standard potentials were used (Fe and S in the VASP distribution), and for Sn a potential, in which $d$ states were
treated as valence states, is used ($S_{nd}$). We use a $12 \times 12 \times 12$ $k$-point grid and an energy cutoff of 500 eV. The $+U$ correction is not included because Co$_3$Sn$_2$S$_2$ is an itinerant magnetic system. We use SeekK-path $^{44,45}$ to generate the primitive unit cell, that contained minimal number of atoms, i.e., three Co, two Sn and two S.

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**Author contributions**  Q.Z. initialized the project. J. Y. prepared single crystal, M.A.M. synthesized powder sample. Q.Z., M.M., V.O.G., D.A.T. conducted powder and single crystal neutron experiments. Q.Z. did Rietveld analysis on the powder neutron diffraction patterns, analyzed the single crystal neutron data and did the symmetry analysis. Y.Z. did the RMC analysis on total scattering data, S.O. performed the DFT calculations, Q.Z. wrote the paper with the input from all the authors.
**Competing Interests**  The authors declare that they have no competing financial interests.

**Correspondence**  Correspondence and requests for materials should be addressed to Q.Z. (email: zhangq6@ornl.gov) and Y.Z. (email: zhangy3@ornl.gov)
(a) Rhombohedral $R-3m$

- FM
- AFM
- Spin glass; exchange bias;
- Phase separation

$T_A$ (135 K)

$T_C$ (175 K)

(b) $T > T_C$
- Global symmetry: $R-3m$
- Local symmetry: $R-3m$

$T < T_C$
- Global symmetry: $R-3m$
- Local symmetry: $Cm$

(c) $P1$, #1
- 250 K

$Cm$, #8
- 160 K

$R-3m$, #166
- 6 K

0.01 → Atomic position tolerance in Å → 0.06
Figure 1  (a). Crystal structure and the magnetic properties of Co$_3$Sn$_2$S$_2$ based on previous reports. The color change of the horizontal bar in $T_A < T < T_C$ indicates the temperature evolution of the FM volume fraction (aqua) and AFM volume fraction (purple). (b). The global and local symmetries of Co$_3$Sn$_2$S$_2$ in $T > T_C$ and $T < T_C$ temperature regions. The gray hexagonal unit cell is also displayed in the middle panel to show the conversion between the hexagonal $R-3m$ and distorted monoclinic $Cm$ unit cells. The arrows in the monoclinic unit cell illustrate the schematic displacements of Co1 and Co2 atoms in the $a_mC_m$ plane and one 3D direction, respectively. The right panel illustrates a tendency of the local spin reorientation from the $c_H$ axis to the $a_m$ axis in $T < T_C$. (c). Space group extraction from collapsed unit cells for the neutron diffraction and PDF datasets at 250, 160 and 6 K in Co$_3$Sn$_2$S$_2$.

Figure 2  Half-polarized neutron diffraction results from the HB1 triple-axis spectrometer: $\theta - 2\theta$ scan of the up-spin diffraction intensity $I^+$ and down-spin diffraction intensity $I^-$ of (110) peak at (a) 210 K, (b) 158 K and (c) 1.5 K. Temperature dependence of (d) the integrated intensity of $I^+$ and $I^-$, (e) the flipping ratios $R=I^+/I^-$ (HYSPEC data was added for comparison), and the difference $I^+-I^-$. (f) Contour plot of the PDF patterns vs temperature.

Figure 3  Temperature dependence of the lattice constants $a$ and $c$, (b) the volume of one unit cell, (c) the bond lengths Co-Sn(1), Co-Sn(2) and Co-S within one octahedra, and (d) the Co-Sn(1)-Co, Co-S-Co bond angles.
Figure 4  (a) Overview of PDFs at various temperature points and the insets are zoom-in over the regions around 5 Å. (b) Partial PDFs of all atomic pairs extracted from the fitted model corresponding to datasets at 250 K, 160 K and 6 K. (c), (d) and (e) shows the PDF fitting results for the datasets at 250 K, 160 K and 6 K, respectively - the $R$-factor of the fitting, as calculated by $\sum_i (y^o_i - y^c_i)^2 / \sum_i (y^o_i)^2$ (where $y^o_i$ and $y^c_i$ represents the observed and calculated PDF intensity, respectively), is presented in each figure.

Figure 5  (a) & (b) The Co-Co-Co and Co-Sn-Co bond angle extracted from the structural model obtained through RMC fitting to the datasets at 250, 160 and 6 K. The inset in each figure illustrates the corresponding bond angle, given the kagome lattice in the $a_H b_H$ plane (refer to the axis illustration on the left). (c), (d) and (e) is the KDE result for the atomic distribution for the cobalt kagome lattice and the central Sn2 atom in the collapsed unit cell at 250 K, 160 K and 6 K, respectively. The inset in each of (c-e) is a zoom-in of the KDE for the central Sn2 atom in the cobalt kagome lattice (scales for the color maps are $1E - 5 \rightarrow 1E - 3$, $1E - 5 \rightarrow 1E - 3$, and $1E - 5 \rightarrow 5E - 2$, respectively)

Figure 6  (a) Magnetic structure and (b) electronic band structure of the most stable FM$_a$ phases in the distorted structure of Co$_3$Sn$_2$S$_2$. Red (blue) lines are majority (minority) spin bands without SOC, and black lines are results with SOC in the FM$_a$ phase, where Co moment is parallel to the $a_m$ axis. SOC lifts the degeneracy at the band crossing as shown as green circles. (c) First Brillouin zone of the distorted structure. Green lines indicate a nodal ring protected by the mirror symmetry without SOC. The mirror plane is indicated
as a gray plane. Red (blue) spheres are Weyl points with positive (negative) chirality in the FM$_a$ phase. There are two Weyl points along the nodal ring. Reciprocal lattice vectors $\mathbf{b}_{1,2,3}$ as well as Cartesian coordinates $x,y,z$ are also indicated. Crystallographic axes $a$ and $b$ for the primitive cell are parallel to $x$ and $y$, respectively. For comparison, panel (d-f) show the corresponding figures of undistorted structure. Three mirror planes are indicated by blue, pink and gray planes.
Table 1: Refined structural parameters for the local monoclinic symmetry with space group $Cm$ (No.8) at 6 K. The lattice constants are refined to be $a_m = 9.2752 \, \text{Å}$, $b_m = 5.3594 \, \text{Å}$ and $c_m = 5.3561 \, \text{Å}$, $\beta = 125.2^\circ$.

| atom | Wyckoff Site | x       | y       | z         |
|------|--------------|---------|---------|-----------|
| Co1  | 2a           | 0.00162 | 0       | 0.50067   |
| Co2  | 4b           | 0.75136 | 0.24971 | 0.00040   |
| Sn1  | 2a           | -0.00008| 0.00000 | -0.00243  |
| Sn2  | 2a           | 0.49945 | 0.00000 | 0.50133   |
| S1   | 2a           | 0.71915 | 0.00000 | 0.28943   |
| S2   | 2a           | 0.28424 | 0.00000 | 0.70960   |
Supplementary Information for

Local symmetry breaking with correlation to ferromagnetism and the Weyl state in $\text{Co}_3\text{Sn}_2\text{S}_2$

Qiang Zhang$^{1,*}$, Yuanpeng Zhang$^{1,*}$, Masaaki Matsuda$^1$, Vasile O Garlea$^1$, Jiaqiang Yan$^2$ Michael A. McGuire$^2$, D. Alan Tennant$^{1,3,4,*}$, & Satoshi Okamoto$^2$

$^1$Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA

$^2$Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA

$^3$Quantum Science Center, Oak Ridge, Tennessee 37831, USA

$^4$Shull Wollan Center, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA

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**Figure 1**  Rietveld analysis on the high resolution neutron diffraction patterns in a wide Q region at (a) 300 K, (b) 150 K and (c) 15 K

**Figure 2**  (a-c) The F(Q) fitting results for datasets corresponding to 250 K, 160 K and 6 K, respectively. (d-f) The Bragg fitting results for datasets corresponding to 250 K, 160 K and 6 K, respectively. The $R$-factor of the fitting, as calculated by $\sum_i (y^o_i - y^c_i)^2 / \sum_i (y^o_i)^2$ (where $y^o_i$ and $y^c_i$ represents the observed and calculated PDF intensity, respectively), is presented in each figure.

**Figure 3**  Electronic band structures in FM states. (a) without SOC (red and blue lines are for majority spin bands and minority spin bands, respectively), (b) FM$_a$ with SOC (Co spins along the $a_m$ axis), (c) FM$_b$ with SOC (Co spins along the $b_m$ axis), and (d) FM$_c$ with SOC (Co spins along the $c_m$ axis). (e) and (f) are magnified views of panel (c) near the nodal ring.

**Figure 4**  Nodal lines, Dirac points, and Weyl points in FM states. (a) Results without SOC. Green lines in the mirror plane (gray plane) form a nodal ring. Green dots away from the mirror plane are Dirac points that are not protected by the mirror symmetry. (b) results for FM$_a$ with SOC, (c) FM$_b$, and (d) FM$_c$. In panels (b) and (d), green lines are the nodal lines appearing without SOC. Red (blue) spheres are Weyl points with positive (negative) chirality. In these cases, there is one pair of Weyl points in the mirror plane or on the nodal ring without SOC. Away from the mirror plane, there are four pairs of Weyl points in FM$_a$ and six pairs of Weyl points in FM$_c$. In FM$_b$, nodal ring is protected by the
mirror symmetry as indicated by green lines in panel (c), with additional ten pairs of Weyl points away from the mirror plane.