Evolving Devil’s Staircase Magnetization from Tunable Charge Density Waves in Nonsymmorphic Dirac Semimetals

Ratnadwip Singha, Tyger H. Salters, Samuel M. L. Teicher, Shiming Lei, Jason F. Khoury, N. Phuan Ong, and Leslie M. Schoop*

While several magnetic topological semimetals have been discovered in recent years, their band structures are far from ideal, often obscured by trivial bands at the Fermi energy. Square-net materials with clean, linearly dispersing bands show potential to circumvent this issue. CeSbTe, a square-net material, features multiple magnetic-field-controllable topological phases. Here, it is shown that in this material, even higher degrees of tunability can be achieved by changing the electron count at the square-net motif. Increased electron filling results in structural distortion and formation of charge density waves (CDWs). The modulation wave-vector evolves continuously leading to a region of multiple discrete CDWs and a corresponding complex “Devil’s staircase” magnetic ground state. A series of fractionally quantized magnetization plateaus is observed, which implies direct coupling between CDW and a collective spin-excitation. It is further shown that the CDW creates a robust idealized nonsymmorphic Dirac semimetal, thus providing access to topological systems with rich magnetism.

1. Introduction

The quest for finding novel quantum phases drives the discovery of new materials and phenomena in condensed matter physics. The introduction of topological band structure in material classification is perhaps the most notable example in the last decade. Starting from the identification of the topological insulating state in a handful of materials,[1–4] this field has been enriched by subsequent realization of a plethora of unconventional quantum states such as Dirac,[5,6] Weyl,[7–9] nodal-line semimetals,[10,11] and topological superconductors.[12] While the idea of exploring the dynamics of relativistic particles in low-energy electronic systems is already quite enticing, the prospect of probing new quasiparticle excitations beyond the realm of particle physics[13] is even more attractive. Motivated by these predictions, researchers have utilized data mining to identify and categorize a huge number of compounds hosting different types of nontrivial topological band structures.[14,15] To realize new topological phases, another effective route is to introduce additional tuning parameters in these materials such as strong electron correlations[16,17] and magnetism. Incorporation of magnetism in topological systems is particularly interesting as it can break the inherent time-reversal symmetry (TRS) of the crystal structure. TRS broken topological semimetals are extremely rare and have been the major focus of a number of studies.[18–21] Contrary to the approach of scanning through material databases, it has been shown that simple structural motifs can provide strong indications of topological nontrivial states in a compound.[22] A great example of this is square-net materials. In 2015, Young and Kane proposed nonsymmorphic symmetry protected Dirac semimetal states in a 2D square-net lattice.[23] In recent times, a vast array of topological semimetals has been discovered, all hosting the 2D square-net motif in their crystal structure.[24] Probably the most extensively studied member of this family is ZrSiS, which shows the largest reported energy range (≈2 eV) of linearly dispersive bands.[11] It hosts multiple Dirac nodes near the Fermi energy, which form a diamond shaped nodal-line in momentum space.[12,25,26] In addition, both far below and above the Fermi energy, there are nonsymmorphic symmetry protected Dirac nodes, which, in contrast to other Dirac cones, remain gapless even under spin–orbit coupling.[11] Thus, ZrSiS presents a unique platform to explore different types of topological physics. CeSbTe is an isostuctural compound, which in addition to having a rich band structure similar to ZrSiS, introduces magnetism as an additional controllable parameter.[27] In fact, it has been reported that this material can be driven through distinct topological phases while changing the magnetic ordering by modulating the strength of an applied magnetic field, resulting in a tunable topological...
2. Results and Discussion

2.1. Evolution of the Crystal Structure and the Charge Density Wave

Powder X-ray diffraction (XRD) spectra along with LeBail fitting for the crushed single crystals with different compositions are shown in Figure S1a, Supporting Information. Within the experimental resolution, no secondary phases are detected. Figure 1 illustrates the extracted lattice parameters as functions of Sb content at the square-net. These parameters are also listed in Table S1, Supporting Information. We could not prepare any CeSb$_{0.91}$Te$_{0.91}$ crystal with $x > 0.91$. Therefore, the parameters for CeSb$_{0.91}$Te$_{0.91}$ are obtained from an earlier report. Several other parameters from ref. [27] are also used throughout this work in order to put the results in context. CeSb$_{0.91}$Te$_{0.91}$ crystallizes in the ZrSiS-type tetragonal structure (spacegroup $P4/nmm$). Our data for CeSb$_{0.91}$Te$_{0.91}$ are similar to those for ideal stoichiometric CeSbTe. With decrease in the Sb content, the distorted crystal structure is better described using the orthorhombic spacegroup $Pmmn$. This also explains the orthorhombic structure observed by Wang et al. [34] and Lv et al. [35] for “CeSbTe” crystals, which had a significant vacancy at the Sb-site. From our analysis, we identified a boundary between $x = 0.63$ and 0.74 for the tetragonal to orthorhombic phase transition. Without more data point in this doping range, however, it is not possible to precisely locate this phase boundary. We note that the orthorhombic structure has also been reported in few other members of the LnSbTe-family. [31,36,37] The lattice parameters undergo a smooth transition across this phase boundary. Two local maxima at $x = 0.91$ and 0.34 are observed for both lattice constants $c$ and $b$, respectively. While the peak in $c$ might indicate a deviation from ideal stoichiometry for CeSbTe crystals in ref. [27], the maximum in $b$ represents a phase transition from multiple to one CDW modulation vectors, as we will show later. Signatures of satellite peaks in the XRD spectra, which indicate the presence of the CDW, can be seen in all orthorhombic samples.

The structural evolution of the CDW in CeSb$_{x}$Te$_{2-x}$-δ is investigated by single-crystal XRD. The presence of a CDW is confirmed by the weak intensity satellite peaks as observed in precession images (Figure 2a, Figure S1b, Supporting Information). The solved crystal structures for all the compositions are investigated by single-crystal XRD. The presence of a CDW is confirmed when there is a deviation from nominal stoichiometry. For example, in LaSb$_{2-x}$Te$_{2}$, the CDW has been observed to evolve continuously with antimony substitution.[31] On the other hand, in GdSb$_{2}$Te$_{2}$-δ, the CDW is shown to have an important role in designing new Dirac semimetals.[32] Recently, the signature of a fivefold CDW modulation has also been reported in an off-stoichiometric single crystal of CeSbTe.[33] However, the detailed evolution of this phase and its impact on the topological band structure remains unexplored. Such an investigation is important to reveal the true potential of doped LnSbTe, in respect to their magnetic, structural, and topological properties as well as their interplay.

Herein, we combine structural and magnetic measurements with theoretical calculations to probe the interplay of magnetic ordering, CDW, and topological band structure in single-crystalline CeSb$_{x}$Te$_{2-x}$-δ ($\delta$ represents the vacancy concentration in the crystal). We show that there are two distinct regimes in this material with formation of either one or multiple CDW orderings. Moreover, it is possible to continuously tune the modulation wave-vector by controlling the electron filling at the square-net position. The corresponding unique distortions of the square-net motif lead to modifications in the electronic band structure. While CeSbTe has a simple antiferromagnetic ordering, CDW, and topological phases, accessible via Fermi level engineering by electron filling.

The distorted lattice supports the formation of charge density waves (CDWs), which open a bandgap at the Fermi level. In a few rare earth antimony tellurides (LnSbTe; Ln = lanthanides), the presence of CDWs has been confirmed when there is a deviation from nominal stoichiometry. For example, in LaSb$_2$Te$_2$, the CDW has been observed to evolve continuously with antimony substitution.[30] On the other hand, in GdSb$_2$Te$_2$-δ, the CDW is shown to have an important role in designing new Dirac semimetals.[32] Recently, the signature of a fivefold CDW modulation has also been reported in an off-stoichiometric single crystal of CeSbTe.[33] However, the detailed evolution of this phase and its impact on the topological band structure remains unexplored. Such an investigation is important to reveal the true potential of doped LnSbTe, in respect to their magnetic, structural, and topological properties as well as their interplay.

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single modulation wavevector $\mathbf{q}$ is observed in the $b$-direction (the longer axis of the $ab$-plane) of the parent structure and it corresponds to a threefold expansion in CeSb$_{0.11}$Te$_{1.90}$ and a fivefold expansion in the CeSb$_{0.51}$Te$_{1.40}$ unit cell. As can be seen in Figure 1, the lattice constant $b$ also shows a change in slope around this threefold to fivefold transition ($x = 0.51$). More complex behavior emerges in the low Sb-composition range. CeSb$_{0.11}$Te$_{1.90}$ exhibits three different $\mathbf{q}$-vectors (one within the $ab$-plane, $\mathbf{q}_1 = b*/3$; and two having component along the $c$-axis, $\mathbf{q}_2 = a*/3 + b*/3 + c*/2$ and $\mathbf{q}_3 = a*/3 + b*/3 - c*/2$, where $a^*$, $b^*$, and $c^*$ are the reciprocal lattice vectors) corresponding to multiple distinct CDW orderings, which result in an overall fivefold expansion in the CeSb$_{0.51}$Te$_{1.40}$ unit cell. As evident from Figure S1b, Supporting Information, no CDW ordering is observed in tetragonal CeSb$_{0.79}$Te$_{1.05}$. The results of the structural solution for all compositions are summarized in Table S2, Supporting Information.

2.2. Magnetic Phase Diagrams

In Figure 3a, we have plotted the temperature dependence of the magnetic susceptibility ($\chi$) of tetragonal (unmodulated) CeSb$_{0.91}$Te$_{0.91}$ for different magnetic field strengths, applied along the crystallographic $c$-axis. In general the results are very similar to those observed in CeSbTe,

$$
\chi = \frac{N_s \mu_{eff}^2 k_B}{3 k_B (T - \theta_{CW})}
$$

for some representative
compositions (Figure S2, Supporting Information). While $\theta_{\text{CW}}$ is negative for higher Sb-compositions as expected for AFM ordering, it changes sign below $x \approx 0.50$ indicating some FM component (Figure 3d). This might not be unexpected as we note that the magnetization in CeTe$_2$ has been reported to be quite complex. There are conflicting reports of a ferrimagnetic ground state with positive $\theta_{\text{CW}}$[39,40] as well as a long range AFM ground state coexisting with a short range FM ordering.[38] From Curie–Weiss fitting for all the compositions, the effective moment ($\mu_{\text{eff}}$) is calculated to be close to the moment ($2.54 \mu_B$) of the free Ce$^{3+}$ ion. Nevertheless, some deviation from the theoretical value is observed, which is possibly due to the strong crystal electric field effect.[41]

To get the complete picture, we have constructed magnetic phase diagrams from the first-order derivative of $\chi(T)$ curves with the field parallel to the $c$-axis for the entire electron filling range (Figure 4a–c; Figure S3a, Supporting Information). From the results, it is clear that there are three distinct regimes, represented by three compositions in Figure 4a–c. For $x = 0.11$ (orthorhombic with three $q$-vectors), in addition to the AFM and FM states that are similar to the ones in tetragonal CeSbTe, we observe two new phases, namely “Phase I” and “Phase II”. Among these, Phase II is a field-induced state, whereas the boundary for Phase I extends toward lower fields and becomes indistinguishable from the AFM phase boundary. To identify the true magnetic ground state, we have performed a zero-field heat capacity ($C_P$) measurement on a CeSb$_{0.11}$Te$_{1.90}$ crystal (Figure S4a, Supporting Information). From the enlarged view of the low-temperature region in Figure S4b, Supporting Information, we can clearly see an additional peak in $C_P$ adjacent to the AFM transition, confirming that Phase I coexists even at zero-field. Upon application of a magnetic field, this second peak becomes more prominent as also evident from the emergence of an explicit boundary with field in the magnetic phase diagram. Both of these peaks in $C_P$ are suppressed completely at 0.5 T, indicating a fully spin-polarized state. With increasing Sb-content at $x = 0.34$ (orthorhombic with threefold modulation along the $b$-axis), Phase II disappears, whereas Phase I still remains. For tetragonal CeSb$_{0.74}$Te$_{1.25}$, however, the phase diagram becomes much simpler and closely related to CeSbTe.[27] In Figure 4d, we have plotted the doping-dependent phase diagram. From this figure, we can correlate the boundaries...
for both new magnetic phases with the structural transitions. Phase II only appears in presence of multiple CDW orderings, whereas Phase I only exists in orthorhombic structures. The previously published magnetization data for CeTe$_2$ suggests that Phase I probably corresponds to short range FM ordering.[38] On the other hand, Phase II represents a more complex magnetic state as we discuss below.

Contrary to the complex magnetic structure for field along the $c$-axis, the phase diagrams for all the compositions turn out to be quite simple, when the magnetic field is applied along the $ab$-plane (Figure S3b, Supporting Information). In this configuration, only three states have been observed (AFM, FM, and PM), similar to the $c$-axis phase diagram of CeSbTe$_2$.[27]

**Figure 5**a shows the magnetization ($M$) as a function of magnetic field ($H \parallel c$-axis) for tetragonal CeSb$_{0.91}$Te$_{0.91}$ at different temperatures both below and above $T_N$. To compare, the $M(H)$ curve at 2 K for $H \perp c$-axis is also plotted in the same graph. The material reaches the FM state at lower field, when $H \parallel c$-axis, confirming that it is the easy axis of magnetization. Similar to in CeSbTe, we resolve the “spin-flip” ($H \parallel c$-axis) and

![Figure 4](image-url)

**Figure 4.** Magnetic phase diagrams of CeSb$_x$Te$_{2-x}$−δ. a–c) Magnetic phase diagram for three different Sb-content samples, showing three distinct magnetization regimes for CeSb$_x$Te$_{2-x}$−δ. The discrete points and red curve, obtained from the magnetization data, represent the boundary between FM and PM states. d) Doping-dependent phase diagram.

![Figure 5](image-url)

**Figure 5.** Field dependence of magnetization for CeSb$_{0.91}$Te$_{0.91}$. a) Magnetization ($M$) at different temperatures as a function of field ($H$), applied along the $c$-axis. For comparison, the $M(H)$ curve at 2 K for $H \perp c$-axis is also plotted. b) The low-field region of the $M(H)$ curves, showing “spin-flip” (top) and “spin-flop” (bottom) transitions for two different applied field directions. The arrows illustrate the spin arrangement for two consecutive Ce-layers.
“spin-flop” ($H \perp c$-axis) transitions (Figure 5b: arrows illustrate the spin arrangements between two consecutive Ce-layers), as well as strong magnetocrystalline anisotropy.

In Figure S5a, Supporting Information, we have compared the $M(H)$ curves of all Sb-compositions for two measurement configurations. It is evident that the magnetocrystalline anisotropy changes systematically with the electron filling, whereas the direction of spin-flip and spin-flop transitions remain unaltered, indicating that the spins are still aligned along the $c$-axis. For $x = 0.11$, the nature of anisotropy is completely opposite to that for $x = 0.91$. This suggests that the electron filling at the Sb square-net position continuously tunes the AFM exchange interaction between two consecutive Ce-layers (nearest neighbors). This is indeed expected, as the square-net motif in CeSb$_{x}$Te$_{2-x}-\delta$ provides the conduction electrons, required for Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction between localized Ce$^{3+}$ moments at two different layers. We find that the anisotropy reverses at a composition close to $x = 0.50$, where the Curie–Weiss temperature also changes its sign indicating predominantly the AFM ground state for $x \geq 0.50$. It is possible that below this critical Sb-content, with higher electron filling, the FM interaction between two next nearest neighbor layers starts to dominate and contributes to a positive Curie–Weiss temperature.

2.3. Fractionally Quantized Magnetization Plateaus

An even more surprising magnetic state appears at high electron filling. Figure S5b, Supporting Information, shows the low-field region ($-0.5 \, T \leq H \leq 0.5 \, T$) of the $M(H)$ curves for samples with $x \leq 0.51$, when $H \parallel c$-axis. For $x = 0.11$ and 0.20 (multiple $q$-vector region), we observe cascades of metamagnetic transitions, represented by a series of plateaus and steps in magnetization, leading up to the spin-flip transition. In addition, there is prominent hysteresis between the increasing and decreasing-field measurements, confirming the presence of a FM component. The metamagnetic transitions significantly weaken and then completely disappear at $x \geq 0.34$. In Figure 6a, we have plotted the $M(H)$ curve at 1.8 K for CeSb$_{0.11}$Te$_{1.90}$, normalized by the saturation magnetization ($M_S$). Remarkably, the plateaus are found to be locked to rational fractions identical to the quantized Hall resistivity in the fractional quantum Hall effect (FQHE). Quantized magnetization plateaus have been previously observed in low-dimensional magnets$^{[42,43]}$ and magnetically frustrated systems,$^{[44,45]}$ forming a structure called “Devil’s staircase” as each step consists of infinite number of steps under magnification, and so forth.$^{[46]}$ We note that such complex magnetic state has also been reported in CeSbSe with a tetragonal structure.$^{[47]}$ Analogous to the FQHE, a devil’s

![Figure 6. Devil’s staircase in magnetization of CeSb$_{0.11}$Te$_{1.90}$. a) Normalized magnetization as a function of field, revealing fractionally quantized plateaus. b) Magnetic phase diagram for plateaus corresponding to different quantum numbers. c) Schematic showing the magnetic-field-tunable spin wave, responsible for the devil’s staircase structure.](image-url)
staircase originates from an energy gap in the many body excitation spectra in a compound due to the translational symmetry breaking.\cite{48-51} The plateau appears when a commensurability condition between lattice wave vector and localized excitation is satisfied. For quantum spin systems, this commensurability condition is found to be $n(S-m) = \text{integer}$, where $n$, $S$, and $m$ are number of spins in the unit cell, the magnitude of spin, and magnetization per site, respectively.\cite{58} In principle, this staircase structure can be observed for different physical properties in presence of two coupled waves.\cite{48} A continuous variation in frequency (wavelength) of one wave then drives the frequency of other wave such that they go through regimes of phase-locked (plateau) and non-phase-locked (steps) states.

By repeating the magnetization measurements for CeSb$_{0.11}$Te$_{1.90}$ at several temperatures (Figure S6, Supporting Information), we have tracked the evolution of the plateaus corresponding to different fractions. Using these results, in Figure 6b, a phase diagram is constructed. We conclude that this staircase structure is a magnetic field induced state and only appears within the AFM phase. Interestingly, it is also sensitive to the crystallographic directions, as no metamagnetic transition is observed, when the field is applied perpendicular to the $c$-axis (Figure S7, Supporting Information). Moreover, the doping range ($x = 0.11$ and 0.20), where this complex state is observed also suggests that it corresponds to the Phase II in the magnetic phase diagram. We note that only within this region, there are multiple CDW modulation wave-vectors, two with components along the $c$-axis and the other one residing within the $ab$-plane. Below this electron filling, the CDW wave-vectors along the $c$-axis disappear. All these features help us to construct a microscopic picture of the origin of the devil's staircase structure in CeSb$_{0.11}$Te$_{2.2-\delta}$ as illustrated in Figure 6c.

The AFM ordered $\uparrow\downarrow\uparrow\downarrow$ Ce$_{34}$-layers in CeSb$_{0.11}$Te$_{2.2-\delta}$ form a spin wave along the $c$-axis. With application of a magnetic field perpendicular to the layers, the down spins try to flip in order to align with the field. However, under a low enough field strength, this transition does not occur at all down-spin layers simultaneously. Instead, different layers undergo spin-flip transitions one after another, thus, effectively creating a spin-wave with continuously field-tunable wavelengths. As the magnetic exchange interaction between these layers is mediated by conduction electrons through the RKKY interaction, the spin-wave is already coupled to any modulation in the electron density along the $c$-axis. Moreover, the $c$-axis component of the two CDW wave-vectors is commensurate with the lattice, hence also with the spin-wave modulation-vector. So, by controlling the magnetic field, these coupled waves can be driven to a series of sequential phase-locked and non-phase-locked states. Once all the spins are aligned along the magnetic field (fully spin-polarized state), there is no longer a spin-wave and hence, no more steps are observed. On the other hand, for $x > 0.2$, the absence of a $\gamma$-vector along the $c$-axis causes the steps to disappear. We note that in cubic CeSb, a continuous modulation of the spin-wave with both temperature and field, and its commensurability with lattice wave-vector, have been confirmed to be the origin of the devil’s staircase in $M(T,H)$.\cite{52,53} In particular, the “1/3 plateau” is observed when the layers are arranged in a $\uparrow\uparrow\downarrow$ configuration. Interestingly, two extremely weakly interacting spin-excitations with one of them having analogous properties to astrophysical “dark matter”, have recently been observed within the “1/3 plateau” of CeSb,\cite{54} showing the possibility of exploring new quantum states in the fractional magnetization plateaus. In the case of CeSb$_{0.11}$Te$_{2-\delta}$ we observe another signature of the spin-wave in the magnetic entropy ($S_m$), calculated from the heat capacity data (Figure S4c, Supporting Information). For $x = 0.51$, $S_m$ reaches a saturation value $R\ln 2$, expected for localized Ce$^{3+}$ moments, whereas it is smaller for $x = 0.11$, indicating a gapped spin excitation spectrum.\cite{55,56} It is worth mentioning that the observed reduction in magnetic entropy for $x = 0.11$ could be caused by an enhanced Kondo lattice hybridization between the cerium $f$-electrons and the conduction electrons. However, the electronic coefficient ($\gamma$) of the heat capacity is not very large (Figure S8, Supporting Information), which points to a different origin. We have calculated $\gamma$ for three different compositions ($x = 0.11$, 0.51, and 0.91) using the relation $C_p = \gamma + \beta T^3$, where $\beta$ is the coefficient for the lattice contribution. Linear fits were performed in the temperature range 10–17 K to avoid any magnetic contribution to the heat capacity. The obtained $\gamma$ values (24–40 mJ mol$^{-1}$ K$^{-2}$) are quite small and remain similar throughout the electron filling range. This suggests that there is no enhanced hybridization effect for $x = 0.11$ and the electronic correlation is not significant in CeSb$_{0.11}$Te$_{2-\delta}$. Thus, based on our initial assessment, the Kondo effect and hybridization play a minor role in the system. We do however hope that further studies will look into this possibility more deeply.

Finally, let us discuss the quality of the magnetization plateau in CeSb$_{0.11}$Te$_{1.90}$ as compared to other systems. Such materials usually show a finite number (one or two) of prominent plateaus accompanied by a series of faint transitions. For example, in CeSb, a pronounced plateau is observed at 1/3 of the saturation magnetization that remains flat for a wide field range.\cite{54} On the other hand, in the low-dimensional magnet NH$_4$CuCl$_3$, clean plateaus are found at 1/8 and 3/8 of the saturation magnetization.\cite{52} In the case of CeSb$_{0.11}$Te$_{1.90}$, the first order derivative of $M/M_s$ (Figure S9, Supporting Information) suggests that the 1/3-plateau is the most prominent one, as in CeSbSe.\cite{47} As the AFM transition temperature (4.27 K) is quite low in CeSb$_{0.11}$Te$_{1.90}$, this particular plateau could become more pronounced as we reach a temperature much below $T_N$.

2.4. Topological Properties of the Electronic Band Structure

Next, we investigate the topological nature of CeSb$_{0.11}$Te$_{2-\delta}$. As mentioned above, CeSbTe has been shown to be of interest for several distinct topological semimetal phases, which can be modulated by an applied magnetic field. In fact, non-trivial topological states are either predicted or confirmed in different members of the LnSbTe family.\cite{16,57-59} Recently, the influence of electron count on the topological band structure has been studied in GdSb$_{0.46}$Te$_{1.48}$, where it was shown that an idealized nonsymmorphic Dirac semimetal can be achieved in GdSb$_{0.46}$Te$_{1.48}$.\cite{13} So, it is of interest to also study the band structures of CeSb$_{0.11}$Te$_{2-\delta}$ with respect on their topological nature. We have performed first-principle calculations for three different compositions, representing different regimes of the structural phase diagram (orthorhombic structure with multiple
q-vectors, one q-vector, and tetragonal structure). In the
tetragonal state (CeSb$_{0.91}$Te$_{0.91}$), the band structure (Figure 7a)
unsurprisingly closely resembles one of CeSbTe$_x$,[27] with only a
slight difference in electron filling. For the single q-vector range,
the band structure of CeSb$_{0.51}$Te$_{1.40}$ is shown in Figure 7b, which
features a fivefold supercell along the b-axis. Orthorhombic
CeSb$_{0.51}$Te$_{1.40}$ is isostructural to GdSb$_{0.46}$Te$_{1.48}$[32] and analogous
to this compound, it also hosts close to ideal nonsymmorphic
Dirac semimetal band structure. Nonsymmorphic symmetry
protected gapless Dirac nodes reside near the Fermi energy
($E_F$) at high symmetry points X and U (yellow arrows), whereas
almost all other band crossings gapped out by the CDW and do
not contribute to the low-energy transport response (Figure 7c).
While it seems that some bands still cross $E_F$ (as CDW bandgap
is smaller than in its Gd-counterpart), there are small gaps in
their spectra exactly at the Fermi level. This is interesting for
two reasons: i) it suggests that the mechanism reported in ref. [32] can be universally applied to LnSb$_{x}$Te$_{2-x-\delta}$ phases that are in the single q-vector region with $q \approx 0.20b^*$; ii) it provides
a second idealized nonsymmorphic Dirac semimetal band structure.

Finally, in Figure 7d, we show the band structure of CeSb$_{0.11}$Te$_{1.90}$, which has multiple q-vectors. In this compound,
due to the infinite number of magnetic phases that arise from
the devil’s staircase, the tunability of topological phases becomes
extremely rich. As can be seen in Figure 7d, the band structure
of CeSb$_{0.11}$Te$_{1.90}$ features both nonsymmorphically enforced Dirac
crossing (yellow arrow) as well as nodal-line crossings (green
arrows). Especially, a nodal-line crossing resides exactly at $E_F$
along $\Gamma - S$. In addition, there are multiple nodal-line crossings just above
$E_F$, which should be easily accessible by fine tuning of the electron
filling at the Te-site. Future studies could further investigate
the interplay of the topological band structure and devil’s staircase
magnetism in the multiple q-vector region of LnSb$_{x}$Te$_{2-x-\delta}$.

Although we only tuned the electron filling at the Sb square-
net, the band structure for different compositions does not
follow a complete rigid band model that resembles CeSbTe$_x$.[27]
The reason lies in the structural distortion and formation of
CDW with changing q-vectors. Nonetheless, it is possible to track some of the band degeneracies, notably the non-
symmorphic Dirac node (yellow arrow) at the high symmetry
devil’s staircase point X, which moves from above to below the Fermi energy as
we increase the electron count.

3. Conclusion
We have probed the detailed structural and magnetic properties of
topological semimetals CeSb$_{x}$Te$_{2-x-\delta}$ as a function of electron
filling at the Sb-square-net site. We found that the crystal
structure increasingly transforms from a tetragonal structure at ideal stoichiometry to an orthorhombic one with electron
calculations confirm that CeSb0.51Te1.40 is an ideal magnetic Dirac semimetal with a nonsymmetric Dirac node at the Fermi energy, while almost all other bands are gapped out. Thus we present a unique template material, where the topological band structure can be controlled by electron filling (tuning chemical potential), CDWs (gapping out non-essential band crossings), or magnetic field (changing the magnetic ordering). This also encourages future studies to design new topological states in CeSb1−δTe1+δ and other members of the LnSbTe-family. Furthermore, CeSb0.51Te0.40 also presents the possibility of exploring a wide range of f-electron physics in a topological material. For example, a pressure induced superconducting phase coexisting with AFM ordering, has been reported in off-stoichiometric CeTe1−x[61]. This superconductivity is extremely sensitive to the tellurium vacancy and only exists in very narrow doping range. Therefore, it would be interesting to investigate if such a state can be observed in CeSb1−δTe0.51, which can pave the way for combining topological bands with superconductivity.

4. Experimental Section

**Single-Crystal Growth and Determination of Stoichiometry:** Single crystals of CeSb1−δTe0.51 were grown by chemical vapor transport. Stoichiometric amounts of high-purity Ce (Sigma-Aldrich 99.99%), Sb (Alfa Aesar 99.999%), and Te (Alfa Aesar 99.9999%) along with a few milligrams of iodine (Sigma-Aldrich 99.999%) were placed into a quartz tube. The tube was then evacuated, sealed, and put into a gradient furnace. The furnace was heated such that the hot end of the quartz tube, containing the materials, remained at 950 °C and the other end at 850 °C for 7 days. After cooling, the crystals were mechanically extracted from the cold end of the tube. The elemental composition of the crystals was determined by energy-dispersive X-ray spectroscopy (EDX) in a Verios 460 scanning electron microscope, operating at 15 keV and equipped with an Oxford EDX detector. To determine the elemental compositions, EDX measurements were performed at several randomly selected regions of each crystal. While the obtained values were very close to each other, the averaged numerical values are used as the exact composition throughout the paper. To check the doping consistency in a batch, the EDX measurements were performed on several crystals from one tube. Though the amount of tellurium vacancies changed randomly within a batch as expected, the x values vary only by ~2% in different crystals. Particular care was taken about the composition for the samples used for the magnetic measurement. EDX was performed on the same crystals after finishing the magnetic measurements. Furthermore, it was confirmed that different crystals from one batch show similar magnetic properties and hence the tellurium vacancy did not affect the magnetization in CeSb0.51Te0.40.

**X-ray Diffraction and Magnetic Measurements:** The powder XRD measurements were performed on a STOE STADI P diffractometer operating in transmission geometry using Mo-Kα (λ = 0.71073 Å) source. The single crystals were crushed into powder in an argon-filled glovebox and sealed in glass capillary to use for XRD experiments. The XRD spectra were analyzed by LeBail fitting using FULLPROF software package.

The single-crystal XRD data were collected at 250(1) K with either a Bruker Kappa Apex2 CCD diffractometer or a Bruker D8 VENTURE equipped with a PHOTON CMOS detector, using graphite-monochromatized Mo-Kα radiation. Raw data were corrected for background, polarization, and Lorentz factors as well as a multi-scan absorption correction were applied. Structure solution was carried out via either intrinsic phasing as implemented in the ShelXT program or via charge-flipping as implemented in SUPERFLIP. Commensurately modulated phases for CeSb0.11Te0.90 and CeSb0.51Te0.40; and tetragonal structure of CeSb0.79Te1.05 were able to be indexed fully and could be refined in conventional 3D space groups, using the ShelXL least-squares refinement package in the Olex2 program. In the modulated phase CeSb0.11Te0.90, the satellite peaks refined to a slightly incommensurate modulation vector q = 0.201δ. Refinement was thus carried out using the superspace approach, where the displacive distortion of atomic positions is expressed by a periodic modulation function, yielding a 3 + 1 dimensional space group.[62] Refinements with the superspace approach were carried out in JANA2006. For the refinement, the modulation vector was rounded to a commensurate q = b*/5 without any significant changes to the fitting compared to a fully incommensurate treatment. Determination of Sb/Te occupancy and ordering within the square-net was limited by near-identical scattering power due to their closeness in atomic number (Z = 51 and 52, respectively).[63] At laboratory-accessible X-ray wavelengths, the two species were indistinguishable if present on the same crystallographic site. Thus, for all refinements, atomic occupancies within the Sb/Te square-net were constrained to the stoichiometry derived from EDX. Assignment of ordering in CeSb0.11Te0.90 was based on the chemical bonding intuitions preferring multiple bonds to Sb.

The magnetic measurements were performed using the vibrating sample magnetometer option of a physical property measurement system (Quantum Design).

**Band Structure Calculations:** Density functional theory (DFT) calculations were performed in VASP v5.4.4[64–66] using the PBE functional.[67] Localization of the Ce f-orbitals was corrected by applying a Hubbard potential of U = 6 eV,[68] as in previous work on tetragonal CeSbTe.[27] Though DFT cannot take into account strong electron correlation effects, the experimental results suggested that such effects were not significant in CeSb0.51Te0.40. PAW potentials[69,70] were based on the v5.2 recommendations. Simulations approximating CeSb0.91Te0.09, CeSb0.51Te0.40 and CeSb0.11Te0.90 were performed on the tetragonal CeSb0.91Te0.09 unit cell with full occupancy and supercells of 1 × 5 × 1 CeSb0.91Te0.09 and 3 × 3 × 2 CeSb0.91Te0.09 with Fermi levels adjusted based on the electron counts of the true experimental cells. Calculations employed a plane wave energy cutoff of 400 eV and a k-mesh density, ℓ = 30; corresponding to 7 × 7 × 3 and 7 × 1 × 3 and 2 × 2 × 2 T-centered k-meshes for the tetragonal CeSb0.91Te0.09 subcell and the CeSb0.51Te0.40 and CeSb0.11Te0.90 supercells, respectively. Unfolded spectral functions for the supercells in the subcell BZ were calculated using the method of Popescu and Zunger[71] in VASPBandUnfolding. The Γ–X high symmetry line in CeSb0.51Te0.40 (the only elongated cell for which this is ambiguous) was chosen to lie along the distortion axis. Crystal structures were visualized with VESTA.[72] Spin–orbit coupling effects were not incorporated. See additional computational details in the Supporting Information.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.
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Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

R.S. and L.M.S. initiated the project. R.S. synthesized single crystals and characterized them with input from S.L. R.S. measured and analyzed magnetization data, with input from N.P.O. and L.M.S. T.H.S. performed the band structure calculations. The data that support the findings of this study are available from the Data Availability Statement contributed to writing the manuscript. L.M.S. supervised the project. All authors discussed the results and input from J.F.K. S.M.L.T. performed single-crystal X-ray diffraction experiments and solved structures with magnetization data, with input from N.P.O. and L.M.S. T.H.S. performed characterizations and input from S.L. R.S. measured and analyzed R.S. and L.M.S. initiated the project. R.S. synthesized single crystals and characterized them with input from S.L. R.S. and L.M.S. supervised the project. All authors discussed the results and contributed to writing the manuscript.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

antiferromagnetic dirac semimetals, charge density waves, quantized magnetization plateaus, spin waves

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