A new noncollinear ferromagnetic Weyl semimetal with anisotropic anomalous Hall effect

Hung-Yu Yang, 1 Bahadur Singh, 2 Jonathan Gaudet, 3 Baozhu Lu, 4 Cheng-Yi Huang, 5 Wei-Chi Chiu, 2 Shin-Ming Huang, 6 Baokai Wang, 2 Faranak Bahrami, 1 Bochao Xu, 7 Jacob Franklin, 7 Ilya Sochnikov, 7, 8 David E. Graf, 9 Guangyong Xu, 10 Yang Zhao, 10 Christina M. Hoffman, 11 Hsin Lin, 5 Darius H. Torchinsky, 4 Collin L. Broholm, 3 Arun Bansil, 2, 9 and Fazel Tafti 1, 11

1 Department of Physics, Boston College, Chestnut Hill, MA 02167, USA

2 Department of Physics, Northeastern University, Boston, MA 02115, USA

3 Institute for Quantum Matter and Department of Physics and Astronomy, The Johns Hopkins University, Baltimore, Maryland 21218, USA

4 Department of Physics and Temple Materials Institute, Temple University, Philadelphia, PA 19122, USA

5 Institute of Physics, Academia Sinica, Taipei 11529, Taiwan

6 Department of Physics, National Sun Yat-sen University, Kaohsiung 80424, Taiwan

7 Department of Physics, University of Connecticut, Storrs, CT USA, 06269

8 Institute of Material Science, University of Connecticut, Storrs, CT USA, 06269

9 National High Magnetic Field Laboratory, Tallahassee, FL 32310, USA

10 NIST Center for Neutron Research, National Institute of Standards and Technology, Gaithersburg, MD 20899-6102, USA

11 Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA

(Dated: June 17, 2020)
Abstract

A developing frontier in condensed matter physics is the emergence of novel electromagnetic responses, such as topological and anomalous Hall effect (AHE), in ferromagnetic Weyl semimetals (FM-WSMs). Candidates of FM-WSM are limited to materials that preserve inversion symmetry and generate Weyl crossings by breaking time-reversal symmetry. These materials share three common features: a centrosymmetric lattice, a collinear FM ordering, and a large AHE observed when the field is parallel to the magnetic easy-axis. Here, we present CeAlSi as a new type of FM-WSM, where the Weyl nodes are stabilized by breaking inversion symmetry, but their positions are tuned by breaking time-reversal symmetry. Unlike the other FM-WSMs, CeAlSi has a noncentrosymmetric lattice, a noncollinear FM ordering, and a novel AHE that is anisotropic between the easy- and hard-axes. It also exhibits large FM domains that are promising for both device applications and an interplay between the Weyl nodes and FM domain walls.
I. INTRODUCTION

Weyl nodes are protected linear crossings of two non-degenerate bands that lead to chiral relativistic quasiparticles [1, 2]. In Weyl semimetals (WSMs), the presence of Weyl nodes at the Fermi level enables Berry phase engineering in the bulk, creates Fermi arcs on the surface, and leads to a host of emergent electromagnetic responses such as topological Hall effect and anomalous Hall effect (AHE) [3–13]. There are two main routes to generating Weyl semimetals, either by breaking inversion symmetry [14], or time-reversal symmetry [15]. The former has been realized in the original discovery of non-magnetic Weyl semimetals in TaAs family [16, 17]. The latter approach has recently led to the discovery of a few ferromagnetic Weyl semimetals (FM-WSMs) such as Co$_3$Sn$_2$S$_2$, Fe$_3$GeTe$_2$, and Co$_2$MnGa [18–21]. These FM-WSMs crystallize in a centrosymmetric lattice and exhibit a collinear FM ordering. They have been intensely studied due to a giant AHE that results from the Berry curvature around Weyl nodes, as confirmed by first-principle calculations [5, 19–21].

In this article, we introduce CeAlSi as a new type of FM-WSM that combines both routes mentioned above to generate Weyl nodes. CeAlSi crystallizes in the noncentrosymmetric space group $I4_1md$ that stabilizes the Weyl nodes by breaking inversion symmetry, as we will confirm from second-harmonic-generation (SHG) experiments and first-principles calculations. In addition, the local $f$-moments of Ce$^{3+}$ interact within the noncentrosymmetric lattice and lead to a noncollinear FM order. The breaking of time reversal symmetry in CeAlSi shifts the nodal positions and controls the magnitude of the AHE. We observe two different AHE responses in this material by orienting the magnetic field along the easy- and hard-axes. The lack of inversion symmetry, the in-plane noncollinear FM order, and the novel anisotropic AHE makes CeAlSi a new FM-WSM candidate distinguished from its predecessors.

II. CENTRAL FINDINGS

Figure 1 summarizes our main results: the discovery of a new noncentrosymmetric FM-WSM with anisotropic AHE. The body-centered tetragonal unit cell of CeAlSi (Fig. 1A) has two vertical mirror planes ($\sigma_v$) but lacks a horizontal mirror plane ($\sigma_h$), thus breaking inversion symmetry. The viability of a FM-WSM in such a structure (space group $I4_1md$)
Non-collinear order, non-centrosymmetric structure, and anomalous Hall effect in CeAlSi. (A) Non-collinear FM order in the tetragonal unit cell of CeAlSi (space group $I\bar{4}1md$). (B) Second harmonic generation data refined in the noncentrosymmetric $C_{4v}$ point group. (C) Anisotropic magnetic susceptibility as a function of temperature showing in-plane easy-axis. (D) Magnetization curves with field along the [110], [100], and [001] directions. Inset shows hysteresis due to FM domains with a coercive field of 70 Oe. (E) Two distinct Hall responses are observed when a magnetic field is applied along the easy-axis ($\sigma_{yz}^A$: anomalous Hall effect) and hard-axis ($\sigma_{xy}^L$: loop Hall effect). (F) Evolution of the magnetic ordering parameter, $\sigma_{yz}^A$, and $\sigma_{xy}^L$ with temperature.

was first proposed by DFT calculations in CeAlGe [22, 23]; however, experimental data revealed an antiferromagnetic (AF) order in that material, instead of an FM one [24, 26]. From neutron diffraction and magnetization measurements, we found that CeAlSi has an FM order with net magnetization along the crystallographic [110] direction and an in-plane non-collinear spin texture as illustrated in Fig. 1A.

An important structural detail is the possibility of site mixing between Al and Si, which could nullify the proposal of CeAlSi being a noncentrosymmetric FM-WSM. Inter-site mixing can restore the $\sigma_h$ mirror plane and change the space (point) group from noncentrosymmetric...
$I4_{1}md$ ($C_{4v}$) to centrosymmetric $I4_{1}/amd$ ($C_{4h}$). Neither X-ray nor neutron diffraction can reliably distinguish between the two space groups (see Sec. M1 in the Supplemental Material). However, SHG discriminates between them, because the SHG signal predominantly originates from a bulk electric dipole in a noncentrosymmetric unit cell. Figure 1B shows a strong SHG signal ($\chi_{xxx} = 200$ pm/V) that is commensurate with the pronounced signal in GaAs [27] and fits to the point group $C_{4v}$. Thus, we confirm the noncentrosymmetric space group $I4_{1}md$ as the correct structure (details in Sec. M2).

CeAlSi is ferromagnetic and has a strong magnetic anisotropy with in-plane easy-axis. As seen in Fig. 1C, the anisotropy makes the in-plane magnetic susceptibility (blue) 200 times larger than the out-of-plane susceptibility (red). The field dependence of magnetization (Fig. 1D) specifies the [110] crystallographic direction as the easy-axis. A gradual saturation of the $M(H||[100])$ curve from $M_{sat}(H||[110])/\sqrt{2}$ to $M_{sat}(H||[110])$ implies zero-field magnetic domains with $M\parallel [110]$, [1-10], [-110], and [-1-10] directions.

Due to the in-plane easy-axis orientation, we expected to observe an AHE when the magnetic field was oriented in the $ab$-plane. Figure 1E confirms such anomalous Hall conductivity (the step in $\sigma_{yzz}^{A}$), but it also reveals an unexpected signal ($\sigma_{xy}^{L}$) which is observed when the field is along the hard-axis. The superscript $L$ in $\sigma_{xy}^{L}$ stands for its loop-shape behavior which will be explored in detail. Figure 1F shows the parallel temperature dependence of $\sigma_{yzz}^{A}$, $\sigma_{xy}^{L}$, and the magnetic order parameter determined by neutron diffraction, suggesting that both Hall responses are controled by the FM order. We examine these findings in detail in the rest of the paper.

III. IN-PLANE NONCOLLINEAR FM ORDER AND LARGE FM DOMAINS

The magnetic heat capacity ($C_{m}$) of CeAlSi is plotted in Fig. 2A with a sharp FM transition at $T_C = 8.2(3)$ K and a broad (Schottky) peak at 80 K due to the crystal electric field (CEF) splitting of Ce$^{3+}$ atomic levels. As shown in Fig. 2B, the $J = 5/2$ sextet of Ce$^{3+}$ splits into a doublet ground-state and a quadruplet excited-state, leading to two plateaus at $R \ln(2)$ and $R \ln(6)$ in the magnetic entropy $S_{m}$. From a fit to the $C_{m}$ data in Fig. 2A, we estimate a gap of $\Delta = 25$ meV between the doublet and quadruplet, and identify the ground-state of CeAlSi as a Kramers doublet with effective spin-1/2.

The in-plane noncollinear FM order of CeAlSi was determined by neutron diffraction.
Figure 2. **Magnetic structure.** (A) Magnetic specific heat as a function of temperature with a fit to the CEF levels. Inset shows a magnified view of the FM transition and a fit to the temperature dependence of the order parameter (magnetic moment per Ce atom). (B) Magnetic entropy as a function of temperature. Inset shows the CEF levels with a doublet ground-state. (C) Neutron Bragg peak at $Q = (004)$. (D) Neutron Bragg peak at $Q = (1-10)$ is observed below $T_C = 8.2(3)$ K in zero-field and suppressed by applying a field of 0.1 T in the [110] direction. (E) Scanning SQUID image of FM domains obtained at $T = 6$ K under zero field. (F) Large in-plane domains develop under a small in-plane field (16 Oe).

Figure 2C shows the $Q = (004)$ peak corresponding to the FM ordering vector $k = (000)$. The magnetic moment per Ce$^{3+}$ (order parameter) is extracted from the intensity of this peak and plotted as a function of temperature in the inset of Fig. 2A along with the low-$T$ heat capacity. These data are consistent with a second-order mean-field transition with the critical exponent $\beta = 0.48(4)$. Thus, the magnetic structure of CeAlSi belongs to a single irreducible representation (irrep) of the $I4_1md$ space group. A combination of a symmetry analysis (details in Sec. M3) and the observation of several (00L) peaks concludes that CeAlSi orders in the $\Gamma_5$ manifold, where all spins lie in the $ab$-plane (Fig. 2C).

As illustrated in the inset of Fig. 2C, the $\Gamma_5$ manifold allows for a complete decoupling
of the Ce spins between adjacent \((0,0, z+1/4)\) layers. We define \(S_1\) to be the Ce spins at \((0,0,0)\) and \(S_2\) to be those at \((0,1/2,1/4)\). The intensity for the neutron Bragg peaks with \(k = (000)\) and \((110)\) ordering vectors is proportional to \(S_1 + S_2\) and \(S_1 - S_2\), respectively. Thus, the observation of both ordering vectors in Figs. 2C,D suggests that both \(S_1 + S_2\) and \(S_1 - S_2\) are finite, and the angle between \(S_1\) and \(S_2\) defined by \(\theta = \cos^{-1}\left(\frac{S_1 \cdot S_2}{\|S_1\| \|S_2\|}\right)\) must be nonzero. Detailed refinement of the spin structure was then performed against 40 symmetrically distinct Bragg peaks collected at both 1.4 K and 10 K in zero-field (see Sec. M3 for details). Assuming \(\|S_1\| = \|S_2\|\), the refinement suggests a moment size of \(1.2(2) \mu_B\) and \(\theta = 70(30)^\circ\), confirming the in-plane noncollinear FM order in CeAlSi (inset of Fig. 2D).

We performed scanning SQUID microscopy \([28–30]\) to visualize the FM domain structure of CeAlSi. The images in Figs. 2E,F were obtained by scanning a SQUID sensor over the \(ab\)-surface of a polished crystal to measure the out-of-plane stray field from the in-plane domains. Whereas the domains are small in zero-field-cooling (Fig. 2E), a weak in-plane field of a few Gauss is enough to develop large in-plane FM domains that are hundreds of microns across (Fig. 2F). The development of large domains is also implied by the selection of a single domain revealed by neutron diffraction. Under a small in-plane field \(H||[110]\), the system selects a single domain with \(M||[110]\) among all symmetrically equivalent ones. As a result, the vector \(S_1 - S_2\) only points along [1-10] and the \(Q = (1-10)\) Bragg peak is suppressed accordingly, as evidenced in Fig. 1D when a field of 0.1 T is applied in the [110] direction. The magnitude of the observed DC flux is on the order of a few \(\Phi_0\), consistent with the remanent magnetization determined from the \(c\)-axis bulk magnetization measurements. According to our estimates based on the remanent \(a\)-axis magnetization, if the domains were to have flipped magnetization from the in-plane to the out-of-plane, it would have produced DC signal on the order of hundreds of \(\Phi_0\), which is clearly not the case in Figs. 2E,F (see Sec. M4 for details). The emerging picture from our neutron scattering and scanning SQUID measurements is a noncollinear in-plane FM order with large domains in CeAlSi.

### IV. BAND STRUCTURE, SHIFTED WEYL NODES, AND INTRINSIC ANOMALOUS HALL CONDUCTIVITY

The band structure of CeAlSi (Fig. 3A) consists of tiny hole and electron pockets with a nearly vanishing density of states (DOS) at \(E_F\) (Fig. 3B). The density of the majority
Figure 3. **Band structure and Weyl nodes.** (A) Band structure of CeAlSi (fully relativistic calculation including spin-orbit coupling). (B) Density of the majority (green) and minority (purple) spin states. (C) Electron (red) and hole (blue) pockets are shown in the left Brillouin zone (BZ), and the Weyl nodes are shown in the right BZ. (D) The effect of breaking inversion symmetry (left) and time-reversal symmetry (right) on the positions of Weyl nodes. (E) Theoretical values of the anomalous Hall conductivity with the field in plane ($\sigma_{xy}^A$) and out of plane ($\sigma_{yz}^A$). (F) The scaling behavior $\sigma_{yz}^A \propto \sigma_{xx}^2$ in CeAlSi, confirming an intrinsic AHE.

and minority spin states peak at different energies (Fig. 3B), leading to the FM ordering. The residual electron and hole pockets are illustrated in Fig. 3C. Also, Fig. 3C shows 12 pairs of Weyl nodes next to the $k_x = 0$ and $k_y = 0$ mirror-planes. We denote the 4 pairs of nodes located on the $k_z = 0$ plane as $W_1$, and the other 8 as $W_2$. Whereas the $W_1$ nodes are 80 – 120 meV away from $E_F$, the $W_2$ nodes are within 25 meV of the $E_F$ (details in Sec. M5). All $W_1$ and $W_2$ Weyl fermions have linear energy dispersions in all $k$-directions, suggesting a type-I WSM (Sec. M5). Note that the Weyl nodes in CeAlSi result from a broken inversion symmetry ($I$) and the effect of broken time-reversal symmetry ($T$) at $T < T_C$ is to shift the position of Weyl nodes in the Brillouin zone (Fig. 3D) [6, 22]. In this regard, CeAlSi is a new FM-WSM, in sharp contrast to the centrosymmetric systems such as Co$_3$Sn$_2$S$_2$ [18, 19], Fe$_3$GeTe$_2$ [20], and Heusler alloys [31] where the Weyl nodes result from broken $T$. 
We calculated the anomalous Hall conductivity (AHC) along the easy-axis ($\sigma_{yz}^A$) and hard-axis ($\sigma_{xy}^A$) as a function of Fermi energy in Fig. 3E [5]. The theoretical magnitude of the AHC along the easy-axis in Fig. 3E is comparable to the experimental values ($\sigma_{xy}^A = -\rho_{xy}^A/\rho_{xx}^2$; $\rho_{xy}^A = \rho_{xy} - R_0 H$) [13, 32] in Fig. 3F. The scaling behavior between $\sigma_{xy}^A$ and $\sigma_{xx}^2$ in Fig 3F is consistent with intrinsic and side-jump AHE [4]. Notice that according to DFT, we do not expect an AHC ($\sigma_{xy}^A = 0$) along the magnetic hard-axis $H \parallel c$. Therefore, the observation of a loop-shape Hall signal with $H \parallel c$ (as explained next) is a novel electromagnetic response.

V. ANISOTROPIC ANOMALOUS HALL EFFECT

CeAlSi displays two different anomalous Hall responses when measured under in-plane (easy-axis) and out-of-plane (hard-axis) magnetic fields. We start by examining the Hall resistivity $\rho_{yz}$ as a function of in-plane field $H \parallel a$ in Fig. 4A. We separate the conventional and anomalous Hall signals by fitting the data at $H > 1$ T to $\rho_{yz} = R_0 H + \rho_{yz}^A$. The conventional Hall effect ($R_0 H$) has a slope $R_0 = -3.9 \mu\Omega \cdot cm^{-1}$ corresponding to a small electron concentration $n_e = -1.6 \times 10^{20} \ cm^{-3}$ (0.0003 electrons per unit cell), consistent with the small DOS at $E_F$ in Fig. 3B. The anomalous Hall resistivity $\rho_{yz}^A$ is plotted as a function of field in Fig. 4B. The anomalous Hall conductivity is calculated from $\sigma_{yz}^A = -\rho_{yz}^A/\rho_{xx}^2$ and plotted in Fig. 1E and Fig. 3G. The magnitude of $\sigma_{yz}^A$ is in agreement with DFT results in Fig. 3E.

Next, we study the Hall resistivity $\rho_{xy}$ as a function of out-of-plane field $H \parallel c$ (hard-axis) in Fig. 4C, where an unusual loop is observed. The loop corresponds to different traces of $\rho_{xy}(H)$ between field sweeps in the positive and negative directions (see arrows in Fig. 4C). It extends over a region of $\pm 2$ T, two orders of magnitude larger than the magnetic coercive field (70 Oe, inset of Fig. 1D). Notice that the loop-shape Hall effect (LHE) does not scale with magnetization $M(H)$ and appears only when measured along the magnetic hard-axis, unlike the AHE that follows the magnetization curve ($\rho_{yz}^A = R_S M(H)$) and appears when the field is parallel to the easy-axis. To study the temperature dependence of the LHE, we subtract the positive field sweep from the negative one and plot the loop Hall resistivity as $\rho_{xy}^L = \rho_{xy}(3 \rightarrow -3 \ T) - \rho_{xy}(-3 \rightarrow 3 \ T)$ at several temperatures in Fig. 4D. The loop Hall conductivity $\sigma_{xy}^L$ in Fig. 1E,F was calculated as $\sigma_{xy}^L = \rho_{xy}^L/\rho_{xx}^2$ (see Sec. M6 for $\rho_{xx}$ data).

We establish a link between the LHE and Weyl nodes by measuring samples with different
Figure 4. **Magnetic anisotropy and the loop Hall effect.** (A) Hall resistivity $\rho_{yz}$ measured at several temperatures with the field along the magnetic easy-axis ($H\parallel a$). (B) Anomalous Hall resistivity $\rho_{yz}^A$ derived from $\rho_{yz}$. (C) Hall resistivity $\rho_{xy}$ measured at several temperatures with the field along the hard-axis ($H\parallel c$). (D) Loop Hall resistivity $\rho_{xy}^L$ derived from $\rho_{xy}$. (E) The normalized resistivity plotted as a function of temperature in samples S1-S6 with similar residual resistivity ratio. (F) Hall resistivity $\rho_{xy}$ of samples S1-S6 measured at $T = 1.8$ K. (G) Quantum oscillations (QO) in samples S2, S4, and S6, showing an evolution of the oscillation period. (H) Illustrated band structure of CeAlSi showing an electron band, a Weyl crossing, and the Fermi levels of samples S2, S4 and S6. The decreasing QO frequency observed in panel (G) originates from the electron pocket (left) and the shift of Fermi level among S2, S4 and S6.

We selected six samples (S1 to S6) with comparable residual resistivity ratios $RRR = R(300K)/R(2K)$ (Fig. 4E). A slight off-stoichiometry of Si and Al in these samples (Sec. S7) causes a shift of the Fermi level relative to the Weyl nodes [33]. The variation of $E_F$ between samples S1 to S6 is evident in Fig. 4F that shows three categories of Hall curves $\rho_{xy}(H)$ in CeAlSi samples: a linear $\rho_{xy}$ with negative slope in S1 and S2; a moderately nonlinear $\rho_{xy}$ with negative slope at all fields in S3 and S4; and, a strongly nonlinear $\rho_{xy}$ with positive slope at low fields and negative slope at high fields in S5 and S6. The slope of the Hall effect is related to the
sign of charge carriers. Therefore, it is conceivable that $E_F$ crosses only electron pockets in S1/S2, nearly crosses another hole pocket in S3/S4, and crosses both the electron and hole pockets in S5/S6 as illustrated in Fig. 4H. The LHE is observed only in S3 and S4 with $E_F$ near the crossing of electron and hole bands, i.e. near the Weyl node (Fig. 4H).

To confirm the scenario in Fig. 4H, we used Schubnikov-de Haas (SdH) oscillations to locate the $E_F$ with respect to the Weyl nodes by following prior studies in Weyl and magnetic semimetals [34, 35]. Figure 4G shows quantum oscillations between magnetic field 15 and 33 T in samples S2, S4, and S6. According to the Onsager relation, $F = A \left( \frac{h}{2\pi e} \right)$, the frequency of SdH oscillations $F$ is proportional to the extremal orbit area $A$, and both $A$ and $F$ can be calculated from the DFT-obtained band structure. As we shift the $E_F$ in the band structure, the theoretical $F$ given by DFT will change, and the $E_F$ for each sample is pinned down once the theoretical $F$ of the electron pocket (the left object in Fig. 4H) matches the experimental $F$ observed in Fig. 4G. As a result of our analysis, the $E_F$ for samples S2, S4, and S6 are determined to be 32, 23, and 12 meV above the DFT-obtained $E_F$ (see Sec. M8 for details). Having fixed the correct $E_F$ from the SdH oscillations, we found that all Weyl nodes are away from the $E_F$ of samples S2 and S6, but a set of Weyl nodes $W_2$ is located within 1 meV from the $E_F$ of sample S4, as illustrated in Fig. 4H (see also Table M2 and M4). Thus, we conclude that the LHE is observed only in samples with $E_F$ nearly crossing the Weyl nodes.

VI. OUTLOOK

In summary, CeAlSi is a unique noncentrosymmetric FM-WSM with an in-plane non-collinear FM order and novel anisotropic anomalous Hall responses along the easy- and hard-axes. Specifically, the LHE is anomalous in a sense that it appears when the applied field is along the hard-axis, it does not scale with either field or magnetization, and it is a pure outcome of having the Weyl nodes nearly at the Fermi level. A similar loop-shape Hall signal was observed in Nd$_2$Ir$_2$O$_7$ and attributed to the Fermi arcs projected from the Weyl nodes on the Domain Walls [36, 39]. The magnetic ordering in both Nd$_2$Ir$_2$O$_7$ and CeAlSi is noncollinear with both AF and FM components. However, Nd$_2$Ir$_2$O$_7$ is an overall AF system and does not exhibit AHE; it only shows the LHE. But CeAlSi has an overall FM order and exhibits both the AHE (along the easy-axis) and LHE (along the hard-axis).
The large FM domains in CeAlSi are specifically suitable for device engineering and tuning the Fermi arcs by straining the domain walls [40]. An exciting future research direction is to explore exotic electromagnetic responses from the Fermi arcs projected on the domain walls instead of the trivial sample surfaces. CeAlSi would be a promising material for such explorations due to its large FM domain and the proximity of the Weyl nodes to the Fermi level.

ACKNOWLEDGMENTS

We thank Chunli Huang, Hiroaki Ishizuka, Bohm-Jung Yang and Ying Ran for helpful discussions. F.T. acknowledges funding by the National Science Foundation under Award No. NSF/DMR-1708929. The work at Northeastern University was supported by the US Department of Energy (DOE), Office of Science, Basic Energy Sciences Grant No. DE-SC0019275, and benefited from Northeastern University’s Advanced Scientific Computation Center and the National Energy Research Scientific Computing Center through DOE Grant No. DE-AC02-05CH11231. Neutron scattering was supported as part of the Institute for Quantum Matter, an Energy Frontier Research Center funded by the U.S. Department of Energy, Office of Science, Basic Energy Sciences under Award No. de-sc0019331. A portion of this research used resources at the High Flux Isotope Reactor and Spallation Neutron Source, a DOE Office of Science User Facility operated by the Oak Ridge National Laboratory. The National High Magnetic Field Laboratory is supported by National Science Foundation through NSF/DMR-1644779 and the State of Florida. The work by I.S. was in part supported by the US Department of Defense, and the US State of Connecticut. B.X. and J.F. were supported through graduate assistantship provided by the University of Connecticut’s College for Liberal Arts and Sciences. We acknowledge the support of the National Institute of Standards and Technology, U.S. Department of Commerce. Certain commercial equipment, instruments, or materials (or suppliers, or software, etc.) are identified in this Letter to foster understanding. Such identification does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

The authors declare no competing financial interests.

H.-Y. Y. and B. S. contributed equally to this work. H.-Y. Y. grew the crystals, performed
magnetization and transport experiments. H.-Y. Y. and D. E. G. performed high-field experiments. B. S. performed first-principles calculations and theoretical analysis with assistance and guidance from C.-Y. H., W.-C. C., S.-M. H., B. W., H. L., and A. B. J. G. and C. L. B. performed neutron scattering, B. L. and D. H. T. performed SHG experiments, F. B. analyzed X-ray data. I. S., B. X., and J. F. performed the scanning SQUID microscopy. F. T. and A. B. conceived the research. All authors discussed the results and contributed to writing the manuscript.

Appendix: Methods

Crystal Growth.

The CeAlSi single crystals were grown by a self-flux method in both regular alumina crucibles and the Canfield crucible sets \([41]\). Both methods produced a similar crystal quality based on the PXRD, SHG, EDX and resistivity measurements. In both methods, the starting materials were weighed in a ratio Ce:Al:Si = 1:10:1, placed inside a crucible in an evacuated quartz tube, heated to 1000°C at 3°C/min, stayed at 1000°C for 12 h, cooled to 700°C at 0.1°C/min, stayed at 700°C for 12 h, and centrifuged to decant the residual Al flux.

Band Structure.

Density functional theory (DFT) calculations were performed using the experimental lattice parameters \((a = 4.252 \text{ Å}; c = 14.5801 \text{ Å})\) and a projector augmented wave (PAW) method as implemented in the Vienna ab-initio simulation package (VASP) \([42]\). The exchange-correlations were included using a generalized gradient approximation (GGA), and the spin-orbit coupling (SOC) was included self-consistently \([43,44]\). The on-site Coulomb interaction was added for Ce \(f\)-electrons within the GGA+U scheme with \(U_{\text{eff}} = 6\) eV. A Wannier tight-binding Hamiltonian was obtained from the ab-initio results, using the VASP2WANNIER90 interface, which was subsequently used in our topological properties calculations \([45]\).
Transport, Heat capacity, and Magnetization Measurements.

Electrical resistivity was measured with the standard four-probe technique and the heat capacity was performed with the relaxation time method in a Quantum Design Physical Property Measurement System (PPMS) Dynacool. The magnetic heat capacity $C_m$ is derived by first measuring the heat capacity of the non-magnetic LaAlSi, and then subtracting it from the heat capacity of CeAlSi. DC magnetization experiments were conducted on the vibrating sample magnetometer in a Quantum Design MPMS3. The high-field experiments were performed using a 35 T DC Bitter magnet and a $^3$He fridge with base temperature of 300 mK at the MagLab in Tallahassee. The comparison of the quantum oscillation frequencies between theory and experiment was done using the DFT-generated bxsf file and the program SKEAF.

Neutron Diffraction.

The nuclear structure of CeAlSi was characterized by a single-crystal time-of-flight experiment at 100 K on TOPAZ at the Oak Ridge National Lab. A 3D diffraction map was acquired from 14 different sample positions allowing measurements of 6946 Bragg peaks where the nuclear structure factors were extracted following the method of Schultz et al. The structural refinements were performed using GSAS-II. The magnetic structure was determined by diffraction experiments at the NIST Center for Neutron Research. The magnetic structure factors were determined using the thermal triple-axis spectrometer BT-7 by collecting rocking scans at various Bragg positions with an incident and scattered neutrons energies of 14.7 meV. Two single crystals were inserted in a top-loading CCR and into a 7 T magnet to measure Bragg peaks in both the (H0L) and (HHL) planes. The order parameter measurement in Fig. 1F was performed with the SPINS spectrometer using 3.7 meV incident and scattered neutrons.

Second Harmonic Generation.

The SHG data in Fig. 1B were taken at normal incidence on the [101] face of as-grown crystals for incoming/outgoing wavelengths 1500/750 nm as a function of the incoming field polarization and measured for emitted light polarized parallel to [010] crystalline axis.
In this geometry, all bulk contributions to SHG from a $I4_1/amd$ space group are forbidden.

**Scanning SQUID Imaging.**

We used scanning SQUID susceptometers with two gradiometric field coils and pick up loops [28]. The SQUID pick-up loop and the field coil average radii were 3.25 and 7 $\mu$m formed from Nb lines of 0.5 and 1 $\mu$m width, respectively. The scanning SQUID apparatus was housed in a closed-cycle Montana Instruments Fusion cryostat (Bozeman, Montana, USA) with the cryostat base temperature of 3 K.

---

[1] N. Armitage, E. Mele, and A. Vishwanath, Weyl and Dirac semimetals in three-dimensional solids, *Reviews of Modern Physics* **90**, 015001 (2018), publisher: American Physical Society.

[2] A. Bansil, H. Lin, and T. Das, Colloquium: Topological band theory, *Reviews of Modern Physics* **88**, 021004 (2016), publisher: American Physical Society.

[3] Y. Tokura, M. Kawasaki, and N. Nagaosa, Emergent functions of quantum materials, *Nature Physics* **13**, 1056 (2017).

[4] N. Nagaosa, J. Sinova, S. Onoda, A. H. MacDonald, and N. P. Ong, Anomalous Hall effect, *Reviews of Modern Physics* **82**, 1539 (2010), publisher: American Physical Society.

[5] Y. Yao, L. Kleinman, A. H. MacDonald, J. Sinova, T. Jungwirth, D.-s. Wang, E. Wang, and Q. Niu, First Principles Calculation of Anomalous Hall Conductivity in Ferromagnetic bcc Fe, *Physical Review Letters* **92**, 037204 (2004), publisher: American Physical Society.

[6] K.-Y. Yang, Y.-M. Lu, and Y. Ran, Quantum Hall effects in a Weyl semimetal: Possible application in pyrochlore iridates, *Physical Review B* **84**, 075129 (2011).

[7] A. Burkov, Anomalous Hall effect in Weyl metals, Physical review letters **113**, 187202 (2014).

[8] K. Ueda, R. Kaneko, H. Ishizuka, J. Fujioka, N. Nagaosa, and Y. Tokura, Spontaneous hall effect in the weyl semimetal candidate of all-in all-out pyrochlore iridate, Nature communications **9**, 1 (2018).

[9] S. Nakatsuji, N. Kiyohara, and T. Higo, Large anomalous Hall effect in a non-collinear antiferromagnet at room temperature, *Nature* **534**, S5 (2016), number: 7607 Publisher: Nature Publishing Group.
A. K. Nayak, J. E. Fischer, Y. Sun, B. Yan, J. Karel, A. C. Komarek, C. Shekhar, N. Kumar, W. Schnelle, J. Kübler, C. Felser, and S. S. P. Parkin, Large anomalous Hall effect driven by a nonvanishing Berry curvature in the noncolinear antiferromagnet Mn₃Ge, *Science Advances* **2**, e1501870 (2016).

X. Li, C. Collignon, L. Xu, H. Zuo, A. Cavanna, U. Gennser, D. Mailly, B. Fauqué, L. Balents, Z. Zhu, and K. Behnia, Chiral domain walls of Mn₃Sn and their memory, *Nature Communications* **10**, 1 (2019), number: 1 Publisher: Nature Publishing Group.

D. Destraz, L. Das, S. S. Tsirkin, Y. Xu, T. Neupert, J. Chang, A. Schilling, A. G. Grushin, J. Kohlbrecher, L. Keller, P. Puphal, E. Pomjakushina, and J. S. White, Magnetism and anomalous transport in the Weyl semimetal PrAlGe: possible route to axial gauge fields, *npj Quantum Materials* **5**, 1 (2020), number: 1 Publisher: Nature Publishing Group.

H.-Y. Yang, B. Singh, B. Lu, C.-Y. Huang, F. Bahrami, W.-C. Chiu, D. Graf, S.-M. Huang, B. Wang, H. Lin, D. Torchinsky, A. Bansil, and F. Tafti, Transition from intrinsic to extrinsic anomalous Hall effect in the ferromagnetic Weyl semimetal PrAlGe₁₋ₓSiₓ, *APL Materials* **8**, 011111 (2020), publisher: American Institute of Physics.

H. Weng, C. Fang, Z. Fang, B. A. Bernevig, and X. Dai, Weyl Semimetal Phase in Noncentrosymmetric Transition-Metal Monophosphides, *Physical Review X* **5**, 011029 (2015).

X. Wan, A. M. Turner, A. Vishwanath, and S. Y. Savrasov, Topological semimetal and Fermi-arc surface states in the electronic structure of pyrochlore iridates, *Physical Review B* **83**, 205101 (2011), publisher: American Physical Society.

B. Lv, H. Weng, B. Fu, X. Wang, H. Miao, J. Ma, P. Richard, X. Huang, L. Zhao, G. Chen, Z. Fang, X. Dai, T. Qian, and H. Ding, Experimental Discovery of Weyl Semimetal TaAs, *Physical Review X* **5**, 031013 (2015), publisher: American Physical Society.

S.-Y. Xu, I. Belopolski, N. Alidoust, M. Neupane, G. Bian, C. Zhang, R. Sankar, G. Chang, Z. Yuan, C.-C. Lee, S.-M. Huang, H. Zheng, J. Ma, D. S. Sanchez, B. Wang, A. Bansil, F. Chou, P. P. Shibayev, H. Lin, S. Jia, and M. Z. Hasan, Discovery of a Weyl fermion semimetal and topological Fermi arcs, *Science* **349**, 613 (2015), publisher: American Association for the Advancement of Science Section: Research Article.

Q. Wang, Y. Xu, R. Lou, Z. Liu, M. Li, Y. Huang, D. Shen, H. Weng, S. Wang, and H. Lei, Large intrinsic anomalous Hall effect in half-metallic ferromagnet Co₃Sn₂S₂ with magnetic Weyl fermions, *Nature Communications* **9**, 3681 (2018).
[19] E. Liu, Y. Sun, N. Kumar, L. Muechler, A. Sun, L. Jiao, S.-Y. Yang, D. Liu, A. Liang, Q. Xu, J. Kroder, V. Süß, H. Borrmann, C. Shekhar, Z. Wang, C. Xi, W. Wang, W. Schnelle, S. Wirth, Y. Chen, S. T. B. Goennenwein, and C. Felser, Giant anomalous Hall effect in a ferromagnetic kagome-lattice semimetal, *Nature Physics* **14**, 1125 (2018).

[20] K. Kim, J. Seo, E. Lee, K.-T. Ko, B. S. Kim, B. G. Jang, J. M. Ok, J. Lee, Y. J. Jo, W. Kang, J. H. Shim, C. Kim, H. W. Yeom, B. I. Min, B.-J. Yang, and J. S. Kim, Large anomalous Hall current induced by topological nodal lines in a ferromagnetic van der Waals semimetal, *Nature Materials* **17**, 794 (2018).

[21] I. Belopolski, K. Manna, D. S. Sanchez, G. Chang, B. Ernst, J. Yin, S. S. Zhang, T. Cochran, N. Shumiya, H. Zheng, B. Singh, G. Bian, D. Multer, M. Litskevich, X. Zhou, S.-M. Huang, B. Wang, T.-R. Chang, S.-Y. Xu, A. Bansil, C. Felser, H. Lin, and M. Z. Hasan, Discovery of topological Weyl fermion lines and drumhead surface states in a room temperature magnet, *Science* **365**, 1278 (2019), publisher: American Association for the Advancement of Science Section: Report.

[22] G. Chang, B. Singh, S.-Y. Xu, G. Bian, S.-M. Huang, C.-H. Hsu, I. Belopolski, N. Alidoust, D. S. Sanchez, H. Zheng, H. Lu, X. Zhang, Y. Bian, T.-R. Chang, H.-T. Jeng, A. Bansil, H. Hsu, S. Jia, T. Neupert, H. Lin, and M. Z. Hasan, Magnetic and noncentrosymmetric Weyl fermion semimetals in the RAlGe family of compounds (R = rare-earth), *Physical Review B* **97**, 041104 (2018).

[23] S.-Y. Xu, N. Alidoust, G. Chang, H. Lu, B. Singh, I. Belopolski, D. S. Sanchez, X. Zhang, G. Bian, H. Zheng, M.-A. Husanu, Y. Bian, S.-M. Huang, C.-H. Hsu, T.-R. Chang, H.-T. Jeng, A. Bansil, T. Neupert, V. N. Strocov, H. Lin, S. Jia, and M. Z. Hasan, Discovery of Lorentz-violating type II Weyl fermions in LaAlGe, *Science Advances* **3**, e1603266 (2017).

[24] H. Hodovanets, C. J. Eckberg, P. Y. Zavalij, H. Kim, W.-C. Lin, M. Zic, D. J. Campbell, J. S. Higgins, and J. Paglione, Single-crystal investigation of the proposed type-II Weyl semimetal CeAlGe, *Physical Review B* **98**, 245132 (2018).

[25] P. Puphal, V. Pomjakushin, N. Kanazawa, V. Ukleev, D. J. Gawryluk, J. Ma, M. Naamneh, N. C. Plumb, L. Keller, R. Cubitt, E. Pomjakushina, and J. S. White, Topological Magnetic Phase in the Candidate Weyl Semimetal CeAlGe, *Physical Review Letters* **124**, 017202 (2020), publisher: American Physical Society.
[26] T. Suzuki, L. Savary, J.-P. Liu, J. W. Lynn, L. Balents, and J. G. Checkelsky, Singular angular magnetoresistance in a magnetic nodal semimetal, *Science* **365**, 377 (2019).

[27] S. Bergfeld and W. Daum, Second-Harmonic Generation in GaAs: Experiment versus Theoretical Predictions of $\chi^{(2)}_{xyz}$, *Physical Review Letters* **90**, 036801 (2003).

[28] I. Sochnikov, L. Maier, C. A. Watson, J. R. Kirtley, C. Gould, G. Tkachov, E. M. Hankiewicz, C. Brüne, H. Buhmann, L. W. Molenkamp, and K. A. Moler, Nonsinusoidal Current-Phase Relationship in Josephson Junctions from the 3D Topological Insulator HgTe, *Physical Review Letters* **114**, 066801 (2015), publisher: American Physical Society.

[29] I. Sochnikov, A. J. Bestwick, J. R. Williams, T. M. Lippman, I. R. Fisher, D. Goldhaber-Gordon, J. R. Kirtley, and K. A. Moler, Direct Measurement of Current-Phase Relations in Superconductor/Topological Insulator/Superconductor Junctions, *Nano Letters* **13**, 3086 (2013), publisher: American Chemical Society.

[30] B. W. Gardner, J. C. Wynn, P. G. Björnsson, E. W. J. Straver, K. A. Moler, J. R. Kirtley, and M. B. Ketchen, Scanning superconducting quantum interference device susceptometry, *Review of Scientific Instruments* **72**, 2361 (2001), publisher: American Institute of Physics.

[31] Z. Wang, M. Vergniory, S. Kushwaha, M. Hirschberger, E. Chulkov, A. Ernst, N. Ong, R. J. Cava, and B. A. Bernevig, Time-Reversal-Breaking Weyl Fermions in Magnetic Heusler Alloys, *Physical Review Letters* **117**, 236401 (2016).

[32] Y. Tian, L. Ye, and X. Jin, Proper Scaling of the Anomalous Hall Effect, *Physical Review Letters* **103**, 087206 (2009).

[33] C. Y. Guo, F. Wu, Z. Z. Wu, M. Smidman, C. Cao, A. Bostwick, C. Jozwiak, E. Rotenberg, Y. Liu, F. Steglich, and H. Q. Yuan, Evidence for Weyl fermions in a canonical heavy-fermion semimetal YbPtBi, *Nature Communications* **9**, 4622 (2018).

[34] R. Schönemann, N. Aryal, Q. Zhou, Y.-C. Chiu, K.-W. Chen, T. J. Martin, G. T. McCandless, J. Y. Chan, E. Manousakis, and L. Balicas, Fermi surface of the Weyl type-II metallic candidate WP$_2$, *Physical Review B* **96**, 121108 (2017).

[35] H.-Y. Yang, J. Gaudet, A. A. Aczel, D. E. Graf, P. Blaha, B. D. Gaulin, and F. Tafti, Interplay of magnetism and transport in HoBi, *Physical Review B* **98**, 045136 (2018), publisher: American Physical Society.

[36] S. M. Disseler, S. R. Giblin, C. Dhital, K. C. Lukas, S. D. Wilson, and M. J. Graf, Magnetization and Hall effect studies on the pyrochlore iridate Nd$_2$Ir$_2$O$_7$, *Physical Review B* **87**, 060403
[37] K. Ueda, J. Fujioka, Y. Takahashi, T. Suzuki, S. Ishiwata, Y. Taguchi, M. Kawasaki, and Y. Tokura, Anomalous domain-wall conductance in pyrochlore-type Nd$_2$Ir$_2$O$_7$ on the verge of the metal-insulator transition, *Physical Review B* **89**, 075127 (2014), publisher: American Physical Society.

[38] Y. Yamaji and M. Imada, Metallic Interface Emerging at Magnetic Domain Wall of Antiferromagnetic Insulator: Fate of Extinct Weyl Electrons, *Physical Review X* **4**, 021035 (2014), publisher: American Physical Society.

[39] E. Y. Ma, Y.-T. Cui, K. Ueda, S. Tang, K. Chen, N. Tamura, P. M. Wu, J. Fujioka, Y. Tokura, and Z.-X. Shen, Mobile metallic domain walls in an all-in-all-out magnetic insulator, *Science* **350**, 538 (2015).

[40] R. Ilan, A. G. Grushin, and D. I. Pikulin, Pseudo-electromagnetic fields in 3D topological semimetals, *Nature Reviews Physics* **2**, 29 (2020), number: 1 Publisher: Nature Publishing Group.

[41] P. C. Canfield, T. Kong, U. S. Kaluarachchi, and N. H. Jo, Use of frit-disc crucibles for routine and exploratory solution growth of single crystalline samples, *Philosophical Magazine* **96**, 84 (2016).

[42] G. Kresse and J. Furthmüller, Efficient iterative schemes for ab initio total-energy calculations using a plane-wave basis set, *Physical Review B* **54**, 11169 (1996).

[43] G. Kresse and D. Joubert, From ultrasoft pseudopotentials to the projector augmented-wave method, *Physical Review B* **59**, 1758 (1999).

[44] J. P. Perdew, K. Burke, and M. Ernzerhof, Generalized Gradient Approximation Made Simple, *Physical Review Letters* **77**, 3865 (1996).

[45] N. Marzari and D. Vanderbilt, Maximally localized generalized Wannier functions for composite energy bands, *Physical Review B* **56**, 12847 (1997).

[46] S. Julian, Numerical extraction of de Haas–van Alphen frequencies from calculated band energies, *Computer Physics Communications* **183**, 324 (2012).

[47] A. J. Schultz, M. R. V. Jørgensen, X. Wang, R. L. Mikkelsen, D. J. Mikkelsen, V. E. Lynch, P. F. Peterson, M. L. Green, and C. M. Hoffmann, Integration of neutron time-of-flight single-crystal Bragg peaks in reciprocal space, *Journal of Applied Crystallography* **47**, 915 (2014), number: 3 Publisher: International Union of Crystallography.
[48] B. H. Toby and R. B. Von Dreele, GSAS-II: the genesis of a modern open-source all purpose crystallography software package, Journal of Applied Crystallography 46, 544 (2013) number: 2 Publisher: International Union of Crystallography.

[49] B. Lu, J. D. Tran, and D. H. Torchinsky, Fast reflective optic-based rotational anisotropy non-linear harmonic generation spectrometer, Review of Scientific Instruments 90, 053102 (2019).