Field-dependent Shubnikov-de Haas oscillations in ferromagnetic Weyl semimetal $\text{Co}_3\text{Sn}_2\text{S}_2$

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We report a study of Shubnikov-de Haas oscillations in high quality single crystals of ferromagnetic Weyl semimetal $\text{Co}_3\text{Sn}_2\text{S}_2$. The Fermi surfaces resolved in our experiments are three-dimensional and reflect an underlying trigonal crystallographic symmetry. Combined with density functional theoretical calculations, we identify that the majority of the Fermi surfaces in the system – of both electron and hole nature – arise from the strong energy dispersion of the (spin-orbit gapped) mirror-protected nodal rings. We observe that an in-plane magnetic field induces a continuous evolution of Fermi surfaces, in contrast to field perpendicular to the kagome lattice planes which has little effect. Viewed alongside the easy-axis anisotropy of the system, our observation reveals an evolution of the electronic structure of $\text{Co}_3\text{Sn}_2\text{S}_2$ – including the Weyl points – with the ferromagnetic moment orientation. Through the case study of $\text{Co}_3\text{Sn}_2\text{S}_2$, our results provide concrete experimental evidence of an anisotropic interplay via spin-orbit coupling between the magnetic degrees of freedom and electronic band singularities, which has long been expected in semimetallic and metallic magnetic topological systems.

Topological semimetals (TSM) refer to a class of bulk gapless topological phases that host singular electronic excitations in three-dimensional momentum space [1]. A particularly versatile subclass of TSMs are those who in the meantime possess magnetic order [2–3]; in such magnetic TSMs, coexisting band topology and magnetic order parameters in principle allows the manipulation of the former using the latter. Early materials proposals of magnetic TSMs have built heavily on insights gained from the non-magnetic sector – pioneering ideas include modulation-doped topological insulating hetero-layers [4] and stoichiometric magnetic materials whose non-magnetic analogues are known as host to topologically non-trivial bands [5–6]. In those systems band topology and magnetism are typically carried by separate orbitals from distinct elements: itinerant $p$ orbitals often form topological bands while localized $d$ and $f$ electrons support magnetism [5–7]. A particularly exciting recent advancement is the discovery of a class of systems where the correlated 3$d$-electrons play the dual role of driving magnetic orders and composing topological bands [8–13]. This not only enables studies of topological responses of magnetic TSMs beyond room temperature [8–11], but also in principle dictates an intrinsic coupling between magnetic order and electronic topology.

In this Letter, via a combined Shubnikov-de Haas (SdH) oscillation and density functional theory (DFT) study, we explore the intertwined ferromagnetic order and the Weyl semimetallic phase in $\text{Co}_3\text{Sn}_2\text{S}_2$. The emergence of Weyl fermions in $\text{Co}_3\text{Sn}_2\text{S}_2$ [12–15] can be viewed in a similar manner with inversion-symmetry-breaking non-magnetic Weyl semimetals [16–18] where an inverted pair of bands first cross and generate nodal rings; then the spin-orbit coupling (SOC) in the presence of ferromagnetism gaps out the ring except at isolated points, the Weyl nodes. Here via high field ferrommetry studies on high quality single crystals, we experimentally demonstrate the evolution of the electronic structure with the orientation of the ferromagnetic moments in the system. With DFT we show that the observed evolution manifests an intrinsic connection between magnetism and topology: a strong dependence of the SOC-induced gap along the nodal ring with the magnetic moment orientation.
Quantum oscillations and Fermi surfaces of Co$_3$Sn$_2$S$_2$

Co$_3$Sn$_2$S$_2$ crystallizes in a Shandite Ni$_3$Pb$_2$S$_2$-type structure (Fig. 1(a)); the Co atoms form kagome lattices that are A-B-C stacked along the c-axis. Co$_3$Sn kagome layers spaced by additional Sn and S atoms can be identified in element-resolved Transmission Electron Microscopy images of our single crystals in Fig. 1(b,c). We show in Fig. 1(d) the resistivity $\rho$ of a typical CVT crystal as a function of temperature $T$. The residual resistivity ratio (RRR) defined as $\rho(T = 300 K)/\rho(T = 2 K)$ shows an enhanced value 235 and a low residual resistivity 3.5 $\mu$Ω cm compared to previous reports [12][19]. The magnetoresistance at $T = 1.6 K$ is shown in Fig. 1(d) inset where Shubnikov-de Haas oscillations are resolved from 5 T (see Supplementary Materials [20]). Together with the Hall traces, using a two band model we can fit $\rho_{xx}$ and $\rho_{yx}$ simultaneously with $n_1 = 9.5 \times 10^{19}/cm^3, \mu_1 = 6.3 \times 10^4 cm^2/V \cdot s, n_2 = -8.1 \times 10^{19}/cm^3, \mu_2 = 7.3 \times 10^4 cm^2/V \cdot s$ [20]. The high electronic quality in our CVT crystals allows us to experimentally study of the Fermi surfaces of the system via magneto-quantum oscillations.

In Fig. 1(f) we show the magnetoresistance traces measured at the NHMFL 45 T Hybrid magnet. The magnetic field $H$ is rotated from the trigonal to either the binary or the bisectrix axis, defining respectively $\theta_1$ and $\theta_2$ rotations schematically illustrated in Fig. 1(e). At $T = 0.4 K$, we show the evolution of resistance with $H$ at selected $\theta_2$ in Fig. 2(f), where prominent Shubnikov-de Haas (SdH) oscillations can be identified. When $H$ is applied close to the kagome planes (see e.g. the trace with $\theta_2 = 83^o$ in Fig. 2(f)), we can identify a kink in the magnetoresistance near 20 T, which we assign to a critical field beyond which the alignment of the magnetic moments with $H$ in the hard plane is complete (this is comparable with that extrapolated from the low field magnetization [21]). We return to the consequences of the reorientation process of the ferromagnetic moments below. We show post-background-subtraction SdH oscillations for $\theta_1$ and $\theta_2$ rotations in Figs. 2(g) and (h), respectively. That we identify SdH oscillations at all angle suggests a three-dimensional nature of the underlying Fermi surfaces in the system.

In Fig. 2(a) we summarize the frequencies obtained from a Fast Fourier Transformation (FFT) of the SdH oscillations between 11.5 - 45 T (FFT spectra themselves are shown in [20]) with the orientation of $H$. We show $\theta_1$ ($\theta_2$) between $0^o - 90^o$ ($0^o - 180^o$) because the oscillation frequencies appear symmetric (asymmetric) with respect to $90^o$ for $\theta_1$ ($\theta_2$) [20], consistent with the trigonal sym-
FIG. 2. (a) Experimental quantum oscillation frequencies as a function of the orientation of applied field $H$. (b) Comparison between the theoretical quantum oscillation spectra derived from DFT (colored lines) and experimental points (gray circles). (c,d) DFT Fermi surfaces of $\text{Co}_3\text{Sn}_2\text{S}_2$ $\alpha, \beta, \gamma, \delta$ and $\epsilon$ shown along with extremal cyclotron orbits for an out-of-plane magnetic field (c) and in the BZ (d). For clarity we show in (d) a subset of symmetry-related Fermi pockets. In (c,d) the hole Fermi surfaces ($\alpha, \beta$) are shown in red and electron pockets ($\gamma, \delta, \epsilon$) in blue. (e,f) Energy dispersion at the reflection-symmetry plane $k_y = 0$ for a $\mathbf{k} \cdot \mathbf{p}$ model obeying the material symmetries for zero and finite SOC, respectively. (g) Energy dispersion of $\text{Co}_3\text{Sn}_2\text{S}_2$ along the nodal ring; blue and orange lines are obtained without and with SOC, respectively. 

The Fermi surfaces giving rise to oscillations can be attributed to the proximity of van Hove singularity and thus a Lifshitz transition; as such Fermi surface topology of $\gamma$ can be captured by $\text{Co}_3\text{Sn}_2\text{S}_2$ along the nodal ring with respect to the horizontal dashed line starting from $L$ as illustrated in (d). We also mark the Fermi points corresponding to $\gamma, \beta$ and $\alpha$ pockets in (g).
The orientation of the magnetization ($\mathbf{M}$) is a crucial factor in determining the electronic structure in Co$_3$Sn$_2$S$_2$. A majority of Fermi surfaces in the system are determined by the orientation of $\mathbf{M}$ in its immediate neighborhood, with a transition to the framework of the magnetic anisotropy and the Zeeman effect. The complexity of the real Fermi surface originates mainly in the energy dispersion (~200 meV) of the nodal ring, which deforms the Weyl bands from the ideal case, yielding a rich energetic landscape (Fig. 2(g)). In addition to $\beta$ that encloses the Weyl points, $\alpha$ and $\gamma$ can also be attributed to the nodal ring energy dispersion. Additional electron pockets can be found near both $\Gamma$ ($\epsilon$ node) and $\alpha$ ($\gamma$ node) as well as for without SOC (gray line).

**Fermi surface changes due to rotation of the magnetization.** The orientation of the magnetization ($M$) has been shown to tune the electronic topology in various ferromagnets [21][24][28], including the Weyl-node energy in the case of Co$_3$Sn$_2$S$_2$ [21]. Below we show that the observed SdH oscillations (Fig. 1(g,h)) allow us to examine electronic structure reconstructions associated with a rotation of $M$. To illustrate the experimental window available in our setup, we consider a model consisting of an uniaxial magnetic anisotropy term and a Zeeman energy: $E = DM^2 \sin^2 \phi - MH \cos(\theta - \phi)$. Here $\theta$ ($\phi$) is the angle of $H$ ($M$) from the $c$-axis (Fig. 3(a)) and we take $D$ such that the field required to fully align $M$ with $H$ is 20T. As Fig. 3(b) shows, when $H$ is applied in the kagome plane, we expect a gradual and then accelerated rotation of $M$ into the plane with increasing $H$.

In Fig. 3(c) we show the FFT spectra between 11.5T - 20T and 20 - 45T, where $M$ is expected to be continuously rotating into the $ab$ plane within the former field range, and subsequently pinned along $H$ within the latter. A strong modification of the FFT spectra with the corresponding field range can be identified in Fig. 3(c). In contrast, performing a similar analysis for $H \parallel c$ reveals an FFT spectra with peak frequencies invariant with the range of $H$ (Fig. 3(d)), consistent with $M$ in the latter case always pointing along the $c$-axis in quantizing fields (Fig. 3(b)) (thus no electronic structure reconstruction is expected). Contrasting Fig. 3(c) and (d) suggests that the Fermi surfaces thus the underlying electronic structure are evolving with the orientation of $M$ in Co$_3$Sn$_2$S$_2$.

We now focus on the hole pocket $\alpha$ which exhibits the strongest agreement between the observed quantum oscillation frequencies and DFT. Between the two subbranches of $\alpha$ we consider the lower $\alpha_1$ (cyclotron orbit illustrated in Fig. 3(c) inset). In Fig. 3(e) we show the evolution of $\alpha_1$ obtained with a moving FFT window with $H \parallel ab$. With $H$ along the bisectrix (binary) direction, we observe an increase of approximately 15% (20%) for $\alpha$; both suggest that $\alpha$ grows in size with an in-plane magnetic field.

In Fig. 3(f) we compare the calculated oscillation spectra of $\alpha$ for both $M \parallel c$ (black curves) and $M \parallel ab$ (red curves). Although only a subset of ($M$, $H$) configurations are experimentally accessible due to the strong uniaxial anisotropy, in Fig. 3(f) we examine the theoretical cyclotron orbits perpendicular to all orientations of $H$ to carve out the overall shape of $\alpha$. For $M \parallel ab$ there is an overall increase of the pocket size of $\alpha$ compared to $M \parallel c$; the calculated Fermi pocket size increase (~10%) is comparable with albeit smaller than the experiments (15 ~ 20%). We illustrate in Fig. 3(g) the cross section of $\alpha_1$ along with that of $\gamma$ in its immediate neighborhood: an in-plane $M$ (solid lines in Fig. 3(g)) appear to increase the size of both $\alpha$ and $\gamma$ pockets and decreases the gap between the two as compared to $M \parallel c$ (dashed lines in Fig. 3(g)). Remarkably, the root of this phenomenology lies in a strong suppression of the energy gap along the nodal ring (Fig. 3(h)) when $M$ rotates into the $ab$-plane. Ultimately, our observations provide experimental evidence for an anisotropic coupling between the ferromagnetic moments with the nodal ring states via SOC.

In summary, we have identified three-dimensional Fermi surfaces of Co$_3$Sn$_2$S$_2$ in a combined SdH and DFT study. A majority of Fermi surfaces in the system are found to derive from the strong energy dispersion of the nodal rings on the (110) mirror planes. Our high field fermiology study has allowed the observation of signa-
tures of moment-orientation induced modulation of the SOC gap along the nodal line, supporting the theoretical scenario that new Weyl points can be generated and their energy tuned by an in-plane orientation of the magnetic moments [21]. Although Co$_3$Sn$_2$S$_2$ is widely viewed as a ferromagnetic Weyl semimetal, our study of the bulk Fermi surfaces of Co$_3$Sn$_2$S$_2$ reinforces the central role of the nodal ring in understanding its transport and optical responses, along with its electronic topology [12, 13, 29, 30]. The implications of our quantum oscillation studies in clean samples of Co$_3$Sn$_2$S$_2$ can also be applied to a broad class of magnetic TSMs hosting point and line nodes (e.g., Mn$_3$(Ge,Sn) [10], Co$_2$MnGa [8], Fe$_3$GeTe$_2$ [31]).

Viewed alternatively from the perspective of kagome lattice metals, we may contrast the 3D Fermi surfaces observed here in Co$_3$Sn$_2$S$_2$ with the quasi-two-dimensional Dirac fermiology reported in Fe$_3$Sn$_2$ [11, 32] and FeSn [33]. The tunability of the dimensionality of the topological states in magnetic kagome metals may be attributed to the orbital degrees of freedom of the underlying 3d electrons [33, 34]. With increasing interplane hopping, ferromagnetic kagome metals connect a 3D quantum anomalous Hall insulator phase to a 3D Weyl semimetallic phase, suggesting a topological phase diagram resembling the seminal theoretical proposal of building Weyl semimetallic phases using stacked topological insulating layers [4]. Additionally, rich magnetic phases have been reported for Co$_3$Sn$_2$S$_2$ at low magnetic fields [35, 36] as signatures of putative magnetic frustration in addition to the dominant ferromagnetic interactions; in view of the intertwined magnetic order and the topological electronic state in Co$_3$Sn$_2$S$_2$ revealed in the present study, the system provides an exciting platform to study the interplay between frustration and band topology.

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