Highly anisotropic interlayer magnetoresistance in ZrSiS nodal-line Dirac semimetal

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We instigate the angle-dependent magnetoresistance (AMR) of the layered nodal-line Dirac semimetal ZrSiS for the in-plane and out-of-plane current directions. This material has recently revealed an intriguing butterfly-shaped in-plane AMR whose origin is not well understood. Our aim was to understand the mechanism behind this peculiar shape of AMR and also to probe AMR in the out-of-plane current direction. In contrast to the in-plane AMR, the polar out-of-plane AMR shows a surprisingly different response with a pronounced cusplike feature. The maximum of the cusplike anisotropy is reached when the magnetic field is oriented in the a-b plane. Moreover, the AMR for the azimuthal out-of-plane current direction exhibits a very strong fourfold a-b plane anisotropy. Combining the Fermi surfaces calculated from first principles with the Boltzmann’s semiclassical transport theory, we reproduce all the prominent features of the unusual behavior of the in-plane and out-of-plane AMR. We can conclude that the dominant contribution the cusplike AMR lies in open orbits of the hole pocket and, in general, AMR is strongly influenced by charge compensation effect and the off-diagonal conductivity tensor elements, which give rise to peculiar butterfly-shaped AMR. Finally, the semiclassical model was also able to clarify the origin of strong nonsaturating (subquadratic) transverse magnetoresistance observed in this material, as an effect of imperfect charge-carrier compensation and open orbits.

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I. INTRODUCTION

Square-net crystal structures have been of considerable interest in structural solid-state chemistry [1]. Introducing nontrivial topology and Dirac fermions to the field of condensed-matter physics started a surge in the discovery of new materials with linear energy dispersion [2–10]. Among many square-net structures, two phases have emerged as especially interesting from the topological point of view. The ATB2 phase, where A stands for alkali or rare-earth metal with +2 oxidation state, T is a 3d transition metal and B a pnictogen group element. Typical representatives are Ca/Sr/Ba-MnBi2 that harbor quasi-2D Dirac fermions with a highly anisotropic band dispersion in the Bi-based atomic plane along with antiferromagnetic ordering in the Mn plane [11–15]. Replacing the alkali earth metal with Eu leads to an additional interlayer decoupling and the formation of the half-integer quantum Hall effect [16].

Another interesting phase is MX′X′′, which incorporates a large group of compounds [1] where M is a metal (Zr, Hf, Ta, Nb), X′ is a +2 valence state of Si, Ge, As and X′′ belongs to the chalcogen group. The prototypical representative of this group is ZrSiS, which is the subject of this paper. ZrSiS and isostructural compounds have recently gained a lot of attention due to the glide and screw symmetry-protected crossing of the conduction and valence bands, thus resulting in a nodal-line Dirac semimetal (NLDSM) phase. NLDSMs are the topological phases related to the 3D Dirac and Weyl semimetals with the difference being that the conduction and valence bands do not cross only at isolated points in the k space, but form loop or nodal-line degeneracies that give rise to interesting physical phenomena [17–22].

In the absence of spin-orbit interaction (SOI), ZrSiS has one set of nodal lines close to the Fermi energy (EF) and another set located deep in the valence band [19,23]. It has been argued that the nodal lines located in the vicinity of EF are protected by C2v symmetry and are thus susceptible to a degeneracy lifting due to the SOI that is effectively transforming the system into a weak topological insulator [24]. On the other hand, the deep-lying nodal lines are topologically protected by the nonsymmorphic symmetry. ZrSiS has several theoretically predicted unique properties among 3D Dirac semimetals, including the large interval of linear dispersion (reaching almost 2 eV) without the presence of any trivial bands and the high degree of electron-hole symmetry [23,24].
Recent studies of the Fermi surface (FS) morphology in ZrSiS by means of angle-resolved photoemission spectroscopy (ARPES) and quantum oscillation measurements (QOMs) have confirmed the nature and the position of the two pockets: a large electron and a smaller hole pocket [23,25–29]. The electron pocket has a 3D nature, whereas the hole pocket shows a quasi-2D signature [30–32]. The QOMs have also revealed the signature of another very small pocket with a puzzling Berry phase and the ultraquantum limit at around 10 T [28,29]. Due to its small size and small charge-carrier concentration, as well as its weakly elongated ellipsoidal shape observed by QOM, this pocket is most likely irrelevant for the observed charge transport effects under the rotation of magnetic field [33]. Furthermore, a high-magnetic field study of ZrSiS has revealed an interesting magnetic-breakdown effect and an unusual mass enhancement, whereas in the sister compound HfSiS an effect of Klein tunneling between electron and hole pockets was detected [34,35]. The magnetoresistance (MR) is large and unsaturated with a sub quadratic magnetic field dependence as frequently observed in the Dirac and Weyl semimetals [30,31,36,37]. On the other hand, constant field angular-dependent magnetoresistance (AMR) measurements have observed an unexpected and intricate butterfly-shaped anisotropy for current applied along the in-plane axes [30,32,38,39].

In this paper, we present a detailed study of magnetic field (B) and temperature (T) dependence of the AMR in ZrSiS single crystals for current oriented along high-symmetry directions. The AMR has proved to be a very powerful tool for studying the FS shape of 3D and quasi-2D (q-2D) systems, with many materials exhibiting nonclassical behavior [40–42]. We have performed out-of-plane AMR measurements (inter-AMR) with current along the c direction and in-plane AMR measurements (intra-AMR) with current along the a(b) axis. To the best of our knowledge, ZrSiS and related compounds have not been previously characterized for the current oriented along the c direction. For current along the c direction, polar inter-AMR reveals a large cusplike anisotropy which becomes pronounced close to the a-b plane. Additionally, the azimuthal inter-AMR shows a strong a-b plane anisotropy with fourfold symmetry and a minimum at an angle corresponding to an odd multiple of π/4. In the case of the intra-AMR, the polar scan displays a previously observed butterfly-like shape.

To understand the striking difference of the AMR response for the in-plane and out-of-plane current directions and to elucidate the role of the putative q-2D hole pocket, we have employed a theoretical transport model based on the FS calculated from first principles combined with the semiclassical Boltzmann’s equation. Using this model, we were able to reproduce all features observed in experimental data. Our model explains the intricate butterfly-like AMR and strongly anisotropic inter-AMR in terms of charge-carrier compensation due to electron and hole pockets and the effect of strong off-diagonal elements in the conductivity tensor. Furthermore, by using the model we clarify the origin of large sub quadratic nonsaturating MR as an effect of the imperfect charge-carrier compensation. Finally, combining the experimental AMR results and the theoretical transport model, we were able to refine the shape of the FS and estimate the average scattering time and the mean-free path.

II. RESULTS AND DISCUSSION

Single crystals of ZrSiS were grown by chemical vapor transport and show excellent quality with a low-T in-plane resistivity of only 0.1 μΩcm [32]. Due to their layered crystal structure, ZrSiS commonly grows in as plate-like crystals, with a thickness of around 100 μm. By optimization of the synthesis procedure, we managed to obtain samples of sufficient thickness (in the mm range) which also allowed us to measure the out-of-plane transport properties. All measured samples S1, S2, and S3 are cut from the same bigger single crystal whose quality was verified before cutting by observation of pronounced quantum oscillations. The zero B temperature dependence of the resistivity for the in-plane current direction ρ_{ab} of sample S1 shows metallic behavior [Fig. 1(a)] with a residual resistivity ratio (RRR = ρ_{300K}/ρ_{1.8K}) of around 80. The out-of-plane resistivity ρ_{c} of sample S2 also shows metallic behavior but with a considerably higher resistivity contributing to a moderate anisotropy ρ_{c}/ρ_{ab} of around 50 at the lowest measured temperature. The almost identical T profile for the in-plane and out-of-plane transport points...
the total magnetotransport of the charge carriers should be smaller frequency is not yet determined. Due to its small size and 241 T. ARPES measurements unambiguously related the tum oscillations with frequencies corresponding to 8.5 T Sample S1 displays strong Shubnikov-de Haas (SdH) quan-
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of high mobility charge carriers. The MR of samples S1

Dirac and Weyl semimetals arising from multiband transport

nonsaturating MR is a commonly observed property of 3D

transport model, we were able to understand the origin of

orbital [47]. By using the framework of the semiclassical

magnetically ordered materials where its origin is not purely

to the in-plane orientation, the dip in the AMR appears at the

has a complicated structure, details for sample S3 at 10 T are

is slightly misaligned with respect

vanishing Lorentz force. In the model of a single spherical

FS it should be zero. By tilting B away from the c axis (in the

c-a plane) the AMR shows a strong increase in magnitude that

becomes more pronounced with increasing B and forms a
cusplike feature. The anisotropy ratio of the MR for the

longitudinal and transverse B orientations is around 50 at

10 T. The strong increase of inter-AMR when B is close to

the in-plane direction is a feature commonly observed in

q-2D materials and it is associated with coherent in-plane

transport, i.e. a small warping of the 2D FS [48,49].
The coherence peak is usually accompanied by Yamaji oscillations

in inter-AMR, but are not detected in our samples [50]. The

small oscillations in the AMR profile at higher B originate

from the quantum oscillations, which is supported by the

field dependence of the oscillations peak positions. Recently,
in several Dirac semimetals with square-net structure, the

peaklike response has been observed for B close to the a-b

plane, which was explained by q-2D FS [11,15] or by the

interlayer quantum transport governed by the Dirac point [51].

In our case, as we show later in the text, this feature can be

explained by the semiclassical Boltzmann’s theory. The inset

in Fig. 2(a) shows temperature dependence of the inter-AMR

for sample S2 at 9 T. By increasing T the cusplike shape of

the inter-AMR weakens and, at around 200 K, it acquires the

classical sinusoidal shape. Performing the same polar scans

but now for different angles φ, we observe an indication of

strong in-plane anisotropy [see Fig. 2(b)]. The polar inter-

AMR becomes significantly weaker as φ approaches π/4.

The azimuthal inter-AMR (φ angle rotation) given in

Fig. 3(a) shows a strong fourfold a-b plane anisotropy with

the maxima positioned along the high-symmetry axes a and b

and minima along the bisector axes (odd multiples of π/4).
The strength of the anisotropy (ratio of the maximum and

minimum values) is almost B independent in the measured

range between 5 T and 9 T and its value is around 4. This

is in contrast to the anisotropy of the polar inter-AMR that

is continuously growing with B. The observed a-b plane

anisotropy is fairly large. For comparison, it is roughly a factor of 2 larger than that in Sr2RuO4 [42,52]. For larger B

(>5 T), the AMR becomes truncated close to the bisector axis,

whereas at 2 T the truncation is not observed. The effect of

quantum oscillations is clearly seen at 9 T close to the high-

symmetry axes. Closer inspection reveals that the truncation

has a complicated structure, details for sample S3 at 10 T are

shown in Fig. 3(b). When B is slightly misaligned with respect

to the in-plane orientation, the dip in the AMR appears at the

bisector axis, whereas when B is in-plane, the truncated part

shows oscillatinglike behavior which is B independent and

cannot be related to quantum oscillations. This unusual

behavior is probably related to the local morphology of the

FS. Considering the temperature dependence of the azimuthal

inter-AMR it can be seen that, above 100 K, the truncated part disappears [see Fig. 3(c)] and the anisotropy of the

in-plane AMR weakens. In Fig. 3(d), we present a detailed

spectroscopic mapping of the inter-AMR. It can be seen that

the AMR pattern has a twofold symmetry for φ-angle rotation

and fourfold symmetry for θ-angle rotation. The maximum

towards the coherent in-plane transport, which is expected in

transport dominated by a 3D-FS.

The transverse MR [43] given in Fig. 1(b) shows a very

strong response reaching almost 20 000 % at 10 T for both

orientations (S1: B∥[c, I]/[a, I] and S2: B∥[a, I]/[c]). A strong, non

saturating MR is a commonly observed property of 3D

Dirac and Weyl semimetals arising from multiband transport

of high mobility charge carriers. The MR of samples S1

and S2 has a subquadratic B dependence [44], which has been

recently associated with a B-dependent mobility [45,46].

Sample S1 displays strong Shubnikov-de Haas (SdH)

quantum oscillations with frequencies corresponding to 8.5 T

and 241 T. ARPES measurements unambiguously related the

higher frequency (241 T) to the q-2D tabelike hole pocket,

whereas the position and exact shape of the pocket with the

smaller frequency is not yet determined. Due to its small

size and weakly elongated ellipsoidal shape, its contribution to

the total magnetotransport of the charge carriers should be

negligible. SdH oscillations observed in sample S2 (different

orientation of B) are less pronounced and composed of several

frequencies (17 T, 23 T and 170 T).

Under the polar rotation (angle θ) of sample S1 at B = 9 T the transverse intra-AMR [43], shown in Fig. 1(c) exhibits a peculiar fourfold butterfly-shaped angular dependence with the angle of maximum resistivity at odd multiples of θ = π/4. The origin of this peculiar intra-AMR has been elusive since, previously, this kind of AMR was only observed in magnetically ordered materials where its origin is not purely orbital [47]. By using the framework of the semiclassical transport model, we were able to understand the origin of the intra-AMR in terms of the charge-carrier compensation effects of the electron and hole pockets. Figure 1(d) displays a detailed spectroscopic mapping of the intra-AMR transport. When B is tilted away from the c axis, the AMR increases showing the butterfly-shaped profile for all values of the azimuthal angle φ. On the other hand, for the in-plane rotation (a-b plane) the intra-AMR is small and the anisotropy is weak.

Figure 2(a) presents details of the polar scan of the inter-

AMR at several discrete values of B between 2 T and 10 T for current along the c axis for sample S3. For the longitudinal configuration (B and current are along the c axis), the MR is small for all measured B, which is expected due to the vanishing Lorentz force. In the model of a single spherical FS it should be zero. By tilting B away from the c axis (in the c-a plane) the AMR shows a strong increase in magnitude that becomes more pronounced with increasing B and forms a cusplike feature. The anisotropy ratio of the MR for the longitudinal and transverse B orientations is around 50 at 10 T. The strong increase of inter-AMR when B is close to the in-plane direction is a feature commonly observed in q-2D materials and it is associated with coherent in-plane transport, i.e. a small warping of the 2D FS [48,49].

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FIG. 2. (a) Polar inter-AMR for several constants B at T = 1.8 K and φ = 0°, with the current in the out-of-plane direction (along the c axis) for sample S3, shows strikingly different behavior than the intra-AMR. B is rotated in the c-a plane. The AMR shows a pronounced increase in magnitude as B is rotated toward the a-b plane and forms a cusplike feature. The inset shows the temperature dependence of polar inter-AMR. The anisotropy becomes very weak around 100 K. (b) Polar intra-AMR for several different directions along the azimuthal (φ) angle reveals a strong a-b plane anisotropy.

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in the AMR only appears when $B$ is oriented along the high-symmetry in-plane directions.

To achieve a deeper understanding of the unusual AMR response in ZrSiS we have employed numerical modeling using the semiclassical Boltzmann’s transport theory \cite{53,54} as implemented in WannierTools open-source software package \cite{55}. A detailed description of the methodology along with representative examples is given in Ref. \cite{54}. We have used a FS calculated using DFT to obtain the Boltzmann’s conductivity tensor $\sigma_{ij}$ and then transformed into the total resistivity tensor $\rho_{ij} = |\sigma_{ij}|^{-1}$. Analyzing individual contributions of electron and hole pockets will help us clarifying the physical mechanism underlying the discussed magnetotransport properties.

As a first step, in Fig. 4(a) we present the transverse angular resistivity $\rho_{zz}(\theta)$ calculated at a fixed magnitude of $B$ for the in-plane current direction. The calculations reproduce very well the experimental data (black open circles), and in particular the twofold symmetry butterflylike shape. The green and blue lines in Fig. 4(a) represent individual contributions of the electron ($\rho_{zz}^e$) and hole ($\rho_{zz}^h$) pockets, respectively. Closer inspection reveals that the total angular resistivity is not a simple sum of electron and hole (parallel) channels. Only for the high-symmetry directions, i.e., $B||c$ and $B||a$, the total calculated resistivity is smaller that the resistivity of the contributing channels, giving rise to a noticeable drop in the intra-AMR. For other directions, we cannot apply the parallel channel rule due to a significant contribution of the off-diagonal elements of the conductivity tensor, from which $\rho_{zz}(\theta)$ is obtained.

Next, we use our model to understand the mechanism of the observed large nonsaturating transverse MR for the in-plane current direction. While nonsaturating MR with $B^2$ dependence is usually assigned to systems with perfect electron-hole compensation, deviations from the ideal quadratic scaling often observed in the high mobility systems with electron and hole pockets has recently been attributed to the field-dependent mobility \cite{46}. The transverse MR for $B||c$ scales as $B^{1.65}$, which is in excellent agreement with the measured $B^{1.67(1)}$ dependence \cite{4b}. The strong orbital MR in our case comes from the imperfect compensation effect between electron and hole pockets.

We now proceed with the analysis of inter-AMR with current oriented along the $c$ direction for two distinct measurement configurations—polar ($\theta$-scan) and azimuthal ($\phi$-scan) ones. Figure 4(d) shows the calculated polar angular resistivity $\rho_{zz}(\theta)$ (red solid line) for the current and field orientations defined in the figure. The calculated resistivity has a strong angular dependence with a cusplike shape and a maximum being reached for the in-plane oriented field. Comparison of the calculated and measured (black solid circles) angular dependences again shows good agreement. To understand the origin of this unusual behavior we examine the individual contributions of the electron and hole pockets, $\rho_{zz}^e$ and $\rho_{zz}^h$, respectively. Comparison of the calculated magnetoresistivity $\rho_{zz}(\theta)$ with individual components again shows that the total resistivity cannot be described by combining two parallel channels and the off-diagonal elements play a significant contribution. Both matrix elements have a strong peak for $B$ close to the in-plane orientation, but of distinct origins. The peak in $\rho_{zz}^h$ originates from the open orbits, since the open orbits extend along the $c$ axis in the presence of field $B$ parallel to the $a$-axis, the velocity $v_\parallel$ remains finite in contrast to a very small velocity $v_\perp$, resulting in significant resistivity $\rho_{zz}^h$. Moreover, when the $B$ field is rotated away from the $a$ axis by ca. 15 degrees, the open orbit disappears in the cross-section of the FS, and the large resistivity $\rho_{zz}^h$ reduces to smaller magnitude. On the other hand, the peak in $\rho_{zz}^e$ originating from the electron pocket deviates strongly from the ideal free-electron spherical shape, i.e., it results from the pocket flatness.

As a next step, we aim at understanding the $a$-$b$ plane anisotropy of the intra-AMR [Fig. 4(e)]. Good agreement of the calculated and experimental angular dependencies is achieved as in the previous cases. Comparing $\rho_{zz}(\theta)$ with the contributions of individual pockets $\rho_{zz}^h(\phi)$ (green line) and $\rho_{zz}^e(\phi)$ (blue line), allows us to conclude that the angular dependence of $\rho_{zz}(\phi)$ is mostly due to the hole pocket. The truncated part of the azimuthal AMR most likely also has its origin in the hole pocket since $\rho_{zz}^h$ shows anomalous behavior.
We are now able to comment on the average relaxation time in the AMR is mostly due to the open orbits. It has been argued that in these materials the peaklike structure of open orbits, the charge-carrier compensation effect also plays an important role since open orbits cannot account for the observed azimuthal angular dependence. Recently, strong increase in the inter-AMR close to the $\mathbf{c}$-$\mathbf{a}$ plane has been reported in several similar square-net Dirac materials. These discrepancies can be related to the sensitivity of the FS shape calculated using DFT to the exact path $I-X$, various details of the methodology, such as pseudopotentials used in the calculations. The reported FS shapes have a similar structure of the electron and hole pockets in the $k_z = \pi/c$ plane, but differ significantly in the $k_z = 0$ plane. Figure 4(c) shows our calculated FS characterized by the hole pocket consisting of an elongated tubelike structure that gives rise to open orbits. In the $k_z = 0$ plane, the hole pocket has protruding armlike features extending along the $X-X$ lines. Good agreement between all the peculiar features of the calculated and experimental AMR provides a strong indication that the calculated FS reproduces the real one.

### III. Conclusion

In conclusion, we have presented a detailed study of the AMR in the NLDSM ZrSiS. We have determined the low-temperature, zero-field anisotropy between the in-plane and out-of-plane directions to be moderately strong with typical values around 50. The AMR was measured in two configurations, for current oriented in-plane along the $a$ axis (intra-AMR) and out-of-plane along the $c$ axis (inter-AMR). The intra-AMR shows an unusual butterflylike shape previously reported by other authors. The inter-AMR shows a strong cusplike shape anisotropy for polar angle rotation, with a maximum achieved for the magnetic field oriented in the $a$-$b$ plane.
plane. Additionally, the azimuthal angle rotation shows strong anisotropy with a four-fold symmetry, with the minimum at odd multiples of $\phi = \pi/4$. To understand this intricate AMR, we have employed a theoretical model based on the FS calculated from first principles and the Boltzmann’s semiclassical theory. The model successfully reproduced all observed features of both the inter- and intra-AMR. The cusplike shape of AMR can be attributed to open orbits of the hole pocket which strongly reduce $v_\phi$ velocity component and thus increase the resistivity. Deviating from the in-plane high symmetry axes, the open orbits have less impact and $v_\phi$ is increased which produces the azimuthal anisotropy. Furthermore, our model explains the subquadratic dependence of transverse magnetoresistance as an effect of imperfect charge-carrier compensation for the out-of-plane current direction. We were able to estimate the average relaxation time to be around $\tau \approx 2.6 \times 10^{-14}$ s, the mean-free path $l \approx 15$ nm, and more accurately determine the FS shape.

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**APPENDIX: SEMICLASSICAL BOLTZMANN’S APPROXIMATION**

Within the relaxation time approximation, the bandwise conductivity tensor $\sigma$ is calculated by solving the Boltzmann’s equation in presence of an applied magnetic field as

$$\sigma^{(n)}_{ij}(B) = \frac{e^2}{4\pi^3} \int d\mathbf{k} \tau_n v_n(\mathbf{k}) \tilde{v}_n(\mathbf{k}) \left( -\frac{\partial f}{\partial \epsilon} \right)_{\epsilon = \epsilon_n(\mathbf{k})},$$

where $e$ is the electron charge, $n$ is the band index, $\tau_n$ is the relaxation time of $n$th band that is assumed to be independent on the wave vector $\mathbf{k}$, $f$ is the Fermi-Dirac distribution, $v_n(\mathbf{k})$ is the velocity defined by the gradient of band energy $v_n = \frac{1}{\hbar} \nabla \epsilon_n(\mathbf{k})$, \tilde{v}_n(\mathbf{k}) is the weighted average of velocity over the past history of the charge carrier

$$\tilde{v}_n(\mathbf{k}) = \int_{-\infty}^{0} \frac{dt}{\tau_n} e^{\frac{\epsilon_n(\mathbf{k})}{k_B T}} v_n[\mathbf{k}(t)],$$

The orbital motion of charge carriers in applied magnetic field causes the time evolution of $k_n(t)$, written as

$$\frac{dk_n(t)}{dt} = \frac{-e}{\hbar} v_n[\mathbf{k}(t)] \times B,$$

with $k_n(0) = \mathbf{k}$. The total conductivity is the sum of bandwise conductivities, i.e., $\sigma_{ij} = \sum_n \sigma^{(n)}_{ij}$, which is then inverted to obtain the resistivity tensor $\rho = \sigma^{-1}$.

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