Observation of Open-Orbit Fermi Surface Topology in Extremely Large Magnetoresistance Semimetal MoAs$_2$

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While recent advances in band theory and sample growth have expanded the series of extremely large magnetoresistance (XMR) semimetals in transition metal dipnictides $TmPn_2$ ($Tm = Ta, Nb; Pn = P, As, Sb$), the experimental study on their electronic structure and the origin of XMR is still absent. Here, using angle-resolved photoemission spectroscopy combined with first-principles calculations and magnetotransport measurements, we performed a comprehensive investigation on MoAs$_2$, which is isostructural to the $TmPn_2$ family and also exhibits quadratic XMR. We resolve a clear band structure well agreeing with the predictions. Intriguingly, the unambiguously observed Fermi surfaces (FSs) are dominated by an open-orbit topology extending along both the [100] and [001] directions in the three-dimensional Brillouin zone. We further reveal the trivial topological nature of MoAs$_2$ by bulk parity analysis. Based on these results, we examine the proposed XMR mechanisms in other semimetals, and conclusively ascribe the origin of quadratic XMR in MoAs$_2$ to the carriers motion on the FSs with dominant open-orbit topology, innovating in the understanding of quadratic XMR in semimetals.

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The emergence of novel states in condensed matter is not only classified by the typical spontaneous symmetry breaking, but also by their topology, i.e., the topologically protected quantum states [1–3]. The discovery of such symmetry protected states of matter in two-dimensional (2D) [4–6] and three-dimensional (3D) topological insulators [7], node-line semimetals [8, 9], topological crystalline insulators [10, 11], and Dirac and Weyl semimetals [12–17], has attracted tremendous interests in condensed matter physics and materials science. The magnetotransport behavior of these states is often unusual, such as linear transverse magnetoresistance (MR) and negative longitudinal MR in Dirac and Weyl semimetals [18,24], and more generally, extremely large transverse MR (XMR) in nonmagnetic semimetals [25,30].

Recently, the discovery of XMR in a class of transition metal dipnictides $TmPn_2$ ($Tm = Ta, Nb; Pn = P, As, Sb$) [31–36] has sparked immense interests for understanding the underlying mechanism of quadratic XMR and exploring novel quantum states arising from nontrivial topology. Another two series of semimetals possessing quadratic XMR behavior and rich topological characteristics are the ZrSiS family [37–39] and LnX ($Ln = La, Y, Nd$, or Ce; $X = Sb/Bi$) series [40–48], whose electronic structures have been considerably studied both in theory and experiment [47,53]. While the band structures of the $TmPn_2$ series have been theoretically characterized in several work [32,34,54], experimental observations have not yet been reported. It is widely believed that the large positive MR in semimetals is intimately related to their underlying electronic structures. Therefore, a systematic and unambiguous experimental study on the electronic structure of the $TmPn_2$ family is urgently demanded. Eventually, we suggest the open-orbit Fermi surface (FS) topology as another candidate mechanism to explain the XMR, in addition to the earlier proposed origins like nontrivial band topology [40], forbidden backscattering at zero field [55], and electron-hole compensation [56].

In this Letter, we employ systematic angle-resolved photoemission spectroscopy (ARPES), first-principles calculations, and magnetotransport measurements on MoAs$_2$, which is isostructural to the $TmPn_2$ family and exhibits quadratic MR exceeding $3.2 \times 10^4\%$ at $1.8 \text{ K}$ under 9-T magnetic field. The FS topology is clearly resolved to display two “double ripple”-shaped FSs extending along the [001] direction (a-axis), which do not close along the [001] direction (normal to the $ab$-plane) either, and one pocket at X. Besides, we identify two “handle”-like FSs around $\bar{\Gamma}$, which arise from a trivial massless surface state (SS) along $\bar{\Gamma}$-$\bar{X}$. Our detailed electronic structure of MoAs$_2$ would facilitate a more comprehensive understanding of the quadratic XMR in the $TmPn_2$ family.

High-quality single crystals of MoAs$_2$ with large residual resistance ratio ($RRR = R(300 \text{ K})/R(1.8 \text{ K}) = 1238$) were grown via the chemical vapor transport method. Detailed methods for ARPES and first-principles calculations can be found in Sec. I of Supplemental Material (SM) [57]. MoAs$_2$ crystallizes in a monoclinic structure with space group $C2/m$ (No. 12), as illustrated in Fig. 1(a) [58]. It is isostruc-
FIG. 1: (a) Schematic crystal structure of MoAs$_2$. (b) XRD pattern on the (001) surface. (c) Schematic primitive BZ and 2D projected BZ of the (001) surface. (d) Constant energy ARPES image obtained by $h\nu = 42$ eV at $E_F$ with the mapping center around $\Gamma$ point. Cut 1 indicates the momentum location of the measured bands in Figs. 2(a) and 2(b). Red lines represent the (001) surface BZ. Orange dashed curves are guides to the eye for the “double ripple”-shaped FSs. (e) Calculated FSs for a 10-unit-cell-thick (001) slab with the MoAs-terminated layer. The inner and outer “ripple”-shaped FSs are indicated by green and cyan arrows, respectively. Blue lines represent the (001) surface BZ. The intensity of the color scales the spectral weight projected to the topmost unit cell. (f) Temperature dependence of the resistivity in different magnetic fields. The magnetic field is applied, one can see a prominent up-turn and a saturation at low temperatures. As depicted in Fig. 1(g), MoAs$_2$ exhibits rather large MR exceeding $3.2 \times 10^4 \%$ at 1.8 K under 9-T magnetic field, and there is a drastic suppression of MR with increasing temperature. One can obtain a nearly-quadratic exponent $m = 1.97$ by fitting the $MR-H$ profile at 1.8 K with a power-law function. These fingerprints are consistent with the common features of XMR semimetals [56].

In order to uncover the underlying nature of quadratic XMR and topological characteristics in MoAs$_2$, we investigate the near-$E_F$ band dispersions along $\Gamma - \bar{X}$ and $\bar{Y} - \bar{X}$. The band structure along $\Gamma - \bar{X}$ is presented in Figs. 2(a)-(c). As shown in Figs. 2(a) and 2(b), we identify one Dirac cone-like band centered at $\bar{\Gamma}$ that will be discussed below. Three additional bands crossing $E_F$ are clearly distinguished. The innermost electron band ($\alpha_1$) disperses slowly and crosses $E_F$ at $k_z \sim 0.58 \, \text{Å}^{-1}$, forming the inner “ripple”-shaped FS, then it turns back as the outmost electron band ($\alpha_2$), which is in agreement with the bulk band calculations along $\Gamma - X$ depicted in Fig. 2(c). As the absence of the middle one ($\alpha_3$), which forms the outer “ripple”-shaped FS, in the calculations along $\Gamma - X$, we suggest that $\alpha_3$ may not come from the $k_z = 0$ plane. Due to the short escape length of the photoelectrons excited by the vacuum ultraviolet light in our ARPES experiments, the $k_z$ broadening effect would be prominent [60], demonstrated by previous ARPES studies [51], [52], [61], [64]., we further perform bulk band calculations along $Z - I_1$ (in the $k_z = \pi$ plane) as shown in Fig. 2(c). The consistency between $\alpha_3$ and the calculations helps finding out the origin of this band [65].

We investigate the electronic structure along $\bar{Y} - \bar{X}_1$ to examine the origin of the “double ripple”-shaped FSs. As indicated in Fig. 2(d), the “double ripple”-shaped feature can be resolved more clearly from the FS mapping focusing around $\bar{Y}$ point, while the “handle”-like FSs are relatively weak in this geometry due to the matrix element effect. From the measured band dispersions along $\bar{Y} - \bar{X}_1$ illustrated in Figs. 2(e) and 2(f), one can observe two almost linearly dispersive bands crossing $E_F$ indicated by white arrows. These two bands are denoted as $\beta_1$ and $\beta_2$, which correspond to the inner and outer “ripple”-shaped FSs, respectively. Detailed energy distribution curve (EDC) analysis on $\beta_1$ and $\beta_2$ are shown in Figs. 2(j) and S3 (Sec. IV of SM [57]), respectively. The experimental band structures agree with the slab calculations along $\bar{X}_1 - \bar{Y} - \bar{X}_1$ shown in Fig. 2(g). By compared with the bulk calculations along $Y - X_1$ (near the $k_z = \pi$ plane), displayed as Fig. S2(b) in Sec. III of SM [57], the fact that $\alpha_3$, which is captured by the calculations along $Z - I_1$ rather than $\Gamma - X$, derives from the projection of the $k_z \approx \pi$ plane due to the $k_z$ broaden-
FIG. 2: (a),(b) Photoemission intensity plot and corresponding second derivative plot along \( \Gamma - \bar{X} \) [cut 1 in Fig. 1(d)], respectively. (c) Calculated band structures along \( \Gamma - I_1 \), considering that \( X \) and \( I_1 \) points are approximately projected to one point on the (001) surface. Cuts 2 and 3 indicate the momentum locations of the experimental band structures in (e)-(j). The electronic structures measured along cut 4 are presented in Fig. S4 (Sec. V of SM [57]). (e),(f) Same as (a),(b) but along \( \bar{Y} - \bar{X}_1 \) [cut 2 in (d)]. Red dashed rectangle illuminates the area for EDC analysis in (j). (g) Calculated band structure along \( \bar{X}_1 - \bar{Y} - \bar{X}_1 \) for a 10-unit-cell-thick (001) slab. (h),(i) Same as (a),(b) but along cut 3 in (d). (j) EDC plot of (e). The band gap is highlighted by black curve. Blue dots are extracted peak positions, serving as guides to the eye.

The consistency between measured electronic structures along \( \bar{Y} - \bar{X}_1 \) and theoretical calculations can be proved by the presence of a band gap between \( \beta_1 \) and \( \beta_2 \) (see a quantitative analysis on this gap in Sec. V of SM [57]). To further demonstrate the open character of the in-plane FSs, we study the band structure along cut 3, i.e., \( k_y \sim 0.50 \text{ Å}^{-1} \), the momentum where the inner and outer “ripple”-shaped FSs are closest to each other. In Figs. 2(h) and 2(i), as marked by vertical arrows, the two linearly dispersing bands are clearly separated at \( E_F \).

While most experimental band structures along \( \bar{\Gamma} - \bar{X} \) are in agreement with the bulk band calculations, we observe an extra Dirac cone-like band centered at \( \bar{\Gamma} \). To illustrate the origin of this band, we investigate the band dispersions recorded with different photon energies [66, 67]. We show the constant energy plot in the \( k_x - k_z \) plane at \( k_y = 0 \) at \( E_F \) and \( E = -0.24 \text{ eV} \), which is the binding energy of the Dirac point (DP) at \( \bar{\Gamma} \), in Figs. 3(a) and 3(c), respectively. The corresponding momentum distribution curve (MDC) plots are presented in Figs. 3(b) and 3(d), respectively. The Dirac-like band features, including the Fermi crossings and the DP, do not show noticeable change with a varying photon energy over a wide range, confirming that it is a SS [68].

We further perform \( k_z \)-dependent measurements along \( \bar{\Gamma} - \bar{X}_1 \) focusing on the “double ripple”-shaped FSs. The photon energy variation covers more than one BZ along the \( k_z \) direction (~0.88 \text{ Å}^{-1}), which is sufficient to illustrate the periodicity along \( k_z \). As shown in Figs. 4(a)-4(c), the “double ripple”-shaped open FSs do not close along the \( k_z \) direction either (see Fig. S5 in Sec. VI of SM for more manifestations of the open character [57]). When the FS topology contains open orbits along certain directions, MR in materials is not saturated and parabolically dependent on the magnetic field [69, 70]. To elucidate the possible electronic origin of quadratic XMR in MoAs\(_2\), angular-dependent MR measurements were carried out (see details of the experimental setup and discussions on the anisotropy of MR in Sec. VIII of SM [57]).

So far, immense work has been generated on the origin of quadratic XMR behavior, and several mechanisms have been proposed, including nontrivial band topology [40], forbidden backscattering at zero field [55], and electron-hole compensation [56]. First, the field-induced resistivity upturn and plateau is suggested as the consequences of breaking time-reversal symmetry in topological semimetals [40], whereas the \( Z_2 \) classification of MoAs\(_2\) is (0; 000) [71] based on our first-principles calculations (detailed analysis can be found in Sec. VII of SM [57]), demonstrating that MoAs\(_2\) is a topologically trivial material. This excludes the possibility that the XMR in
MoAs$_2$ is associated with nontrivial band topology. Second, the spin degeneracy is removed by spin-orbit coupling due to the lack of inversion symmetry in WTe$_2$, leading to an exotic spin texture of the bands, which has been claimed to play an important role in the XMR of WTe$_2$ [55]. This explanation is not applicable to MoAs$_2$ either, because the inversion symmetry in MoAs$_2$ preserves that its spins are doubly degenerate at zero field. Third, while electron-hole compensation can be broadly applied in many XMR semimetals with closed FS trajectories based on the two-band model [41, 51, 56, 72], its availability in XMR materials with open FSs is still unclear [73]. From another perspective, it should be noted that the MR tends to a constant, i.e., it saturates, in high fields for FSs consisting of closed electron and hole pockets, unless they have exactly equal volume. This contrasts with the MR for open FSs, which increases as $H^2$ and exhibits non-saturation without the restriction of carrier compensation [74]. For the former, under the influence of a magnetic field, carriers in clean materials with closed FSs will travel several orbits before getting scattered, resulting in velocity average to zero in the plane perpendicular to the magnetic field. As for the carriers on open FSs, for which the product of the cyclotron frequency ($\omega_c$) and the relaxation time ($\tau$) is no longer much larger than 1, thus leading to a finite in-plane velocity. These two conditions cause above two distinct field-dependence [69] [70]. Since the dominant open-orbit FS topology can, by itself, lead to the quadratic XMR in MoAs$_2$, while the dominant closed FSs are necessary for the non-saturated MR induced by carrier compensation, thus, electron-hole compensation mechanism cannot be applied to the XMR in MoAs$_2$.

Hence, it is reasonable to suggest that the quadratic XMR in MoAs$_2$ is attributed to the carriers motion on the FSs with dominant open-orbit topology. Furthermore, despite what topology the FSs are, the ultra-high charge mobilities are the key condition effectively enhancing the MR in semimetals [41] [70], this is also applicable in MoAs$_2$, of which $\mu_e$ and $\mu_h$ is $1.04$ and $3.49 \times 10^4$ cm$^2$V$^{-1}$s$^{-1}$, respectively. They are determined by fitting the Hall conductivity at $T = 1.8$ K using the two-carrier model [75] [76], as illustrated in Fig. S1(b) (Sec. II of SM [57]). The open FS character along certain directions has also been observed in PdCoO$_2$ and ZrSiS exhibiting the XMR behavior [77] [79]. However, for PdCoO$_2$, the field-dependence of MR is not parabolic and the role of open FSs on the XMR has not yet been studied [77]; for Zr-
SiS, the carrier compensation is proposed to be dominant in the quadratic XMR when $H \parallel [001]$, while the open-orbit FS topology serves to enhance the MR (∼50%) when $H \parallel [011]$, as the existence of open FSs when $H \parallel [010]$ \cite{79}.

In conclusion, we present the comprehensive electronic structure of XMR semimetal MoAs$_2$. The observed FSs are dominated by open-orbit topology extending along both the [100] and [001] directions. We demonstrate the trivial topological nature of MoAs$_2$ by bulk parity analysis. Our results unambiguously suggest that the origin of quadratic XMR in MoAs$_2$ is attributed to the carriers motion on the FSs with dominant open-orbit topology, serving as a novel mechanism for XMR in semimetals.

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