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Multiple Topological States in LaAgAs_2 , a Failed Square-Net Semimetal

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Abstract

The rational design of new materials emerges as an important direction to explore new topological materials, which is based on the understanding of the correlation between crystal and electronic structures. In this paper, we perform a comprehensive study on the crystal and electronic structures in LaAgAs_2 through a combination of single-crystal x-ray diffraction (XRD), quantum oscillation, and angle-resolved photoemission spectroscopy (ARPES) experimental measurements, and density functional theory (DFT) calculations. Single-crystal XRD measurements reveal that LaAgAs_2 crystallizes into a HfCuSi_2 -derived structure with the square net distorted into cis-trans chains. Quantum oscillation measurements reveal two frequencies with small effective masses and quasi-two-dimensional (2D) characters. ARPES measurements reveal an electronic structure strikingly different from the square-net-based semimetals, such as LaAgSb_2 . The Fermi surface is quasi-two-dimensional (2D), with Dirac-like hole pockets at the zone center and a quasi-1D elliptical electron pocket at the zone boundary. Based on the DFT calculations, the measured electronic structure can be well understood regarding the cis-trans distortion, which transforms the two-dimensional square net-derived Dirac bands into quasi-1D trivial bands. Intriguingly, multiple topological states can be identified around the zone center, including a nontrivial Z_2 topological surface state and a bulk Dirac state. Our study clarifies the impact of cis-trans distortion and identifies LaAgAs_2 as a topological material with multiple topological states near the Fermi level, providing a guideline for intentionally designing new topological materials.

INTRODUCTION

Topological materials, including topological insulators and topological semimetals, are unconventional phases of matter characterized by topologically nontrivial band structures [1–3]. Owing to their unconventional electronic and transport properties, great effort has been devoted to the discovery of new topological materials [4–8]. One of the most successful strategies is to design the new layered materials using the LEGO-like building block approach, specifically by alternatively stacking structural motifs that host desired band structure and the buffer layers along the out-of-plane direction [9–12]. This strategy is based on the assumption that the electronic structure of the building block can be regarded as an entirety, which could keep its main feature when embedded in real materials [13–15].

However, in real materials, both the crystal and band structures of the building blocks are tunable by local chemical environments, for example, structural distortions and band dispersion. To improve the efficiency and accuracy of designing new topological materials, it is important to clarify the impact of structural distortions and explore new building blocks.

Due to its simplicity and prevalence in real materials, the planar square lattice has attracted extensive theoretical and experimental interest [10]. Theoretically, it has been extensively studied in the context of the Ising model and 2D Dirac semimetal [16, 17]. Experimentally, the square lattice is found to be linked to various intriguing phenomena, such as anisotropic Dirac fermions [18, 19], charge density waves (CDWs) [20, 21], and superconductivity [22], making it an excellent platform for scrutinizing the intertwined orders. Notably, cuprate-, iron-, and nickel-based high-temperature superconductors with complex phase diagrams are associated with the square lattice [23–26]. In real materials, the square lattice might undergo structural distortions, e.g., distorted into zigzag chains or cis-trans trains, which will alter its electronic structure and related physical properties [27]. Therefore, clarifying the distortion effects on the square lattice is crucial to understanding square-net-based materials.

In topological materials, many crystal structures can host the prototype square-net-derived band structure, which can be classified as the square-net-based topological materials [10, 28]. Primary examples include the HfCuSi₂-type structure (e.g., SrMnBi₂ and LaAgSb₂ [29, 30]) and PbFCl-type structure (e.g., ZrSiS and GdSbTe [31, 32]), as well as PtPb₄ [33]. Owing to their structural diversity and the robust square-net-derived bands, HfCuSi₂-type compounds with the general chemical formula $AMPn_2$ (A = alkaline earth or rare earth; transition metal; Pn = Sb or Bi) are a central platform for studying the square-net-based topological semimetals [10]. The crystal structure is typically described as the alternating stacking of the A layer, anti-PbO-type $[MPn]$ layer, and square-net-type Pn planar layer along the stacking direction. Most of the notable phenomena observed in $AMPn_2$ materials arise from the Pn square lattice. A and $[MPn]$ layers are merely treated as buffer layers that influence physical properties through interaction with the Pn square lattice [18].

Previous studies mainly focus on tuning the band structure of Pn square net via manipulating the A and $[MPn]$ layers [34, 35]. However, little attention has been paid to the buffer layers and the distortion of the square net. However, it has been reported that the $[\text{FeAs}]/[\text{FeTe}]$ layer with the same crystal structure as the $[MPn]$ layer could host multi-

ple topological states [36]. Hoffmann *et al.* theoretically pointed out that Pn atoms in $[MPn]$ layer are also arranged in the square net fashion, potentially showing the characteristic square-net bands [14, 15]. Moreover, it has been theoretically demonstrated that the Peierls-like distortions could significantly modify the square net derived band structure [14]. The possible distortion of the square net mainly includes the zigzag chains and cis-trans chains. Zigzag chains have been often observed among $AMnSb_2$ ($A = Sr, Ba, Eu$) compounds, showing a similar band structure with the undistorted square nets [37]. However, experimental validation is still lacking for the cis-trans distortion due to the lack of suitable materials. Therefore, it is of great interest to systematically study the topological properties of buffer layers and the impact of cis-trans distortion.

Recently, ternary rare-earth coinage metal antimonide $LaTSb_2$ ($T = Cu, Ag, \text{ and } Au$) has gotten renewed interest due to the coexistence of charge density wave (CDW), Dirac fermion, and superconductivity [21, 22]. $LaAgAs_2$ shares a similar crystal structure with $LaTSb_2$ but with the planar layer distorted from the Sb square net into As cis-trans chains [38], which is expected to show similar physical properties. However, previous studies mainly focus on the crystal structure [38–40], with the electronic structure and physical properties remaining elusive, possibly due to the lack of high-quality single crystals.

In this study, we successfully synthesized high-quality $LaAgAs_2$ single crystals using the self-flux method. Through comprehensive crystal and electronic structure measurements, our experimental findings reveal that rather than being a square-net-based topological semimetal with symmetry-induced band inversion between As1 p_x/p_y bands, $LaAgAs_2$ is identified as a topological material with multiple topological states.

RESULTS

Sample characterization

The single-crystal XRD measurements revealed that $LaAgAs_2$ crystallizes in an orthorhombic crystal structure (space group: $Pbcm$) with lattice parameters $a = 5.8350(1)$ Å, $b = 21.2926(4)$ Å, and $c = 5.8306(1)$ Å, consistent with those reported by Rutzinger *et al.* [38]. The obtained crystal structure is shown in Fig. 1(a-c) and the refined parameters are listed in Table I. The crystal structure of $LaAgAs_2$ [Fig. 1(a)] consists of alternatively

stacking the anti-PbO type [AgAs] layer [Fig. 1(b)] and As1 cis-trans chains [Fig. 1(c)] along the b axis, interspersed with La layers. The orthorhombic distortion primarily affects the planar As1 layer, which deforms from a square-net configuration into cis-trans chains. In contrast, the La and [AgAs] layers retain a pseudo-tetragonal structure with an a/c ratio of 1.00075.

The out-of-plane x-ray diffraction pattern shown in Fig. 1(d) confirms the pure phase of the crystals with a preferred [010] orientation. The narrow peak width [inset of Fig. 1(d)], along with the bright and sharp Laue diffraction spots [inset of Fig. 1(e)], demonstrates the excellent crystallinity nature of the LaAgAs₂ crystals. Laue diffraction exhibits a four-fold symmetry, attributable to the combination of pseudo-tetragonal symmetry and the twinning effect, which will be discussed in detail in the following sections. Figure 1(e) shows the shallow core levels, indicating that only the constituent elements (La, Ag, and As) were present, with no additional peaks. These results suggest that our LaAgAs₂ crystals are phase pure and of high quality.

As shown in Fig. 1(f), both the in-plane resistivity (ρ_{xx}) and out-of-plane resistivity (ρ_{zz}) of LaAgSb₂ exhibit a typical metallic behavior across the entire temperature range, in contrast with the semiconducting behavior reported by Rutzinger *et al.* [38]. This difference might be attributable to the influence of the grain boundaries in polycrystal pellets. The residual resistivity ratios (RRR) for both ρ_{xx} and ρ_{zz} is about 5. The room-temperature resistivity values are determined to be $\rho_{xx}(300\text{ K}) = 68\ \mu\Omega\text{ cm}$ and $\rho_{zz}(300\text{ K}) = 4.86\text{ m}\Omega\text{ cm}$, yielding a large resistive anisotropy ratio of $\rho_{zz}/\rho_{xx} \sim 70$. This value is approximately an order of magnitude larger than that in LaAuSb₂ [21], reflecting the strong 2D character for the underlying electronic structure in LaAgAs₂.

As shown in Fig. 1(g), the Hall resistivity $\rho_{xy}(B)$ exhibits an approximately linear behavior with a positive slope up to 14 T. A subtle change in slope around $B = 4\text{ T}$ can be recognized for temperatures below 100 K, indicative of multiband behaviors dominated by hole-type carriers. Due to the nearly linear characteristics of the $\rho_{xy}(B)$ curves, accurate fitting using a two-band model is challenging. Consequently, the carrier concentration is estimated by single-band linear fitting of the high-field region of the $\rho_{xy}(B)$ curves [41]. The resulting carrier concentration, presented in the inset of Fig. 1(g), gradually decreases with decreasing temperature. The carrier concentration at 300 K is estimated to be $n = 1.57 \times 10^{21}\text{ cm}^{-3}$, equivalent to 0.15 holes per formula unit (f.u.), which is comparable to

that in LaAgSb₂ [30].

Quantum oscillations

As one of the most powerful experimental techniques that can probe the band topology, quantum oscillation has been widely adopted to study topological materials [42, 43]. In high magnetic fields, the quantization of energy levels into discrete Landau levels leads to periodic oscillations in physical quantities as a function of $1/B$ [44]. These quantities include magnetization and resistivity, corresponding to de Haas-van Alphen (dHvA) oscillation and Shubnikov-de Haas (SdH) oscillation, respectively. The quantum oscillation signal of LaAgAs₂ is probed by both dHvA and SdH oscillation measurements, with the results presented in Fig. 2.

Shown in Fig. 2(a) is the isothermal out-of-plane ($B \parallel b$) magnetization measured at various temperatures for a LaAgAs₂ single crystal. Clear oscillation signals are observed on the paramagnetic/diamagnetic background for $B > 5$ T. The oscillatory components ΔM , extracted by subtracting a smooth polynomial background, are plotted as a function of $1/B$ in Fig. 2(b), where a subtle beat pattern can be identified. As shown in Fig. 2(c), fast Fourier transformation (FFT) analyses of ΔM reveal two oscillation frequencies, i.e., $F_\alpha = 94$ T and $F_\beta = 158$ T. Figure 2(e) shows the magnetic field dependence of MR, defined as $\text{MR} = [\rho(B) - \rho(0)]/\rho(0) \times 100\%$. The MR exhibits a power-law dependence on B ($\text{MR} \propto B^n$, $n = 1.45 \sim 1.6$), reaching a maximum value of 58% at 2 K and 14 T. SdH oscillation signal superimposed on the MR curves can be seen for $B > 7$ T, where the oscillating magnitude is weaker than that in magnetization. After the subtraction of a smooth polynomial background, the FFT analysis of $\Delta\rho$ in Fig. 2(f) gives rise to the FFT spectrum displayed in Fig. 2(g), revealing two oscillation peaks with nearly identical frequencies to those in the dHvA spectrum [Fig. 2(c)]. However, from the FFT spectra shown in Figs. 2(c) and 2(g), it can be seen that the relative amplitude between F_α and F_β is reversed between dHvA and SdH oscillations, which is a common phenomenon and is usually interpreted in terms of different scattering mechanisms [45, 46]. Further details for the comparison of the quantum oscillation in M , ρ_{xx} , and ρ_{xy} can be found in Supplementary Fig. S1 [47].

The geometry of the Fermi surface can be inferred from Quantum oscillation measure-

ments via the Onsager relation [44]: $F = (\frac{\Phi_0}{2\pi^2}) A_F$, where $\Phi_0 = 2.07 \times 10^{-15} \text{ Tm}^2$ is the magnetic flux quantum, and A_F is the cross-sectional area of the Fermi surface normal to the magnetic field. The cross-sectional area of the Fermi surfaces associated with F_α and F_β are calculated to be $A_{F,\alpha} = 0.90 \text{ nm}^{-2}$ and $A_{F,\beta} = 1.51 \text{ nm}^{-2}$, respectively, corresponding to 0.8% and 1.3% of the total area of the ac plane in the Brillouin zone. From the inset in Fig. 2(g), it can be seen that both F_α and F_β show a $\sim 1/\cos(\theta)$ dependence on the orientation of the magnetic field, which is a characteristic of the 2D Fermi surface. These results indicate that F_α and F_β originate from quasi-2D Fermi pockets with small sizes.

The effective mass $m = m^*m_e$ is related to the band dispersion around the Fermi level (E_F) via $m^* = \hbar^2 / [\partial^2 E(k) / \partial k^2]$, which can be extracted by fitting the temperature dependence of the oscillation amplitudes to the thermal damping factor R_T . According to the Lifshitz-Kosevich model, R_T is defined as [44]:

$$R_T = \frac{\alpha T m^* / \bar{B}}{\sinh(\alpha T m^* / \bar{B})} \quad (1)$$

where $\alpha = 2\pi^2 k_B m_e / e\hbar = 14.69 \text{ T/K}$ is a constant, $\bar{B} = 1 / [(1/B_{\max} + 1/B_{\min}) / 2]$ is the average inverse field used in the FFT analysis. As shown in Fig. 2(d), the FFT amplitudes vs temperature curves can be well-fitted by R_T , yielding effective masses of $m_\alpha = 0.094m_e$ and $m_\beta = 0.21m_e$. These effective masses are comparable to those in sister compounds such as $REAgSb_2$ [(0.07 ~ 0.5) m_e] [48], and $AMnSb_2$ [(0.05 ~ 0.1) m_e] [35, 49], as well as La_3ScBi_5 ($\sim 0.2 m_e$) [50], where RE and A represents the rare-earth and alkaline-earth elements, respectively. The small value of effective masses in $LaAgAs_2$ indicates strong band dispersions for both the F_α and F_β pockets, possibly linear Dirac bands as those reported in sister $AMPn_2$ compounds [21].

The general behavior of quantum oscillations for $LaAgAs_2$ is similar to the well-established square-net-based topological semimetals such as $LaAgSb_2$, $LaAuSb_2$, $SrMnSb_2$, and $EuMnSb_2$ [21, 41, 48, 49], indicating a similar band structure and topological properties at first glance. However, the detailed band structure and the origin of F_α and F_β remain elusive. Therefore, the electronic structure of $LaAgAs_2$ is further studied by ARPES, a technique that maps the band structure directly.

ARPES

Figure 3 shows the in-plane Fermi surface (a) and the band dispersions along the BZ diagonal (b), taken with $h\nu = 70$ eV photons. The photon-energy dependence that gives access to the out-of-plane electronic structure is shown in panels (c) and (d). The set of spectra along the zone diagonal, $\bar{\Gamma} - \bar{M} - \bar{\Gamma}$, measured at photon energies ranging from 55 to 90 eV is converted to k_z using the free electron approximation for the final photoemission state, with $V_0 = 10$ eV for the "inner" potential. Panel (c) shows the intensity at the Fermi level (Fermi surface), while panel (d) shows intensity at $E = -0.6$ eV. Almost total absence of dispersion in (c) indicates a highly 2D character of the states forming the Fermi surface, whereas weakly dispersing contours in (d) indicate that deeper states have a certain 3D character.

In Fig. 4 we show the more detailed in-plane electronic structure, recorded at $h\nu = 100$ eV. Panel (a) shows the Fermi surface from the spot on the sample that has two orthogonal orthorhombic domains contributing nearly equally to the ARPES intensity. In panel (b), recorded only ~ 20 μm from the spot probed in (a), one domain clearly dominates. Panel (c) shows the spectrum from the $k_y = 0$ ($\bar{\Gamma} - \bar{Y}$) line of the surface BZ of the twinned spot (a), while panels (d-f) show the spectra along the three different momentum lines, as indicated, recorded from the single-domain spot (b).

There are several important findings that can be immediately deduced from Fig. 4. First, consistent with our XRD results, it is obvious that the crystals of LaAgAs_2 are twinned. The domains are large enough that they can be completely resolved in ARPES at both beamlines, indicating that they are of the order of tens of microns across. Obviously, the twinning will affect the macroscopic transport properties measured on the twinned crystals and remove any anisotropy related to the quasi-1D character of the electron pockets visible in Fig. 4(b). Indeed, as shown in Supplementary Fig. S3 [47], with the magnetic field being rotated in the ac plane, the angular dependence of MR exhibits a four-fold symmetry instead of two-fold symmetry as would be expected for the quasi-1D electron pockets.

Therefore, the states at the Fermi level, that determine transport properties, are either quasi-2D (holes) or quasi-1D (electrons), with almost no out-of-plane dispersion, in good agreement with the large ρ_{zz}/ρ_{xx} ratio and the quantum oscillation measurements [inset of Fig. 2(g)]. The Γ -centered hole states disperse almost linearly within the planes, with

relatively high Fermi velocities, indicating a very light character in good agreement with magnetotransport. This, and the absence of significant broadening with energy or temperature, should be reflected in very high hole mobilities.

Our high-resolution measurements show that the larger contour actually consists of two very close pockets, as can be seen in Fig. 5(a-c) (additional details of both the hole and electron pockets can be seen in Supplementary Fig. S8 [47]). In the inner, circle-like shaped contour, the two states predicted by calculations cannot be resolved. The area of the Fermi surface, which gives the concentration of carriers through the Luttinger count, can be directly measured from the positions of momentum distribution curves (MDC) peaks, k_F [26, 51]. The inner and outer hole contours areas recorded at $h\nu = 100$ eV are $A_i = 2.3$ nm⁻² and $A_o = 8.6$ nm⁻², respectively. If recorded at $h\nu = 70$ eV [Fig. 5(d)], both hole contours enclose slightly smaller areas, $A_i = 1.6$ nm⁻² and $A_o = 7.3$ nm⁻², respectively, indicating a small, but finite k_z warping, in excellent agreement with the DFT calculations. The smaller one, A_i , is very close to the larger orbit F_β in the dHvA oscillations. The estimated Fermi velocities of the inner and outer hole bands along the $\bar{\Gamma} - \bar{X}$ and $\bar{\Gamma} - \bar{Y}$ lines, determined from MDC derived dispersions, are almost the same, $v_F \approx 2.5$ eVÅ (3.8×10^5 ms⁻¹). The only exception is one of the states forming the outer doublet that disperses somewhat faster: $v_F \approx 3.6$ eVÅ (5.5×10^5 ms⁻¹). In the $\bar{\Gamma} - \bar{M}$ direction all holes have nearly the same Fermi velocity, $v_F \approx 3.3$ eVÅ (5×10^5 ms⁻¹). Taking all the hole pockets into account, and their quasi-2D character, we can estimate the concentration to be ≈ 0.34 holes/f.u.

The electron pockets around \bar{X} points are the only indication of a structurally quasi-1D character of the crystal, originating from the cis-trans chains of As1 atoms inside the As1 planar planes. These pockets show more conventional parabolic dispersion. Their Fermi surface encloses an area $A_{el} = 5.3$ nm⁻², somewhat smaller than in DFT calculations. Further disagreement with the DFT is visible at $h\nu = 70$ eV: a very small electron-like band is just touching the Fermi level, enclosing a tiny electron pocket inside the big one [Fig. 5(d,e)]. This state is not present in the DFT calculations. Its area cannot be precisely determined in ARPES, but it could range from 0.5 to 1 nm⁻².

The Fermi velocity of the main electron pocket is $v_F \approx 4$ eVÅ (6.1×10^5 ms⁻¹) and $v_F \approx 1.9$ eVÅ (2.9×10^5 ms⁻¹) along the $\bar{\Gamma} - \bar{X}$ and perpendicular to it, respectively. Corresponding effective masses are $0.07 m_0$ and $0.53 m_0$, respectively. The electron concentration from this pocket is estimated to be 0.18 electrons per site, by assuming that it too is a doublet and

that it is perfectly 2D. The resulting excess hole concentration is then 0.16 per site, in perfect agreement with the Hall coefficient [Fig. 1(g)].

Further, there is a peculiar intensity modulation of photoelectron intensity near the Fermi level measured at different BZs: strong intensity from the 1st BZ, almost vanishing in the 2nd and again strong in the 3rd (Fig.3(a)). The diminishing intensity in the 2nd BZ points to some sort of destructive interference, probably related to the cis-trans structure motif of the As1 chains. Also, the states further away from the Fermi level, $E \leq -0.6$ eV, shown in Fig. 4(c-f), do not show the same periodicity as the As1-derived states forming the Fermi surface. These deeper states are mainly the [AgAs]-derived states that keep the original tetragonal symmetry and due to the weak coupling with the As1 layers, do not repeat in the orthorhombic BZ.

Finally, we note that according to the crystal structure and calculated exfoliation energies for possible cleavage surfaces [see Supplementary Fig. S4 [47]], there should be at least two different terminations when the crystal is cleaved. Due to the very shallow probing depth, ARPES should be sensitive to that. Our calculations show that the La–As1 interface has the lowest exfoliation energy of the three possible interfaces within the crystal. This suggests that the single crystal preferentially exposes the La and the As1 atomic layers upon cleaving. In reality, the actual surface will likely be a mixture of these two terminations [52]. The fact that ARPES could not distinct between different terminations would indicate that 1) the lateral size of each termination domain is smaller than our spot size, or 2) that the spectroscopic difference between the terminations is not significant, or 3) some reconstruction occurs, leading to a surface that is crystallographically and chemically distinct from bulk, but uniform across the surface.

Electronic structure calculations

DFT calculations were performed to understand the electronic structure experimentally detected by quantum oscillation and ARPES measurements. From the density of states (DOS) shown in Fig. 6(a), it is evident that the Fermi level of LaAgAs₂ is dominated by the states of La and As(2,3), differing from the square-net structured Pn layer in $AMPn_2$ materials [21, 30, 35, 53]. Further in-depth DOS analysis can be seen in Supplementary Figs. S5 and S6 [47]. This result implies that cis-trans distortion significantly reduces the

square-net-derived states. Consequently, the states from the buffer layers, i.e., As(2,3) and La, should play a more important role in the physical properties of LaAgAs₂.

Figure 6(b) shows the calculated band structure for LaAgAs₂. It can be seen that the bands cross E_F at Γ point, X point, and the S–Y direction, leading to a Fermi surface as shown in Fig. 6(f). The Fermi surface consists of two 2D hole pockets at the Γ point, a quasi-1D electronic pocket at the X point, and two tiny 3D electronic pockets along the S–Y direction. Further details for each pocket are displayed in Supplementary Fig. S7 [47].

The calculated electronic structure of LaAgAs₂ is in good agreement with that detected by the ARPES measurements except for the size of the 1D electron pocket at X point, an additional tiny pocket inside the 1D electron pocket, and the absence of 3D electron pockets along the S–Y direction. As the electron pockets are derived from the As1-orbitals within the cis-trans chains, they are very sensitive to the structure of these chains and that could be the origin of all discrepancies between the calculated and measured Fermi surfaces.

To uncover the impact of cis-trans distortion on the electronic structure, we also calculate the electronic structure of the hypothetical tetragonal LaAgAs₂, with cis-trans chains artificially arranged to a checkerboard-like square net [Fig. 6(g)]. Because the unit cell of LaAgAs₂ is related to the tetragonal HfCuSi₂-type structure by $\sqrt{2}a_T \times \sqrt{2}a_T \times 2c_T$, the electronic structure of LaAgAs₂ is folded as compared to that of $AMPn_2$, where the relationship between the folded and unfolded Brillouin zone is illustrated in Fig. 6(e). Both the folded [Figs. 6(c) and 6(g)] and unfolded [Figs. 6(d) and 6(h)] versions for the electronic structure of the hypothetical tetragonal LaAgAs₂ are shown to facilitate the comparison with the orthorhombic LaAgAs₂ and typical $AMPn_2$, respectively. From Figs. 6(b) and 6(c), it can be seen that the orthorhombic structure distortion mainly modifies the bands around the zone boundary with the As1 character, with the hole bands around the zone center remaining intact, in good agreement with the ARPES measurements. Specifically, cis-trans distortion changes the 2D linear Dirac bands into quasi-1D parabolic bands, consistent with the theoretical prediction by Tremel *et. al.* [14].

The transport properties of LaAgAs₂ are similar to those in $AMPn_2$ compounds, which are dominated by the Pn square net-derived Dirac bands. However, from the above analysis, the square net-derived bands initially expected to account for the quantum transport phenomena are transformed into quasi-1D trivial bands by the cis-trans distortion. Therefore, bands around the Γ point with primarily As(2,3) orbital character should be responsible

for the observed quantum transport phenomena. Indeed, the F_β with a higher frequency can be assigned to the inner hole pockets around the Γ point. However, no Fermi pocket responsible for the lower frequency F_α can be identified at present.

From Figs. 6(b-d), it can be seen that the linear conduction bands nearly touch the valence bands at the Γ point as observed in Heusler topological insulators [54, 55], indicating the proximity of a topological quantum transition. Upon a close inspection of the band structure without considering SOC at the Γ point [Fig. 7(a)], the valence and conduction bands are separated by a tiny gap about ~ 0.6 meV, which is much smaller than the strength of the SOC. Since the system possesses inversion symmetry, we can calculate the parity values of the occupied states at the eight time-reversal invariant momenta (TRIM) points according to the Fu-Kane formula [56] (see Supplementary Table S1 for details [47]). The results indicate that the system exhibits Z_2 topologically non-trivial characteristics when it is fully occupied by valence electrons (256 valence electrons). Therefore, it can be concluded that the SOC induces a band inversion between the valence and conduction bands and leads to a topological surface state (TSS) with spin-momentum locked Dirac-cone located in the gap between the valence and conduction bands [Fig. 7(b)] [57–61]. We further notice that two valence bands cross around E_F , resulting in a bulk 3D Dirac cone [Fig. 7(b)].

Therefore, both the TSS and topological Dirac semimetal (TDS) states coexist in LaAgAs_2 , as illustrated in Fig. 7(b). The multiple topological states observed in LaAgAs_2 are similar to those in $\text{LiFe}_{1-x}\text{Co}_x\text{As}$, $\text{FeTe}_{1-x}\text{Se}_x$ [36], where both the TSS and TDS are identified around the Γ point [36]. These results were also cross-validated through the application of topological quantum chemistry methods, validating the predictive capabilities of topological quantum chemistry theory in the discovery of topological materials [5, 62, 63].

Because both the TSS and TDS states are located well above the Fermi level, they cannot be captured by ARPES measurements (see the ARPES section). Furthermore, due to the highly delocalized nature of the La's d orbitals and As1's p orbitals, generating well-localized Wannier orbitals for subsequent surface state calculations via the surface Green's function method [64, 65] proved challenging. The quantum oscillation measurements indicate that the F_α bands show the characteristics expected by a nontrivial topological state, i.e., the quasi-2D Fermi surface with small size and small effective mass. Since the TDS cone is located closer to E_F than the TSS cone, F_α likely originates from the TDS cone.

DISCUSSION

Our calculations have clearly demonstrated that LaAgAs_2 is a topological material with multiple topological states, similar to iron-based superconductors, but differs significantly from $AMPn_2$ sister compounds. Our magnetotransport experiments also suggest that the F_α oscillation originates from nearly massless carriers, most likely from the bulk TDS cone. These results can be well understood from the perspective of chemical bonds.

From Fig. 7(a-b) and orbital-resolved DOS shown in Supplementary Figs. S5 and S6 [47], it can be seen that it is As(2,3)- p_x/p_y , La- d_{xy} , and Ag- d_{xz}/d_{yz} that dominate the dispersive bands around Γ , indicative of primarily in-plane bonding interactions. This result differs from the interlayer bonding in iron-based superconductors [36, 66], suggesting a distinct origin of the multiple topological states.

As illustrated in Fig. 7(c), the As(2,3) atoms, with four As(2,3) neighbors in the same layer, are sandwiched by four La(1,2) and four Ag(1,2) atoms. Notice that we do not distinguish the detailed atomic position of La(1,2), Ag(1,2), and As(2,3) atoms due to the pseudo-tetragonal nature of the La and [AgAs] layers. The distance between As(2,3) atoms is $\sim 4.12 \text{ \AA}$, too long to form the square-net-derived bands as predicted by Hoffmann *et al.* [15]. The interatomic distance of As(2,3)-La is $\sim 0.95 \text{ \AA}$, much shorter than $\sim 1.85 \text{ \AA}$ between As(2,3) and Ag layers, indicating that La atoms play a more important role in the in-plane bonding interactions, which is corroborated by the higher orbital weight of La in the valence and conduction bands [Supplementary Figs. S5 and S6 [47]]. This result suggests that the puckered [LaAs(2,3)] layer with an in-plane checkerboard configuration can be treated as an entirety, i.e., a building block [Fig. 7(d)]. Therefore, we replot the crystal structure of LaAgAs_2 in Fig. 7(e), emphasizing the important role played by the [LaAs] layer in the topological electronic structure. Due to their small electronegativity, rare earth elements are typically regarded as spacer atoms that donate electrons to the system, as reported in iron-based superconductors [67, 68]. Our results indicate that the structural motif can be rearranged in specific materials and identify a new structural motif, [LaAs], capable of hosting multiple topological states. Since the puckered [RE Pn] layer is a common structural motif among the square-net-based structures, such as HfCuSi_2 , ThCr_2Si_2 , PbFCl , and ZrCuSiAs , all of which exhibit great structural diversity [10, 69, 70]. Thus, our results can serve as a guideline for discovering new topological materials.

Finally, we want to point out to an interesting possibility that the extremely small orthorhombicity in LaAgAs_2 and its sister compounds, of the order of $\sim 5 \times 10^{-4}$, could make these materials potentially very sensitive to the uniaxial strain [71, 72]. A routinely achievable strain of $\sim 1\%$, 1 – 2 orders of magnitude larger than the orthorhombicity, might not only re-establish tetragonal order, but it might be able to drive the system to an uncharted territory with easily tunable electronic structure, where the topological character could be turned on and off on demand.

METHODS

Single crystal growth. Single crystals of LaAgAs_2 were grown by the self-flux method using excess Ag and As as flux [73]. The starting materials of La chunks, Ag grains, and As lumps were weighted according to the ratio of $\text{LaAg}_{36}\text{As}_{17}$, which were mixed and loaded into an alumina crucible. The crucible was sealed in an evacuated quartz tube and then slowly heated to 970°C in a box furnace. After dwelling at 970°C for 10 h, the sample was cooled to 750°C at a rate of $2^\circ\text{C}/\text{h}$ to grow single crystals. Shiny centimeter-sized single crystals with a typical size of $10 \times 5 \times 0.5 \text{ mm}^3$ can be obtained by decanting the excess flux using a centrifuge.

Structure characterizations. Single-crystal XRD measurements were performed on a high-flux, high-resolution, rotating anode Rigaku Synergy-DW (Mo/Ag) diffractometer using Mo K_α radiation ($\lambda = 0.7107 \text{ \AA}$). The system is equipped with a background-less Hypix-Arc150° detector, which guarantees minimal reflection profile distortion and ensures uniform detection conditions for all reflections. All samples were measured to a resolution better than 0.5 \AA and with the beam divergence set to 5 mrad . The samples exhibited no mosaic spread and no additional reflections from secondary phases, highlighting their high quality and allowing for excellent evaluation using the latest version of the CrysAlisPro software package [74]. The crystal structure of the system was refined using JANA2006 [75]. Out-of-plane x-ray diffraction patterns were collected using a PANalytical powder diffractometer (Cu K_α radiation). Laue diffraction patterns were taken in a backscattering geometry with white incident light exposed from the out-of-plane direction.

Magnetization and transport measurements. Both the magnetization and magnetotransport measurements were performed in a Quantum Design DynaCool Physical Prop-

erties Measurement System (PPMS-14T). The VSM option was used for magnetization measurements, and the DC delta mode with a Keithley 6221 current source and a Keithley 2182A nanovoltmeter was used to record the transport data. In-plane electrical resistivity ρ_{xx} and Hall resistivity ρ_{xy} were measured simultaneously on a polished single crystal with the six-probe configuration. To eliminate the mixture of resistivity and Hall signals arising from the geometrical factor of the electrical contacts, the ρ_{xx} and ρ_{xy} data shown in this paper were corrected according to the following two formulas: $\rho_{xx}(B) = [\rho_{xx}(+B) + \rho_{xx}(-B)] / 2$ and $\rho_{xy}(B) = [\rho_{xy}(+B) - \rho_{xy}(-B)] / 2$. The out-of-plane resistivity ρ_{zz} was measured using a standard four-probe method, where the sample was prepared by cutting a thick single crystal into a needle-shaped bar with the long axis along the crystalline b axis (out-of-plane direction).

ARPES measurements. The ARPES experiments were performed at the Electron Spectro-Microscopy (ESM) 21-ID-1 beamline of the National Synchrotron Light Source II and at BL-10.0.1.2 of the Advanced Light Source. Both beamlines are equipped with a Scienta DA30 electron analyzer, with base pressure $\sim 2 \times 10^{-11}$ mbar. The total energy resolution was ~ 10 and ~ 20 meV at ESM and BL-10.0.1.2, respectively. The photon spot size on the sample is estimated to be $\sim 2 - 3 \mu\text{m}$ and $\sim 50 \mu\text{m}$ at ESM and BL-10.0.1.2, respectively. The angular resolution was $\sim 0.1^\circ$, and $\sim 0.3^\circ$ along the slit and perpendicular to it, respectively, in both facilities. The polarization of the light was always linear and horizontal.

First-principles calculations. First-principles calculations were conducted using the Vienna *ab-initio* Simulation Package (VASP) within the framework of density functional theory (DFT) [76]. The exchange-correlation interactions were described using the Perdew-Burke-Ernzerhof (PBE) functional under the generalized gradient approximation (GGA) [77]. Projector-augmented-wave (PAW) pseudopotentials were employed with a plane-wave basis set energy cutoff of 400 eV [78, 79]. The calculations used an orthorhombic crystal structure based on our experimental lattice parameters of $a_o = 5.8350(1) \text{ \AA}$, $b_o = 21.2926(4) \text{ \AA}$, and $c_o = 5.8306(1) \text{ \AA}$. For structural optimization, all atomic positions were fully relaxed until the residual forces on each atom were less than 0.001 eV/\AA , with convergence achieved within an energy threshold of 10^{-5} eV . A Γ -centered Monkhorst-Pack k -point mesh of $9 \times 9 \times 2$ was utilized to sample the first Brillouin zone. The electronic band structure was computed with and without the inclusion of spin-orbital coupling (SOC) to capture

the relativistic effects. The Fermi surface at different energy levels was analyzed and refined using the iFermi software [80]. The hypothetical tetragonal LaAgAs₂ was calculated with the same method as the orthorhombic one. The tetragonal structure is obtained by artificially setting $a = b = a_o$ and $c = b_o$, and arranging the cis-trans chains into the square net fashion. Parity analysis is conducted using the irvsp code [81].

Data availability: The datasets generated and/or analyzed during the current study are not publicly available due to legal restrictions preventing unrestricted public distribution, but are available from the corresponding authors upon reasonable request.

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Author Contributions: T.V. and A.W. conceived the project. Y.L. and A.W. grew the

single crystals. T.L., X.Y., Z.S., and H.F. performed the DFT calculations. N.M. and M.M. performed the single-crystal XRD measurements. A.V.F., A.K.K., T.Y., E.V., and T.V. performed ARPES measurements. Y.L., X.W., L.Z., M.H., Y.C., and X.Z. performed the transport measurements. M.M., T.V., and A.W. wrote the manuscript with inputs from all co-authors. All authors read and approved the final manuscript.

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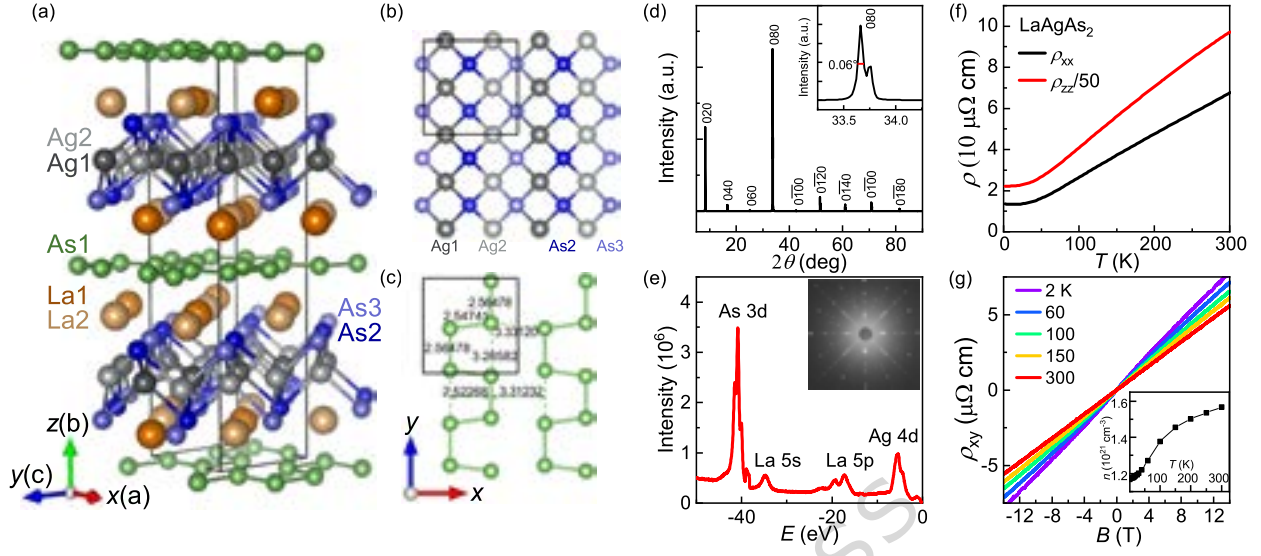


FIG. 1. Crystal structure and basic physical properties of LaAgAs_2 . (a) Crystal structure of LaAgAs_2 . To avoid confusion, the crystallographic direction for a , b , and c were denoted as x , z , and y , respectively. Top view of $[\text{AgAs}]$ layer (b) and As planar layer (c), where panels (b) and (c) use the same coordinate axis as shown in panel (a). Black lines in panels (a-c) indicate the unit cell of LaAgAs_2 . (d) XRD pattern for $(0\ k\ 0)$ surface of a flat LaAgAs_2 crystal. The insert shows the enlarged view of the $(0\ 8\ 0)$ reflection. (e) Core-level electronic structure of LaAgAs_2 , measured using a photon energy of 150 eV. The inset shows an x-ray Laue pattern of the $(H, 0, L)$ reciprocal plane. (f) Temperature dependence of in-plane resistivity ρ_{xx} ($j \parallel ac$) and out-of-plane resistivity ρ_{zz} ($j \parallel b$) measured with $B = 0$ T for LaAgAs_2 , where j is the electric current applied for the resistivity measurements. (g) Typical Hall resistivity $\rho_{xy}(B)$ curves measured at different temperatures with $B \parallel b$. The insert shows the temperature dependence of carrier concentration obtained from the single-band fitting of the high-field part of $\rho_{xy}(B)$ curves.

TABLE I. Crystallographic results of LaAgAs₂ as determined from single-crystal x-ray diffraction at 300, 200, and 80 K. For all temperatures, the structure was refined in the orthorhombic space group *Pbcm*. The lattice parameters, *a*, *b*, *c*, and volume *V* are shown together with the Wyckoff positions of the atoms, and the equivalent atomic displacement parameters U_{eq} . The ADPs were refined anisotropically, but due to space limitations, only the U_{eq} are listed in the Table. *TW* denotes the twinning ratio of the sample. The errors shown are statistical errors from the refinement.

Temperature		300 K	200 K	80 K
	SG	<i>Pbcm</i>	<i>Pbcm</i>	<i>Pbcm</i>
	<i>a</i> (Å)	5.8350(1)	5.8266(1)	5.8198(1)
	<i>b</i> (Å)	21.2926(4)	21.2585(5)	21.2284(5)
	<i>c</i> (Å)	5.8306(1)	5.8206(1)	5.8172(1)
	<i>V</i> (Å ³)	724.4	721.0	718.7
La1	Wyck.	4d	4d	4d
	<i>x</i>	0.01295(6)	0.01300(6)	0.01297(6)
	<i>y</i>	0.38575(2)	0.38571(2)	0.38567(2)
	<i>z</i>	1/4	1/4	1/4
	U_{eq}	0.00848(11)	0.00617(11)	0.00398(10)
La2	Wyck.	4d	4d	4d
	<i>x</i>	0.51428(6)	0.51439(6)	0.51434(6)
	<i>y</i>	0.11949(2)	0.11952(1)	0.11956(1)
	<i>z</i>	1/4	1/4	1/4
	U_{eq}	0.00780(11)	0.00567(11)	0.00388(10)
Ag1	Wyck.	4c	4c	4c
	<i>x</i>	0.26271(8)	0.26286(8)	0.26292(7)
	<i>y</i>	1/4	1/4	1/4
	<i>z</i>	0	0	0
	U_{eq}	0.01164(21)	0.00786(20)	0.00473(18)
Ag2	Wyck.	4c	4c	4c
	<i>x</i>	0.76594(8)	0.76612(8)	0.76601(8)
	<i>y</i>	1/4	1/4	1/4
	<i>z</i>	0	0	0
	U_{eq}	0.01226(20)	0.00889(20)	0.00501(18)
As1	Wyck.	8e	8e	8e
	<i>x</i>	0.28383(7)	0.28405(8)	0.28392(7)
	<i>y</i>	0.00118(3)	0.00116(3)	0.00119(3)
	<i>z</i>	0.03005(7)	0.03016(7)	0.03015(3)
	U_{eq}	0.01246(10)	0.00941(9)	0.00665(9)
As2	Wyck.	4d	4d	4d
	<i>x</i>	0.51406(9)	0.51411(10)	0.51411(10)
	<i>y</i>	0.33591(3)	0.33584(3)	0.33580(3)
	<i>z</i>	1/4	1/4	1/4
	U_{eq}	0.00892(18)	0.00636(19)	0.00417(18)
As3	Wyck.	4d	4d	4d
	<i>x</i>	0.01425(10)	0.01439(10)	0.01440(10)
	<i>y</i>	0.16336(3)	0.16346(3)	0.16354(3)
	<i>z</i>	1/4	1/4	1/4
	U_{eq}	0.00906(18)	0.00668(19)	0.00441(18)
	<i>TW</i> (%)	58/42	58/42	58/42
	wR ₂ (%)	6.64	6.66	6.62
	R ₁ (%)	3.09	3.13	3.19
	GOF	1.85	1.81	1.85

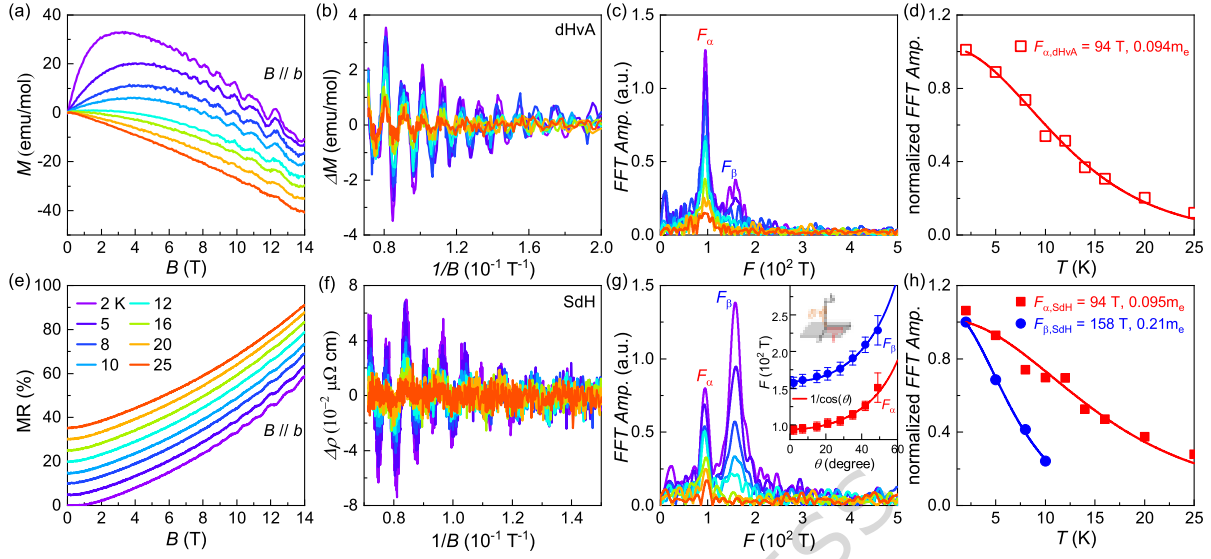


FIG. 2. Quantum oscillations in LaAgAs_2 . (a)(e) Magnetic field dependence of magnetization $[M(B)]$ and magnetoresistance $[\text{MR}(B)]$, respectively. Each subsequent MR curve is shifted upward by 4% for clarity. (b)(f) Inverse field dependence of oscillatory components of magnetization (ΔM , dHvA oscillations) and magnetoresistance ($\Delta\rho$, SdH oscillations), respectively. $\Delta M/\Delta\rho$ are obtained by subtracting the polynomial background from the data in (a)/(e), respectively. (c)(g) FFT spectra of dHvA/SdH oscillations at various temperatures. The inset of (g) shows the angular dependence of FFT peaks and the geometry of the measurements, where θ is defined as the angle between the magnetic field B and the crystallographic b -axis. Further details for the angular dependence of SdH oscillation can be found in Supplementary Fig. S2 [47]. (d)(h) Temperature dependence of the FFT amplitude for F_α and F_β , which are the FFT peaks inferred from (c) and (g), respectively. Solid lines represent the fits with the Lifshitz-Kosevich formula.

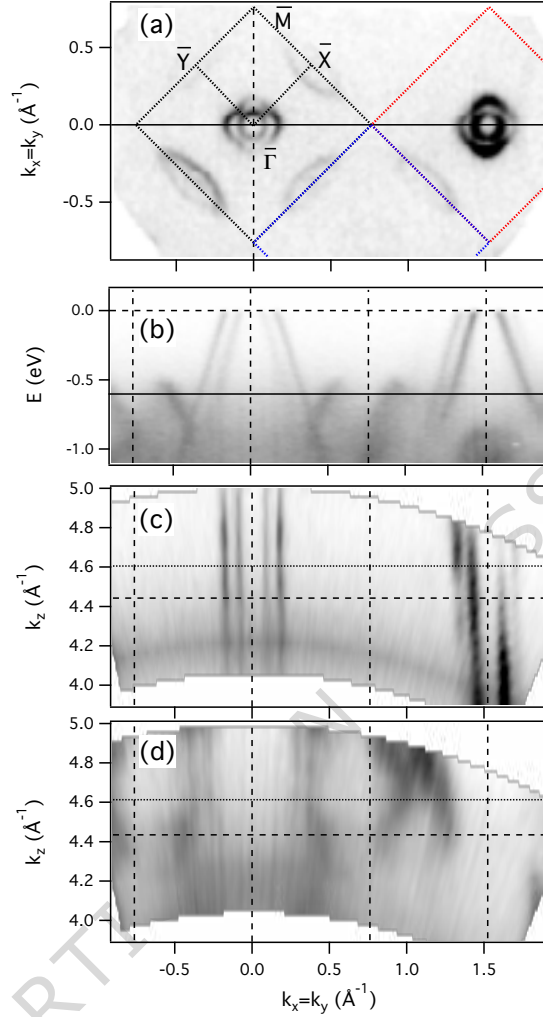


FIG. 3. Electronic structure of LaAgAs₂ from ARPES. (a) The Fermi surface is taken at $h\nu = 70$ eV. The black, blue, and red dotted squares represent the 1st, 2nd, and 3rd surface BZs, respectively, with the symmetry points indicated. (b) The $E(k)$ dispersion along the $\bar{\Gamma} - \bar{M}$ line of the surface BZ (solid horizontal line in (a)). (c) The k_z dependence of the states at the Fermi level ($E = 0$) along the same momentum line. (d) The same as in (b), but at $E = -0.6$ eV marked by the black solid line in (b). The maps in (c, d) are obtained by using the photon energies in the range from 55 to 90 eV and the free-electron approximation for the final electron state, $k_z = 1/\hbar\sqrt{2m_e(E_k\cos^2(\theta) + V_0)}$, where E_k is the kinetic energy of a photoelectron and $V_0 \sim 10$ eV is the inner potential. The dashed and dotted horizontal lines mark the Γ and Z planes in the 16th 3D BZ, respectively. All the spectra were taken at $T = 15$ K using the horizontally linearly polarized light.

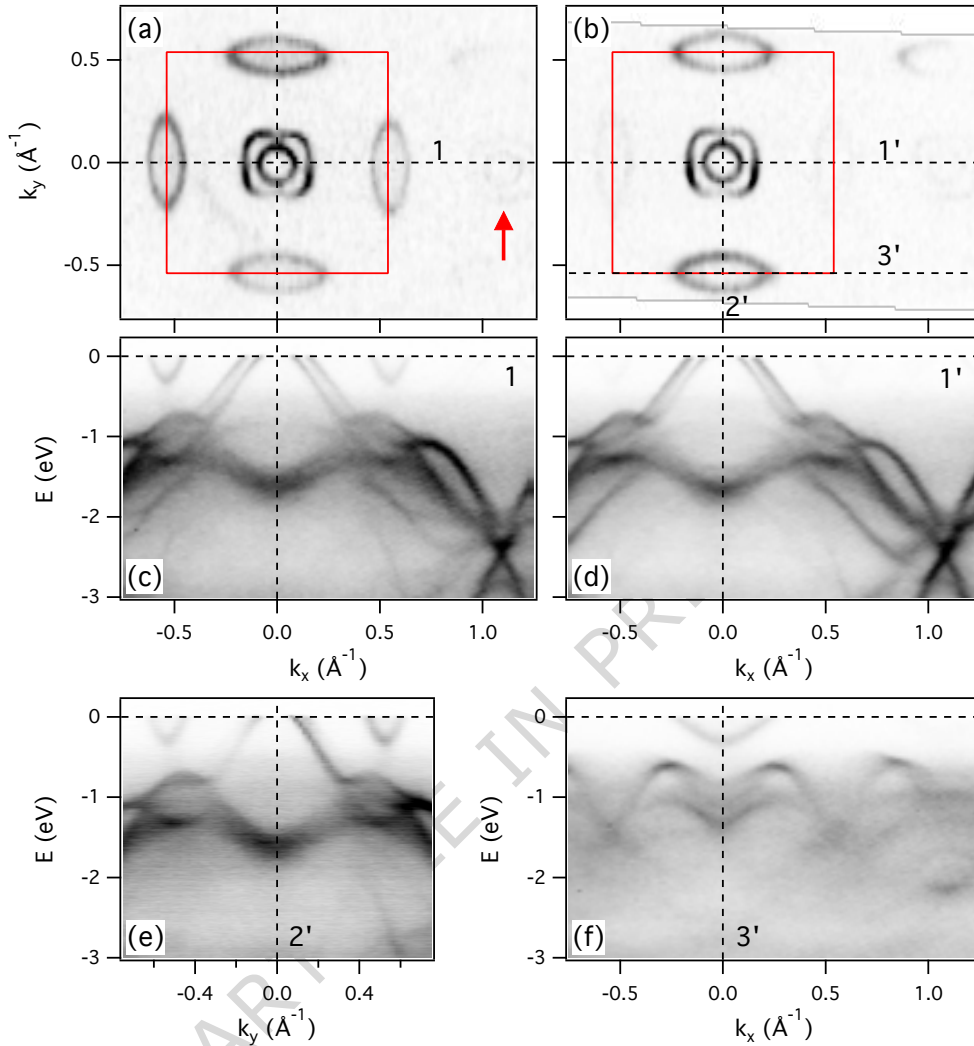


FIG. 4. In-plane electronic structure of LaAgAs_2 . (a) The Fermi surface from the spot with 2 orthogonal domains. (b) The Fermi surface from the spot with a single domain dominating. (c) Band structure along the $k_y = 0$ (marked as 1) line from (a). (d) The same as (b) (marked as 1'). (e) Band structure along the $k_x = 0$ (marked as 2') line from (b). (f) Band structure along the $k_y = \pi/c$ (marked as 3') line from (b). The red arrow points to the suppressed intensity of the 2nd zone states. The red square in (a,b) represents the 1st BZ. All the spectra were taken at $T = 15$ K using the horizontally linearly polarized light at 100 eV.

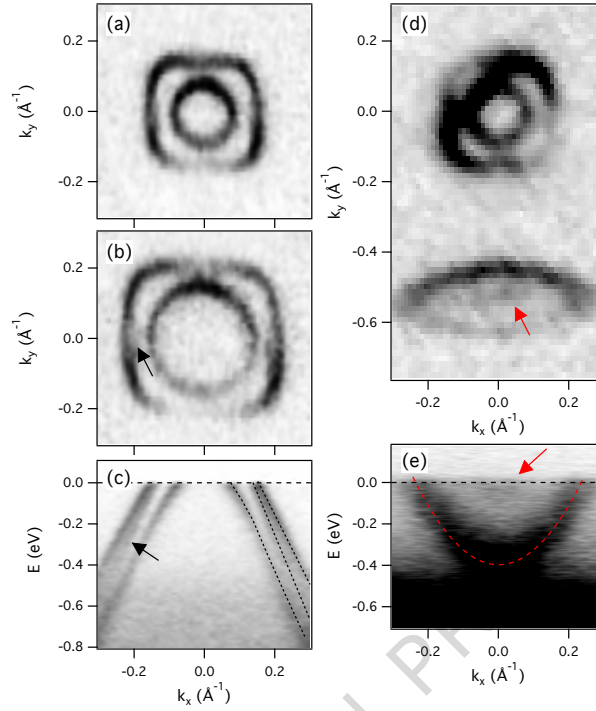


FIG. 5. High-resolution electronic structure of the Fermi pockets. (a) The Fermi surface and (b) the intensity at $E = -0.2$ eV taken at $h\nu = 100$ eV (c) Band structure along the $k_y = 0$ line from (a). The dotted lines at $k_x > 0$ indicate the three resolved hole bands. The black arrows point to the splitting of the outer hole doublet. (d) The Fermi surface taken at $h\nu = 70$ eV (e) Band structure along the $k_y = \pi/c$ line from (d). The red arrows indicate the small electron pocket inside the main one. The red dashed curve represents the parabolic fit to the MDC-derived dispersion.

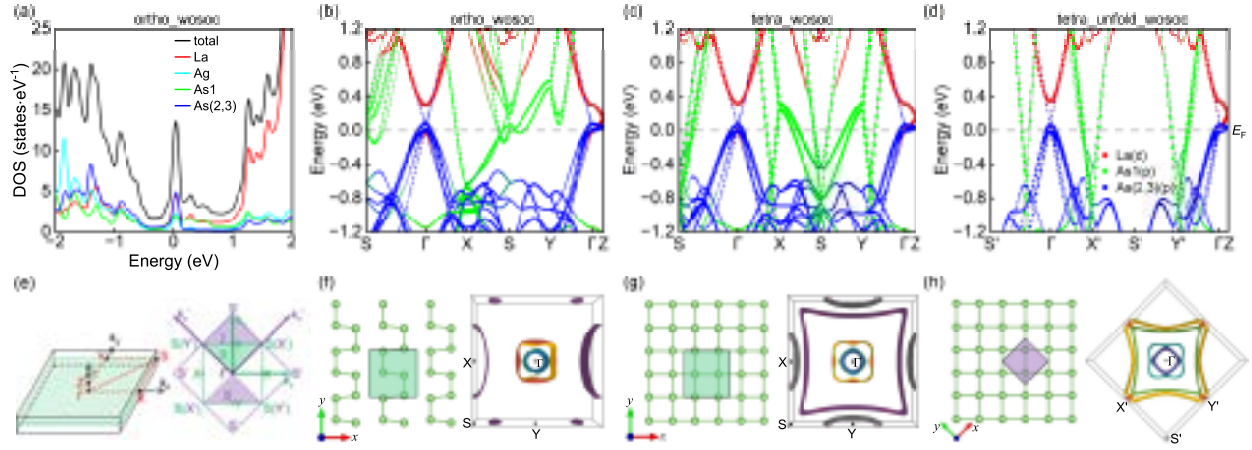


FIG. 6. The calculated electronic structure of LaAgAs_2 . (a) Atom-specific partial density of states (DOS). The orbital-projected band structure without spin-orbit coupling for LaAgAs_2 with pristine orthorhombic structure ($a_o \times b_o \times c_o$) (b), hypothetical tetragonal structure ($a_o \times a_o \times b_o$ or $\sqrt{2}a_T \times \sqrt{2}a_T \times 2c_T$) (c), and unfolded hypothetical tetragonal structure ($a_T \times a_T \times 2c_T$) (d), where $a_o = \sqrt{2}a_T$ and $b_o = 2c_T$, a_T and c_T refer to a HfCuSi_2 -type unit cell. The correspondence of the high symmetry points between the folded (green) and unfolded (purple) BZ is illustrated in (e). (f-h) display the top view of the crystal structure of the As1 layer (left) and the Fermi surfaces (right), where the green/purple shaded square on the left panel indicates the unit cell corresponding to the folded/unfolded BZ.

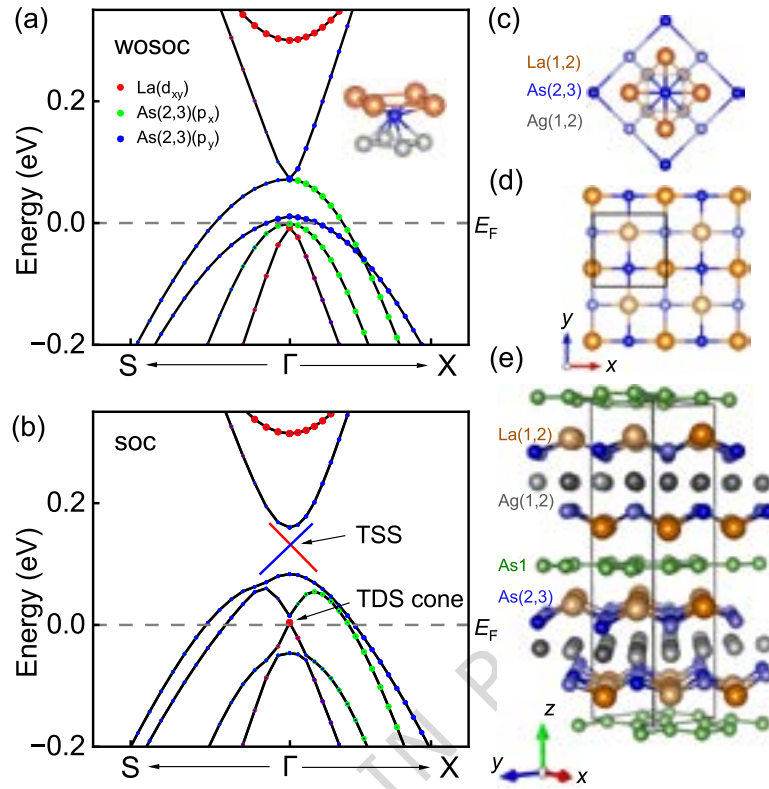


FIG. 7. The enlarged view of the calculated band structure around the Γ point. Panels (a) and (b) show the bands without and with considering spin-orbit coupling, respectively. The sketches of the spin-polarized topological surface state (TSS) and topological Dirac semimetal state (TDS) are marked in (b). (c) An As(2,3) atom with its nearest neighbors. (d) Top view of the puckered [LaAs(2,3)] layer, where the atoms are arranged in the checkboard square net fashion. The crystal structure of LaAgAs₂ is rearranged from Fig. 1(a) into panel (e), emphasizing the role of the [LaAs(2,3)] layer played in the topological electronic structure.