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Pressure-induced superconductivity and topological phase transitions in the topological nodal-line semimetal SrAs₃HPSTAR
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Topological nodal-line semimetals (TNLSMs) are materials whose conduction and valence bands cross each other, meeting a topologically protected closed loop rather than discrete points in the Brillouin zone (BZ). The anticipated properties for TNLSMs, include drumhead-like nearly flat surface states, unique Landau energy levels, special collective modes, long-range Coulomb interactions, or the possibility of realizing high-temperature superconductivity. Recently, SrAs₃ has been theoretically proposed and then experimentally confirmed to be a TNLSM. Here, we report high-pressure experiments on SrAs₃, identifying a Lifshitz transition below 1 GPa and a superconducting transition accompanied by a structural phase transition above 20 GPa. A topological crystalline insulator (TCI) state is revealed by means of density functional theory (DFT) calculations on the emergent high-pressure phase. As the counterpart of topological insulators, TCIs possess metallic boundary states protected by crystal symmetry, rather than time reversal. In consideration of topological surface states (TSSs) and helical spin texture observed in the high-pressure state of SrAs₃, the superconducting state may be induced in the surface states, and is most likely topologically nontrivial, making pressurized SrAs₃ a strong candidate for topological superconductor.

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INTRODUCTION

In recent years, topological semimetals including Dirac, Weyl, and nodal-line semimetals have been theoretically predicted and experimentally verified, opening a new field in condensed-matter physics in which novel properties and new applications can arise from spin-polarized states with unique band dispersion^{1–3}. Unlike the discrete points in momentum space in Dirac or Weyl semimetals^{2,3}, the band crossings in nodal-line semimetals can form closed loops inside the BZ⁴; a nodal chain consisting of several connected loops⁵; or an extended line traversing the entire BZ⁶. These one-dimensional nodal curves are topologically protected by certain discrete symmetries, for example mirror reflection, time-reversal, or spin-rotation symmetries^{2,3}. Upon breaking symmetries in a topological nodal-line semimetal (TNLSM), the nodal line is either fully gapped or gapped into several nodal points⁴. The nodal-line structure is expected to have several intriguing properties³, such as unique Landau energy levels⁷, special collective modes⁸, long-range Coulomb interactions⁹, or drumhead-like nearly flat surface states^{10,11}, which can be considered a higher-dimensional analog of the flat band on the zigzag edge of graphene³. These drumhead states may host interesting correlation effects, and even offer the possibility of realizing high-temperature superconductivity¹².

In the search for nodal-line semimetals, several systems have been theoretically proposed since 2011^{2,3}. However, only a few candidates including PbTaSe₂^{13,14}, ZrSiX (X = S, Se, Te)^{15,16}, CaAgX

(X = P, As)^{17,18}, and MB₂ (M = Ti, Zr)¹⁹ have been verified experimentally. More recently, the CaP₃ family of materials (MA₃, for M = Ca, Ba, and SrX₃ for X = P, As) was proposed as another potential host of TNLSMs²⁰. Among these compounds, only SrAs₃ shows a strongly topological nature at ambient pressure, while others need extra compressing²⁰. SrAs₃ displays semimetallic behavior with the hole carriers dominating^{21,22}. Previously, unusual galvanomagnetic properties and a first-order longitudinal Hall effect have been found in SrAs₃²³, and quantum oscillation experiments have been applied to map out the shape of the Fermi surface, finding two asymmetric, quasi-ellipsoidal Fermi-bodies as well as light cyclotron effective mass²². Recent magnetotransport measurements on SrAs₃ single crystals found a nontrivial Berry phase and a robust negative longitudinal magnetoresistance (MR) induced by the chiral anomaly, which indicates the presence of topological properties in SrAs₃^{24,25}. Subsequently, Song et al.²⁶ observed the complete nodal-line feature around the Y point by means of angle-resolved photoemission spectroscopy, demonstrating the existence of Dirac nodal-line fermions. In contrast to most TNLSMs, the nodal-line structure in SrAs₃ does not coexist with complex topologically trivial Fermi surfaces, which may pave an easy path to potential applications^{20,26}.

Among the topological materials, intense effort has been applied to realizing topological superconductors (TSCs)^{27,28}, one source of Majorana fermions, an effort which suffers from a severe

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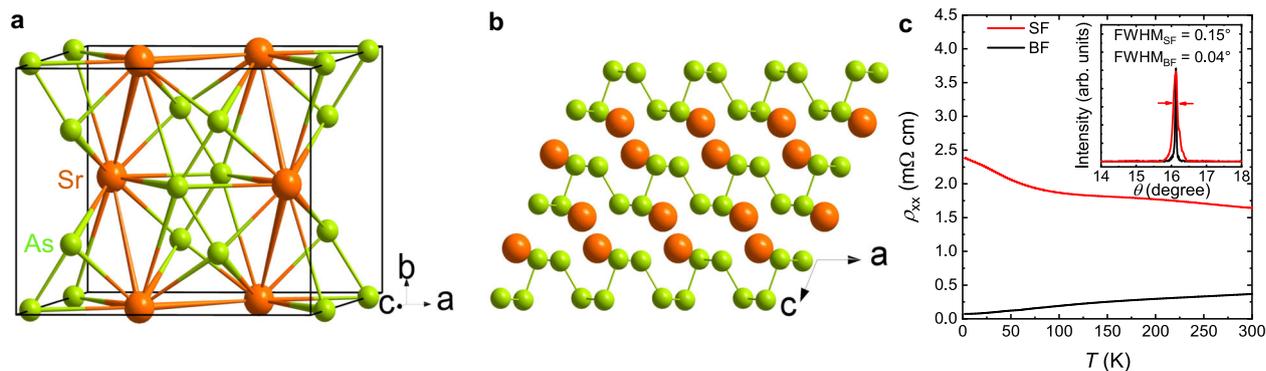


Fig. 1 **Characterization of SrAs₃.** **a** The unit cell of monoclinic SrAs₃. **b** The side view along the *b* axis. **c** Longitudinal resistivity of SrAs₃ single crystal. The black and red curves represent data collected on SrAs₃ single crystals obtained through Bi (BF) and self-flux (SF) methods, respectively. The red curve (SF) shows semimetallic behavior, while the other one (BF) displays metallic behavior. Inset: X-ray diffraction (XRD) rocking curves of the (002) Bragg peak for Bi-flux and self-flux single crystals. The full width at half maximum (FWHM) of bi-flux single crystal is 0.04°, while the FWHM of self-flux single crystal is 0.15°, indicating that the quality of bi-flux single crystals is higher.

lack of suitable materials to study^{27,28}. Experimentally, applying chemical doping or pressure to search for superconductivity in known topological materials are two common methods to obtain new TSC candidates^{27,28}. While chemical doping introduces chemical complexity and disorder, pressure is a clean and effective approach for tuning the interactions among multiple degrees of freedom, and superconductivity has been found in many materials via this route^{29–31}. Among the CaP₃ family of materials, CaAs₃ was proposed to host a single nodal loop due to time reversal, spatial inversion, and accidental degeneracies³². Li et al.³³ reported its transport properties under hydrostatic pressure up to 2.09 GPa, finding a decrease in the resistivity and a possible superconducting transition under pressure. Since SrAs₃ has already been demonstrated to be a TNLSM, the lack of any high-pressure report inspired us to explore its pressure dependence.

In this work, we present the results of high-pressure measurements on single-crystalline SrAs₃. Upon applying pressure, the topologically protected *a* pocket and trivial *β* pocket disappear around 1 GPa, and two higher frequencies denoted as *ε* and *ξ* emerge, indicating a Lifshitz transition. More interestingly, a superconducting transition has been observed from 20.6 GPa, with a dome-like pressure dependence. High-pressure X-ray diffraction (XRD) was conducted to investigate the high-pressure structure of SrAs₃, and a structural transition was found around 20 GPa. DFT calculations on the high-pressure structure of SrAs₃ reveal a TCI state. TCI states have previously been experimentally verified only in narrow-gap IV–VI semiconductors with a rock-salt structure, for example SnTe³⁴ and Pb_{1–*x*}Sn_{*x*}M (*M* = Se, Te)^{35,36}. Thus, the observation of a pressure-induced TCI state in SrAs₃ offers an alternative route to explore this exotic state. Moreover, the finding of superconductivity in this state makes high-pressure SrAs₃ a candidate TSC.

RESULTS

SrAs₃ crystallizes in a triclinic (space group *P* $\bar{1}$) or monoclinic (space group *C2/m*) structure; the latter is proposed to possess topological-nodal-line states protected by time-reversal symmetry, spatial-inversion symmetry, and mirror symmetry²⁰. Figure 1a shows the unit cell of monoclinic SrAs₃. This crystal structure can be viewed as a stack of two-dimensional (2D) infinite polyanionic layers ²_∞[P₃]^{2–} along the *b*-axis²¹. The As layers form channels and the Sr cations are inserted into the channels, as shown in Fig. 1b²¹. The inset in Fig. 1c shows the XRD rocking curves of SrAs₃ single crystals grown from both Bi flux (which we refer to as BF) and self flux (SF). From an X-ray rocking curve of the (002) Bragg peak, a full width at half maximum (FWHM) of 0.04° indicates the high

quality of the SrAs₃ single crystal grown from Bi flux, while a broader FWHM of 0.15° for the SF sample suggests lower quality. In resistivity measurements (Fig. 1c), the SF sample exhibits semimetallic behavior, while the BF sample exhibits metallic behavior with a residual resistivity ratio RRR = $\rho(300\text{ K})/\rho_0$ of 5, and a residual resistivity ρ_0 of 72.7 $\mu\Omega$ cm, which are further indicative of its high quality. For magnetotransport measurements, the higher-quality BF-grown samples were chosen.

Figure 2a shows the temperature-dependent resistivity from 1.8 to 300 K at pressures up to 1.47 GPa on a BF-grown SrAs₃ single crystal. Upon increasing the pressure from 0.14 to 1.18 GPa, the low-temperature resistivity becomes increasingly metallic. The MR, shown in Fig. 2b, is non-monotonic in pressure. Upon increasing the pressure from 0.14 to 1.18 GPa, the MR at 9 T and 1.8 K increases from 5000 to 32,500%. However, further increasing the pressure to 1.47 GPa reduces the MR to 22,000%. The oscillatory component ΔR_{xx} plotted in Fig. 2c, exhibits low-frequency modes for lower pressures and higher-frequency modes at higher pressures, indicating a significant change in the electronic structure at the Fermi surface. Fast Fourier transforms of the MR oscillations, displayed in Fig. 2d contain only the *a* (1.4 T) and *β* (5.5 T) pockets from 0.14 to 0.69 GPa, as previously seen at ambient pressure²⁵. At 0.99 GPa, the *a* and *β* pockets abruptly disappear, replaced by a single frequency of 21.5 T which we assign to a *ξ* pocket. Upon increasing the pressure to 1.47 GPa, the *ξ*-frequency pocket is joined by an even higher frequency of 48.3 T which grows rapidly to 63.2 T, which we assign to an *ε* pocket.

Figure 2e summarizes the pressure-dependent oscillation frequencies, with shading identifying the transition where the Fermi surface changes. The two obvious scenarios for this abrupt transition are a structural transition or a Lifshitz transition. But high-pressure diffraction, discussed in more detail below, indicates that the ambient-pressure structure persists to ~20 GPa, excluding a structural origin. In topological materials, different types of Lifshitz transitions are possible, involving other types of zeroes in the energy spectrum in addition to or instead of the Fermi surface, such as flat bands, Weyl and Dirac nodes, Dirac nodal lines, zeroes in the spectrum of edge states, Majorana modes, etc.³⁷. For example, in rhombohedral graphite, the Lifshitz transition is related to the change of the shape of the Dirac line³⁷. To study the relationship between the Lifshitz transition and the topologically protected nodal-line structure in SrAs₃, a Landau level index fan diagram for the pressure-induced pockets is plotted in Fig. 2f, where no Berry phase has been observed for either the *ε* or *ξ* pocket. To produce this diagram, we assign integer indices to the ΔR_{xx} peak positions in 1/*B* and half-integer indices to the ΔR_{xx} valleys. Infinite-field intercepts around 3/8 to 5/8 indicate trivial

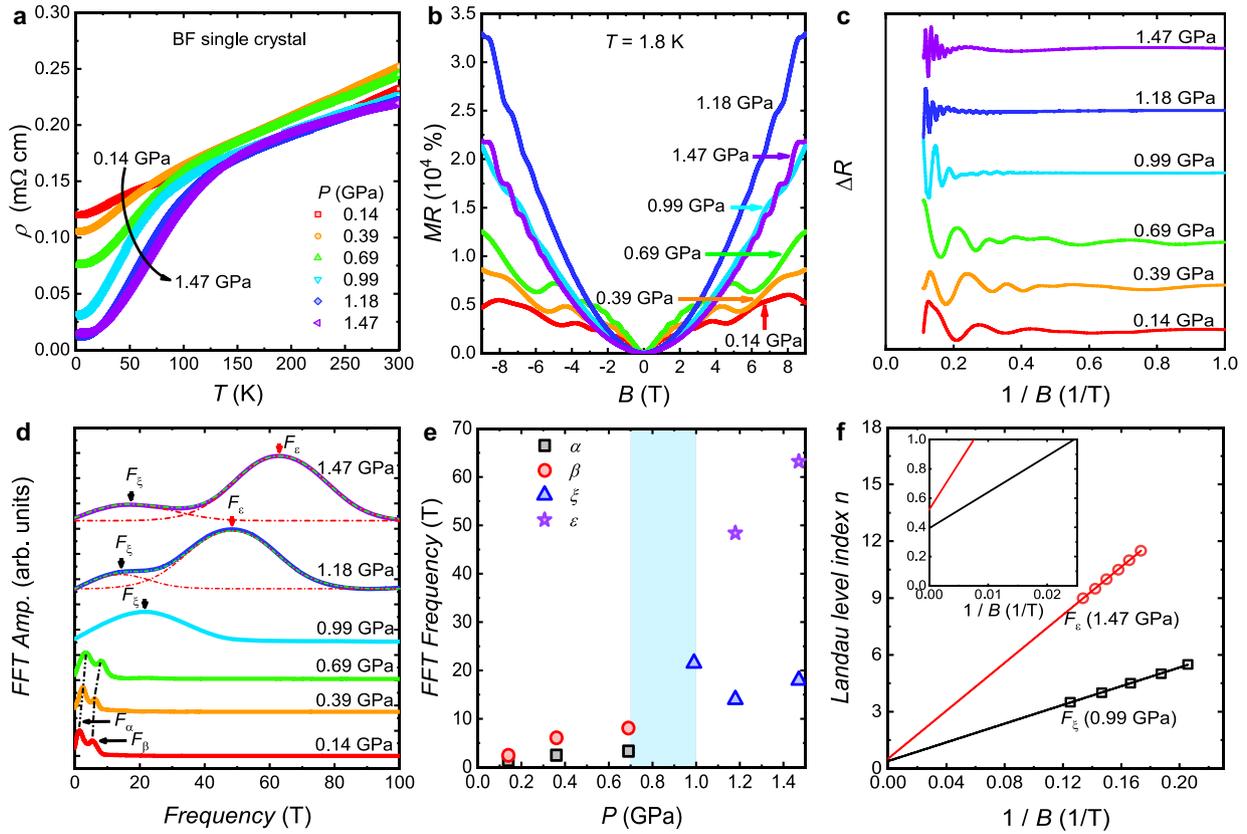


Fig. 2 Shubnikov–de Haas (SdH) oscillation study of SrAs_3 under low pressure. **a** Resistivity of a SrAs_3 single crystal grown from Bi-flux. Upon increasing the pressure to 1.18 GPa, the low-temperature resistivity is monotonously reduced. At 1.47 GPa, the resistivity slightly increases, relative to that at 1.18 GPa. **b** Pressure dependence of magnetoresistance (MR) of a SrAs_3 single crystal at 1.8 K. MR is a non-monotonic, with a maximum at 1.18 GPa and 9 T. **c** The oscillatory component ΔR_{xx} at different pressures, extracted from R_{xx} by subtracting a smooth background. **d** Fast Fourier transform (FFT) results for SdH oscillations at different pressures. From 0.14 to 0.69 GPa, the α and β bands shift slightly to higher frequency, then disappear suddenly at higher pressures, where two higher frequencies ϵ and ξ emerge. **e** Pressure dependence of FFT frequency. The shaded area contains a Lifshitz transition. **f** Landau level index plots for the ϵ band at 0.99 GPa, and the ξ band at 1.47 GPa. Both bands have trivial Berry phase.

band topology. In SrAs_3 , the α pocket is topologically protected, and the nodal line is situated around the Y point in the BZ²⁴. Hence, we surmise that the Fermi surface of SrAs_3 is reconstructed below 1.0 GPa, in a Lifshitz transition which involves the topologically protected nodal-line structure.

As a TNLSM, SrAs_3 possesses 2D drumhead-like nearly flat surface states nested inside the closed nodal line, and it may provide an ideal playground for many interaction-induced nontrivial states, such as superconductivity. In the resistivity under higher pressures in Fig. 2a, there is a clear suppression of the low-temperature resistivity. Pressure-induced superconductivity in topological semimetals often appears as the extremely large MR is suppressed³¹. Given the significant reduction in MR from 1.18 to 1.47 GPa, we suspected that higher pressures may induce superconductivity. Since bismuth displays superconducting transitions from 2.5 to 10.1 GPa with T_c changing from 3.9 to 8.55 K^{38,39}, self-flux-grown SrAs_3 crystals were studied to completely eliminate the possibility of Bi flux inclusions (the results of electrical transport measurements on the Bi-flux-grown SrAs_3 samples (as shown in Fig. S1) and discussions are only displayed in the Supplementary Information file). Figure 3a shows the resistance curves for SrAs_3 under higher pressures. No superconducting transition is observed till 20.6 GPa. At 20.6 GPa, a superconducting transition with $T_c^{10\%}$ of 3.6 K appears, and the T_c increases to 5.8 K at 54.7 GPa. Upon further pressurization to 63.6 GPa, the T_c decreases slightly to 5.5 K. The pressure dependence of the superconducting transition,

summarized in Fig. 3c, is clearly dome-shaped. To verify that this is a superconducting transition, the effect of magnetic field at 39.4 GPa was studied, as plotted in Fig. 3b—the transition is gradually suppressed by magnetic field, as expected for superconductivity. Figure 3d plots the temperature dependence of the upper critical field ($\mu_0 H_{c2}$). We used the Ginzburg–Landau (GL) formula $\mu_0 H_{c2}(T) = \mu_0 H_{c2}(0) (1 - (T/T_c)^2) / (1 + (T/T_c)^2)$ to fit the data at 39.4 GPa; the $\mu_0 H_{c2}(0)$ values are estimated to be 2.58(2), 2.00(2), and 1.48(3) T for $T_c^{10\%}$, $T_c^{50\%}$, and T_c^{zero} , respectively, yielding coherence length $\xi_{\text{GL}}(0)$ of 11.3, 12.9, and 14.9 nm. These fields are much lower than the Pauli limiting fields^{40–42} $H_p(0) = 1.84T_c \sim 9.5, 7.9$, and 6.9 T, respectively, indicating that Pauli pair breaking is not relevant.

To verify whether the pressure-induced superconductivity arises from a structural phase transition, we performed high-pressure synchrotron XRD measurements on the self-flux-grown samples, shown in Fig. 4a for different pressures. The ambient-pressure (denoted as AP) phase persisted up to ~ 20 GPa. New diffraction peaks (marked with a dashed line and asterisk) appear at ~ 23.3 GPa, indicating a pressure-induced structural phase transition, and these continue to strengthen upon further increasing pressure. Figure 4b depicts the crystal structure of SrAs_3 under ambient conditions. There are two Wyckoff positions (4i and 8j) for As atoms in each unit cell, and Sr (4i) atoms are coordinated to seven As atoms. The crystal structure of the ambient-pressure phase below 20.4 GPa was refined according to the initial model

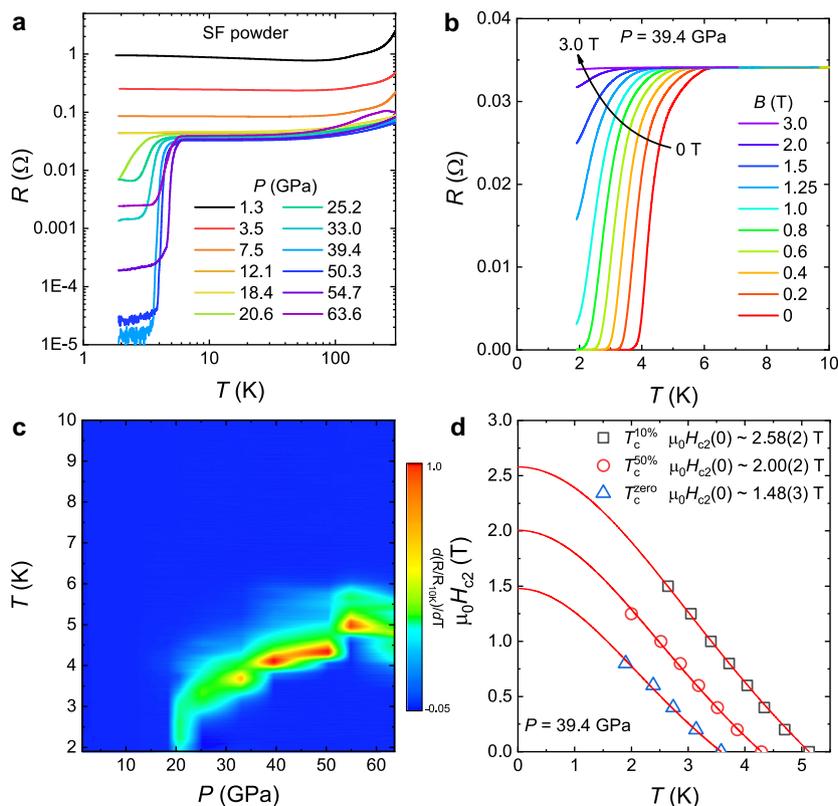


Fig. 3 Pressure-induced superconductivity in SrAs₃. **a** Temperature dependence of the resistance of SrAs₃ powder obtained by crushing a self-flux-grown single crystal. **b** Magnetic field dependence of the superconducting transition of SrAs₃ at 39.4 GPa. **c** Contour plot of the pressure dependent of the superconducting transition. The color represents the magnitude of $d(R/R_{10\text{K}})/dT$. **d** Temperature dependence of the upper critical field $\mu_0 H_{c2}$. The superconducting transition temperatures (T_c s) are defined according to the 10% drop of the transition ($T_c^{10\%}$), the 50% drop of the transition ($T_c^{50\%}$), and zero resistance (T_c^{zero}), respectively. The red line is the fit according to the Ginzburg–Landau theory, $\mu_0 H_{c2}(T) = \mu_0 H_{c2}(0) (1 - (T/T_c)^2)/(1 + (T/T_c)^2)$.

determined by room temperature single-crystal XRD. Typical GSAs refinements for SrAs₃ under 3.6 GPa are illustrated in Fig. 4c. In general, applying pressure tends to increase the coordination number (CN) of cations, the so-called “pressure-coordination” rule, as seen for instance in Bi₂Se₃, where $R\bar{3}m$ (CN = 6) transforms to $C2/m$ (CN = 7) and eventually $I4/mmm$ (CN = 8)⁴¹ with pressure. A further effect, the “corresponding static principle”, states that compounds containing light elements tend at higher pressures to adopt the structures observed at lower pressures in analogous compounds with heavy elements of the same group. A typical example is found in the binary antimonides Li₃X (X = N, P, As, Sb, and Bi): Li₃N crystallizes in the hexagonal $P6/mmm$ structure at ambient pressure and transforms into the hexagonal $P6_3/mmc$ structure (which is the ambient-pressure structure of Li₃P, Li₃As, and Li₃Sb) around 0.5 GPa. It then undergoes a second structural phase transition to a cubic $Fm\bar{3}m$ structure (the structure of Li₃Bi at ambient pressure) around 36–45 GPa⁴³.

At ambient conditions, SrP₃ and SrAs₃ have the same crystal structure ($C2/m$), while SrBi₃ forms in the cubic Cu₃Au-type structure (space group: $Pm\bar{3}m$, No. 221)⁴⁴. Thus the corresponding static principle suggests the Cu₃Au structure type as a strong candidate for the high-pressure structure. The crystal structure model for high-pressure SrAs₃ was deduced by testing several candidates, and was ultimately refined with a SrBi₃-like structure (space group: $Pm\bar{3}m$, No. 221)⁴⁴. The schematic crystal structure of the high-pressure phase is depicted in Fig. 4d. Sr and As atoms occupy 1a (0, 0, 0) and 3c (0, 0.5, 0.5) Wyckoff positions, respectively, and the CN for Sr has increased to 12. Figure 4e shows the Rietveld refinement of SrAs₃ under 49.8 GPa, yielding 70% and 30% for the ambient and high-pressure phases,

respectively. The coexistence of high and ambient-pressure phases up to 51.9 GPa indicates that the pressure-induced structural phase transition in SrAs₃ is first order and that the two phases have only a minute difference in Gibbs free energy.

Since the pressure-induced superconductivity appears long after the topologically protected a pocket is eliminated, it is important to check whether the high-pressure band structure of SrAs₃ is topologically nontrivial, which is a necessary condition for topological superconductivity. To obtain more electronic structure information on the high-pressure phase of SrAs₃, we performed DFT calculations for pressurized SrAs₃ at 34 GPa, as summarized in Fig. 5. The band structure in the absence of spin–orbit coupling (SOC), shown in Fig. 5a, displays metallic behavior. The electron-dominant valence and hole-dominant conduction bands cross near the Fermi level along the Γ – M line. Owing to the cubic symmetry of this material, there are twelve band crossing points at symmetrically equivalent points in the full BZ. Upon turning on SOC, a gap will be opened at these crossing points, resulting in a continuous SOC gap with a curved chemical potential between valence and conduction bands at each K point, as shown in Fig. 5b.

To identify the topological nature of this material, the Fu–Kane parity criterion^{45,46} at eight time-reversal invariant momenta (TRIM) was utilized to determine the Z_2 index. We obtain a trivial Z_2 of (0;000) from the production of the parities of all occupied bands at the eight TRIM points, as shown in Fig. 5c. However, the surface states on the (001)-projected surface contain two surface Dirac cones located at $\bar{\Gamma}$ points, as seen in Fig. 5d. Because of SOC, these Dirac states host a helical spin texture as shown in Fig. 5f. The Wilson loop method was employed to determine the mirror Chern number (MCN), and get MCN = 1, in agreement with the

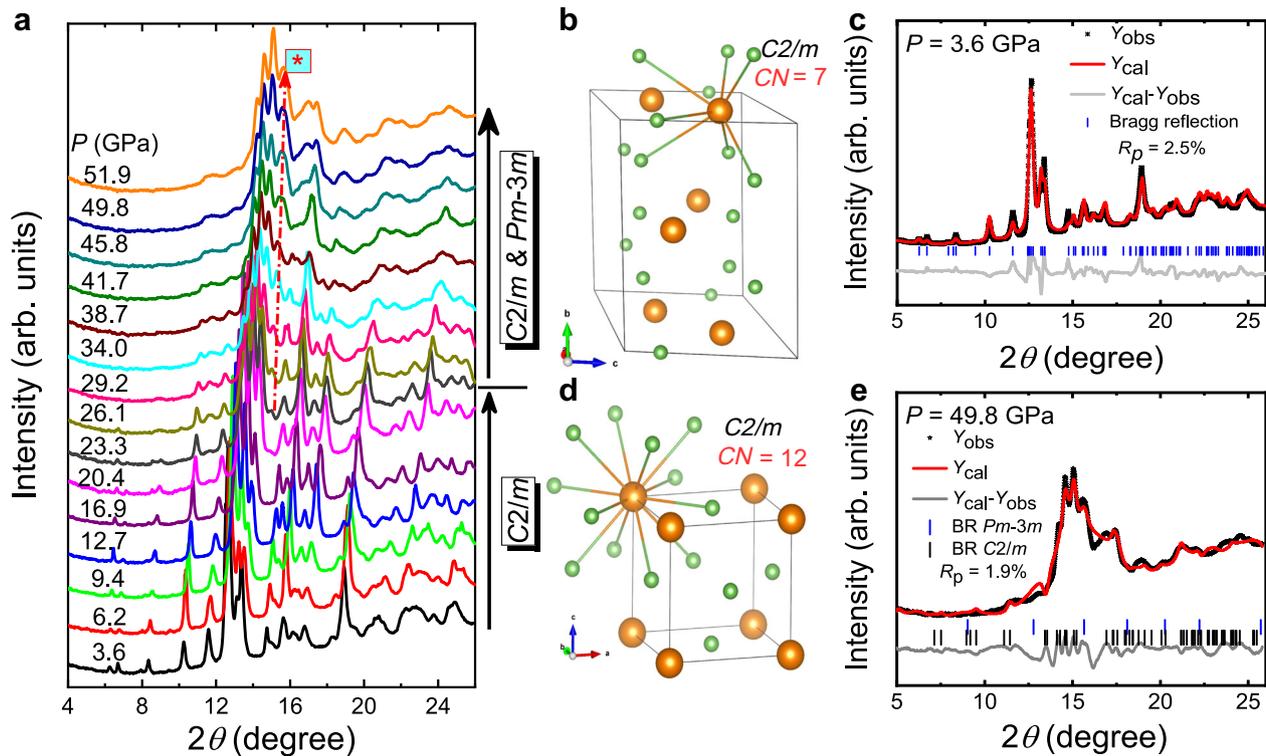


Fig. 4 X-ray diffraction (XRD) results of SrAs₃ powder under pressure. **a** Selected AD-XRD patterns of SrAs₃ at room temperature ($\lambda = 0.6199 \text{ \AA}$) up to 51.9 GPa. The ambient-pressure $C2/m$ structure remains stable up to ~ 20.0 GPa. As the pressure further increasing to 23.3 GPa, the appearance of new diffraction peaks (marked with dashed line and asterisk) indicates the emergence of pressure-induced structural phase transition ($C2/m \rightarrow Pm-3m$ (SrBi₃-like structure)) in SrAs₃. **b, d** The schematic crystal structure of ambient-pressure (AP) and high-pressure (HP) phases. The coordination number (CN) of Sr in two phases is 7 and 12, respectively. **c, e** Rietveld refined XRD patterns for 3.6 and 49.8 GPa, respectively. The AP and HP phases coexist at 49.8 GPa. The vertical lines denote the theoretical positions of the Bragg peaks. The different curves between observed and calculated XRD patterns are shown at the bottom.

surface state behavior observed on the (001) surface^{45–47}. The continuous SOC gap, topologically trivial Z_2 index, nontrivial MCN and even number of surface Dirac points indicate that this material is a TCI^{44–46}. As a counterpart of topological insulators in which crystalline symmetry replaces time-reversal symmetry to enforce topological protection, TCIs possess topological surface states (TSSs) with an even number of gapless Dirac cones on the surface BZ and host a variety of exotic phenomena⁴⁷, for example, large-Chern-number quantum anomalous Hall effect⁴⁸ or strain-induced superconductivity⁴⁹. The superconductivity in high-pressure SrAs₃ may exist in or be induced in these surface states. Owing to their helical spin texture, any superconducting phase in these surface states would most likely be topologically nontrivial^{34–36,47–49}, making high-pressure SrAs₃ a strong candidate for TSC.

DISCUSSION

Searching for Majorana fermions has been fueled by the prospect of using their non-Abelian statistics for robust quantum computation, and they can be realized as a bound state at zero energy, i.e., Majorana bound states, in the vortex core of a TSC^{27,28}. To realize a TSC, two primary routes have been proposed, i.e., bulk spin-triplet superconductivity, and superconductivity in spin-nondegenerate TSSs induced by the proximity effect, for instance through heterostructures stacking conventional s -wave superconductors and topological insulators, quantum anomalous Hall insulators, nanowires, or atomic chains^{27,28,50}. In the former case, due to the breaking of spin degeneracy by asymmetric SOC, the parity-mixed superconducting state in non-centrosymmetric superconductors may also host Majorana fermions if the singlet component is

smaller than the triplet component⁵¹. Recently, the layered non-centrosymmetric compound PbTaSe₂ with strong SOC was reported to possess fully gapped multiband superconductivity⁵². However, its spin-triplet component is small or absent^{53,54}. Bulk topological nodal-line states and fully spin-polarized TSSs have been also demonstrated in PbTaSe₂¹³, which may allow proximity-induced fully-gapped superconducting TSSs, which could pair in $p_x + ip_y$ symmetry and host bound Majorana fermions in the vortices¹⁴.

The superconducting phase of SrAs₃ is centrosymmetric ($Pm-3m$), so barring the exceedingly unlikely possibility of bulk spin-triplet superconductivity, the bulk route to Majorana quasiparticles is not available. However, we propose a TCI state hosting TSSs with helical spin texture. Superconductivity in these states or induced in these states by proximity effect from the bulk could potentially be topologically nontrivial. However, unlike in other TCIs such as SnTe or Pb_{1-x}Sn_xM ($M = \text{Se, Te}$), trivial bulk bands in pressurized SrAs₃ cross the Fermi level, and we are unable to distinguish which bands participate in the superconductivity. Further work will be required to elaborate the contributions from TSSs and bulk states to the superconductivity.

In summary, at ambient pressure, SrAs₃ is a TNLSM with 2D drumhead-like nearly-flat surface states, which may be strongly correlated and are often associated with the enhancement of superconductivity⁵⁵, although pressure did not succeed in driving this material superconducting before changing the electronic structure. A Lifshitz transition has been identified below 1.0 GPa, evidencing a topological phase transition, as the quantum oscillations associated with the TNLSM state vanish. Higher-pressure experiments on a powder sample reveal a dome-like superconducting transition accompanying a structural phase

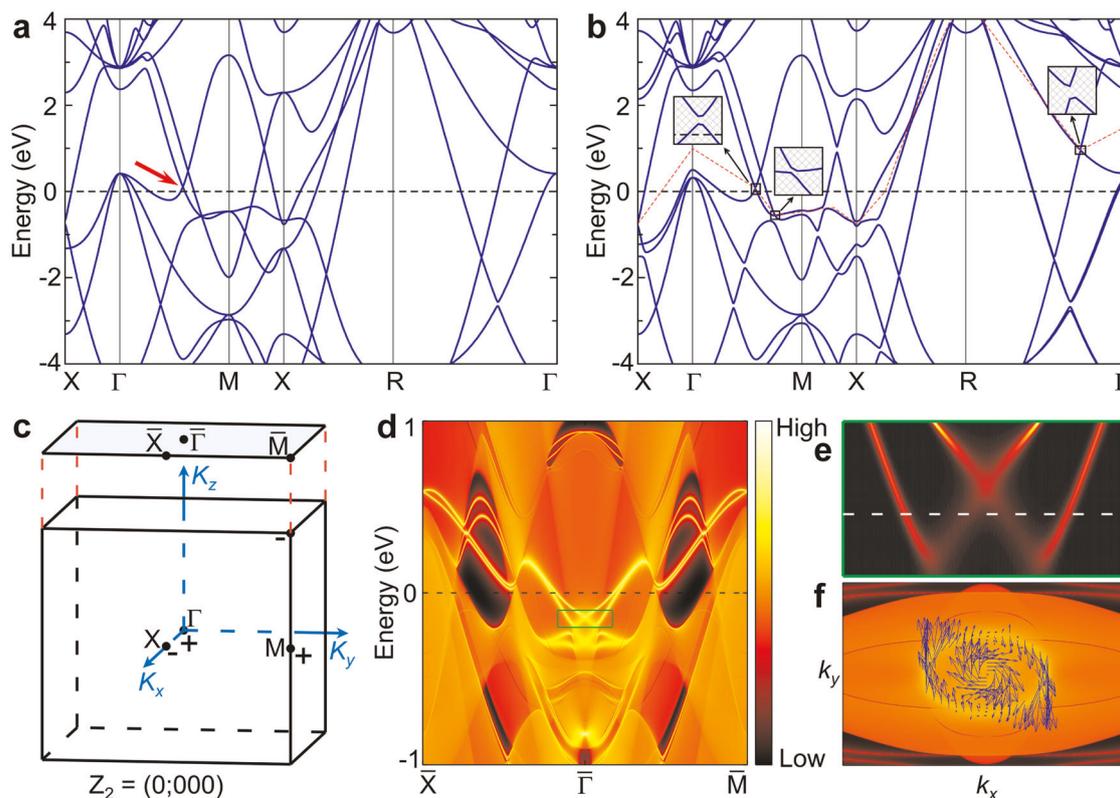


Fig. 5 Band structure for the high-pressure $Pm\text{-}3m$ phase of SrAs_3 . **a** The energy band of SrAs_3 without turning on SOC effect. Between Γ and M , the conduction and valence bands cross linearly close to the E_F . **b** The energy band of SrAs_3 with turning on SOC effect. The insets show the detailed band dispersion near the band crossing points. The red dashed line corresponds to a Fermi curve across the gap. **c** The bulk BZ and the associated (001) projected BZ with high-symmetry points. The \pm symbols represent the parity of all occupied bands at each TRIM. **d** The topological surface states of SrAs_3 on the (001) surface (the green region is enlarged in **e**). **f** The Fermi surface at -0.16 eV (marked in **e**), the blue arrows are the spin texture.

transition into a phase which we predict to host a topological-crystalline-insulator state with TSSs and helical spin texture. Besides its intrinsic interest as a TNLSM, SrAs_3 offers an alternative route to explore the topological-crystalline-insulator state beyond IV–VI semiconductors and, as a superconducting TCI, high-pressure SrAs_3 could serve as a candidate TSC. Doping studies or strain may still be able to induce superconductivity in the low-pressure phase, and should be pursued, and the evolution of the drumhead-like states with pressure remains to be clarified.

METHODS

Sample synthesis

Self-flux method. Sr (99.95 %, Alfa Aesar), and As (99.999 %, PrMat) were mixed in a molar ratio of 1:3 and placed into an alumina crucible. The crucible was sealed in a quartz ampoule under vacuum and subsequently heated to 750°C in 10 h. After reaction at this temperature for 300 h, the ampoule was cooled to 400°C in 50 h and cooled freely to room temperature. SrAs_3 single crystals with black shiny metallic luster were obtained.

BF method. Sr (99.95%, Alfa Aesar), As (99.999%, PrMat) and Bi (99.9999%, Aladdin) blocks were mixed in a molar ratio of 1:3:26 and placed into an alumina crucible. The crucible was sealed in a quartz ampoule under vacuum and subsequently heated to 900°C in 15 h. After reaction at this temperature for 20 h, the ampoule was cooled to 700°C over 20 h, and then slowly cooled to 450°C at $1^\circ\text{C}/\text{h}$. The excess Bi flux was then removed in a centrifuge, and SrAs_3 single crystals with black shiny metallic luster were obtained.

Pressure measurements

Resistance measurements under pressure. For high-pressure experiments, samples were loaded in a piston-cylinder clamp cell made of Be–Cu alloy,

with Daphne oil as the pressure medium. The pressure inside the cell was determined from the T_c of a tin wire. A SrAs_3 single crystal was cut into a bar shape, and the standard four-probe method was used for resistivity measurements, with contacts made using silver epoxy. Higher-pressure measurements were performed on powder samples comprising crushed single crystals using a diamond anvil cell (DAC). The experimental pressures were determined by the pressure-induced fluorescence shift of ruby⁵⁶ at room temperature before and after each experiment. A direct-current van der Pauw technique was adopted. Resistance measurements were performed with a physical property measurement system, Quantum Design.

XRD measurements under pressure. SrAs_3 single crystals grown by the self-flux method were ground into fine powder in a mortar for use in the high-pressure synchrotron angle dispersive XRD (AD-XRD) measurement. The high-pressure synchrotron XRD experiments were carried out using a symmetric DAC with a 260-micron culet diamond. A rhenium gasket was precompressed to 30 microns in thickness followed by drilling the central part by laser to form a 90-micron diameter hole as the sample chamber. The sample chamber was filled with a mixture of the sample, a ruby chip, and silicone oil as the pressure transmitting medium. The experimental pressures were determined by the pressure-induced fluorescence shift of ruby⁵⁶. Synchrotron AD-XRD measurements were carried out at beamline BL15U1 of the Shanghai Synchrotron Radiation Facility (SSRF) using a monochromatic beam of 0.6199 \AA .

Density functional theory (DFT) calculations

DFT calculations were performed based on the Perdew–Burke–Ernzerhof-type generalized gradient approximation⁵⁷, and used the projector augmented wave method⁵⁸, as encoded in the Vienna ab initio simulation package⁵⁹. The cutoff energy for the plane-wave basis taken was 500 eV. The first Brillouin Zone was sampled, using a Γ -centered $12 \times 12 \times 12$ k -point mesh. The energy convergence criteria were defined as 10^{-8} eV.

The lattice constants were fully relaxed using a conjugate gradient scheme until the Hellmann-Feynman forces on the ions were less than $0.001 \text{ eV}/\text{\AA}$. We constructed the maximally localized Wannier functions^{60–62} using Sr d and As s and p atomic orbitals. The topological features of surface state spectra were calculated using the iterative Green's function technique⁶³, as implemented in the Wannier-Tools package⁶⁴.

DATA AVAILABILITY

All data that support the findings of this study are available from the corresponding author upon reasonable request.

CODE AVAILABILITY

The computer codes used to carry out the DFT calculations in this work are available upon request from the corresponding author.

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AUTHOR CONTRIBUTIONS

S.Y.L. and Y.F.G. conceived the idea and designed the experiments. E.J.C. was responsible for high-pressure transport experiments. W.X., Y.Z., X.W., Z.Q.Z., H.S., and X.F.K. performed sample synthesis and partial data analysis. Z.H.Y., N.Y., L.W., L.M.Y., and W.G.Y. conducted the high-pressure X-ray diffraction measurements and structure analysis. C.C.Z. and D.Z.D. helped the data collection. S.Y.L., Y.F.G., and W.W.Z. supervised the project. E.J.C., D.C.P., Y.F.G., and S.Y.L. analyzed the data and

wrote the paper. E.J.C., W.X., and X.B.S. contributed equally to this work. All authors discussed the results and commented on the paper.

COMPETING INTERESTS

The authors declare no competing interests.

ADDITIONAL INFORMATION

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