Robust magnetoresistance in TaAs$_2$ under pressure up to about 37 GPa

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ABSTRACT
The extremely large magnetoresistance (XMR) in nonmagnetic semimetals has inspired growing interest owing to both intriguing physics and potential applications. We report the results of synchrotron X-ray diffraction and electrical transport measurements on TaAs$_2$ under pressure up to ~37 GPa, which revealed an anisotropic compression of the unit cell, formation of unusual As-As bonds above 9.5 GPa, and enhancement of metallicity. Interestingly, the MR of TaAs$_2$ under pressure changed gently, which at 1.7 GPa is 96.6% and at 36.6 GPa is still 36.7%. The almost robust MR under pressure could be related to the nearly stable electronic structure unveiled by the ab initio calculations. The discovery would expand the potential use of XMR even under high pressure.
carrier compensation mechanism. On the other hand, recent studies on NbAs$_2$ under pressure detected superconductivity with the $T_c$ of 2.63 K.\textsuperscript{21} It naturally reminds us of the examination of possible superconductivity in pressured TaAs$_2$. Furthermore, to expand the practical use of the XMR of TaAs$_2$, the pressure stability definitely deserves examination.

Crystals of TaAs$_2$ were grown by a chemical vapor transport method.\textsuperscript{11} The crystallographic phase and quality were examined on a Bruker D8 VENTURE single crystal diffractometer using Mo $K_a$ radiation ($\lambda = 0.7093$ Å) at room temperature. Electrical transport measurements under HP were carried out using a BeCu Diamond Anvil Cell (DAC) with the four-probe method in a 9 T DynaCool physical property measurement system. The HP synchrotron XRD (SXRD) experiments were performed at room temperature by using a symmetric DAC and T301 stainless steel gasket. The 120 $\mu$m diameter sample chamber was filled with a mixture of sample powder, a ruby
chip, and silicone oil as the pressure-transmitting medium. Angle dispersive XRD (AD-XRD) experiments for TaAs2 were performed at the BL15U1 beamline (wavelength: 0.6199 Å) of the Shanghai Synchrotron Radiation Facility (SSRF). The AD-XRD experiments were carried out at room temperature. The pressure determination in our experiments was according to the fluorescence shift of ruby. The AD-XRD experiments were analyzed with Rietveld refinement using the GSAS program package1 with a user interface EXPGUL. The first-principles calculations were performed by the Vienna ab initio simulation package (VASP)26 and the projected augmented-wave (PAW) potential was adopted.27,28 The exchange-correlation functional introduced by Perdew, Burke, and Ernzerhof (PBE) within generalized gradient approximation (GGA) was applied in the calculations.29 The energy cut off for the plane wave basis was set as 520 eV, and the forces were relaxed less than 0.01 eV/Å. The positions of atoms were allowed to relax, while the lattice constants of the unit cells were fixed to the experimental values.

The single crystal XRD analysis confirmed the monolithic NbAs2-type structure (space group C2/m, no. 12) for TaAs2 with a = 9.3385 Å, b = 3.3851 Å, c = 7.7568 Å, and β = 119.70° (Fig. S1 of the supplementary material). Figure 1(a) presents selected AD-XRD patterns of TaAs2 under various pressures. The Bragg peaks exhibit a minute shift toward higher angles caused by the lattice contraction upon increasing pressure. The XRD measurements with the pressure up to 37.6 GPa did not detect any new diffraction peaks arising from other phases or impurities. The Rietveld refinement therefore adopted the C2/m structure as the initial model, and the result of the data at 12.9 GPa is shown in Fig. 1(b), confirming that the model is correct. The detailed Rietveld refinement results are summarized in Table S1.

The formation of unusual As-As bonding states in TaAs2 under HP was observed. The shortest As-As distance at ambient conditions is 2.42 Å, implying that there is no As-As bonding because the covalent radius of As is 1.2 Å. Formation of chemical bonding in metalloid anions such as P, As, and Sb was previously theoretically investigated in transition metal pnictides. The experimental observation of a formation of As-As interlayer bonding was in the collapsed tetragonal NaNbAs2 under pressure.32 The As-As distance for As(1) and As(2) atoms in pressured TaAs2 is presented in Fig. 2(a), showing that all these As-As distances decrease upon increasing pressure. The As(1)-As(1) distance at AP is ~3.0 Å, decreases gently with the increasing pressure, and eventually reaches a minimal value of 2.887 Å at 37.6 GPa, indicating that the As(1)-As(1) bonding could not be formed. The arrangement of the As(2) atoms produces alternately longer (~2.87 Å) and shorter (~2.42 Å) As(2)-As(2) distances at AP. The short As(2)-As(2) distance in Fig. 2(a) exhibits a clear decrease with increasing pressure and becomes smaller than 2.40 Å at ~9.5 GPa, suggesting that the short As(2)-As(2) interactions (<2.4 Å) are actually...
chemical bonds as shown by the inset in Fig. 2(b). The bonding state between metalloid As atoms is one of the intriguing features, and it tends to enhance the metallicity of TaAs₂. Furthermore, the Ta-Ta distance decreases upon the pressure increasing and approaches 2.92 Å at 37.6 GPa, as shown in Fig. 2(b), which indicates that the formation of Ta-Ta bonding is at much higher pressure than that of As(2)-As(2).

The pressure dependent \( R(T) \) of TaAs₂ is presented in Fig. 3(a), displaying a monotonic decrease in magnitude with the increasing pressure. The low temperature upturn of \( R(T) \) at AP is easily completely suppressed with a small pressure. Unfortunately, unlike NbAs₂, no superconductivity in TaAs₂ was traced within the measured pressure range. The lower temperature \( R(T) \) was fitted by the Bloch-Gruneisen equation expressed as \( R(T) = R_0 + A \times T^n \), where \( R_0 \) is the residual resistivity at zero temperature and \( A \) and \( n \) are the fitting parameters. The results of representative fitting to the data at 1.7 and 36.6 GPa are presented in Figs. 3(b) and 3(c). A power law behavior is clearly visible with the exponent \( n \) decreasing from 3.2 at 1.7 GPa to 3.0 at 36.6 GPa. The two values are close to that expected for s-d electron scattering, \( n = 3 \). The results imply that interband scattering within the chain orbitals between Ta and As orbitals plays a crucial role in influencing the transport properties. The \( R(T) \) curves between 20 and 300 K were fitted by using the parallel-resistor model to interpret the observed saturating behavior in TaAs₂, see in Fig. S4, which also yielded the pressure dependence of resistivity determined Debye temperature (Fig. S5). At low pressures, we note that \( R(T) \) of TaAs₂ shows negative curvatures in a high temperature region, seen in Fig. S4(d), which is proposed to originate from the resistivity saturation term of the sample with electron mean free path \( l \) [see Figs. S6(a) and S6(b)] being comparable with the lattice parameters. Upon further increasing the pressure, \( l \) may eventually exceed the progressively compressed lattice parameters, thus leading to the visible transition of \( R(T) \) from the nonlinear to almost linear temperature dependent behavior as is indicated by the marked arrow.

The MR of TaAs₂ at 2 K and 9 T is ~2.1 × 10⁵%, which shows clear suppression by the application of pressure. The value is 96.6% at 2 K, 9 T, and 1.7 GPa but then surprisingly changes gently with continuously increased pressure, which is 90.9%, 81.8%, 77.4%, and 36.7% at 3.4, 6.8, 9.7, and 36.6 GPa, respectively, seen in Fig. 4(a). Li et al. reported that when a 14.6 GPa pressure is applied, MR of NbAs₂ at 2 K and 8 T is effectively suppressed to 8% of that at AP and is almost inhibited with the increasing pressure to 14.6 GPa. Our result implies a robust MR of TaAs₂ under pressure. The power-law relation, \( MR \propto (B^LZ^YX^Y) \), where \( B \) is the averaged mobility and \( Z \) is a fitting coefficient, was used to fit the experimental data as shown in Fig. 4(b), revealing that the averaged carrier mobility clearly decreases with the increasing pressure, which should play an important role in reducing the MR.

The calculated band structures are presented in Fig. 5. Under ambient conditions and without the spin-orbital coupling (SOC), the conduction and valence bands of TaAs₂ cross along three high-symmetry paths \( I \rightarrow Z, I \rightarrow L \), and \( X \rightarrow Y \) [see in Fig. 5(a)], while at other paths, the band dispersion is relatively small which cannot induce a band-inversion. One can easily confirm that these band-crossing paths are parallel to the double-chains. Because of the chainlike structure of TaAs₂, the electrons move more freely along the chains than between the chains, which explains the large-dispersions and band-crossings at \( I \rightarrow Z, I \rightarrow L \), and \( X \rightarrow Y \). When SOC is included, the band crossings at \( I \rightarrow Z, I \rightarrow L \), and \( X \rightarrow Y \) are lifted and energy gaps open at these crossing points. Previous studies have classified the TaAs₂ as a type-II Dirac semimetal without SOC and a weak topological insulator with SOC by computing the Z₂ indices at the time-reversal-invariant-momenta of the Brillouin zone (BZ).

Next, we show that the electronic structure of TaAs₂ is only slightly altered by the HP, and the conduction and valence bands are kept in being inverted. In Figs. 5(b) and 5(c), we show the band structures of TaAs₂ at the pressures of 9.5 and 37.6 GPa, respectively. Due to the shrinking of lattice constants, one sees that the band dispersion of electrons at \( I \rightarrow Z, I \rightarrow L \), and \( X \rightarrow Y \) increases as compared to the band structure of TaAs₂ at ambient conditions in Fig. 5(a), which even further enhances the band inversion at these paths. Therefore, we can conclude that the electronic structure of TaAs₂ is highly stable against the external pressure and the material remains a weak topological insulator.

As a summary, we report the observation of the formation of unusual As-As bonding and the nearly robust XMR in TaAs₂ under high pressure. The formation of As-As bonding highlights the role of high pressure in creating unusual bonding states that would influence the physical properties or give rise to exotic behaviors. Considering the potential applications of the NbAs₂-family of materials, the robust XMR makes TaAs₂ more promising for use even under HP.
TaAs₂ under ambient conditions, TaAs₈ polyhedron stacking along different axes, and the pressure dependence of lattice parameters for TaAs₂ single crystal synthesized in this work, the crystal structure of TaAs₂. The results would also bring valuable clues for understanding the electrical transport properties and intriguing band topology of TaAs₂.

See the supplementary material for the optical image of the TaAs₂ single crystal synthesized in this work, the crystal structure of TaAs₂ under ambient conditions, TaAs₂ polyhedron stacking along different axes, and the pressure dependence of lattice parameters for TaAs₂. Please refer to supporting data guidelines at https://aip.scitation.org/apl/authors/manuscript.

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