Field-induced resistivity plateau and unsaturated negative magnetoresistance in topological semimetal TaSb$_2$

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Several prominent transport properties have been identified as key signatures of topological materials$^{1,2}$. One is the resistivity plateau at low temperatures as observed in several topological insulators (TIs)$^{3,4}$; another is the negative magnetoresistance (MR) when the applied magnetic field is parallel to the current direction as observed in several topological semimetals (TSMs) including Dirac semimetals (DSMs)$^{7,8}$ and Weyl semimetals (WSMs)$^{13,14}$. Usually, these two exotic phenomena emerge in distinct materials with or without time reversal symmetry (TRS), respectively. Here we report the discovery of a new member in TSMs, TaSb$_2$, which clearly exhibits both of these phenomena in a single material. This compound crystallizes in a base-centered monoclinic, centrosymmetric structure, and is metallic with a low carrier density in the zero field. While applying magnetic field it exhibits insulating behavior before appearance of a resistivity plateau below $T_c = 13$ K. In the plateau regime,
the ultrahigh carrier mobility and extreme magnetoresistance (XMR) for the field perpendicular to the current are observed as in DSMs\textsuperscript{18–22} and WSMs\textsuperscript{23–29}, in addition to a quantum oscillation behavior with non-trivial Berry phases. In contrast to the most known DSMs and WSMs, the negative MR in TaSb\textsubscript{2} does not saturate up to 9 T, which, together with the almost linear Hall resistivity, manifests itself an electron-hole non-compensated TMS. These findings indicate that the resistivity plateau could be a generic feature of topology-protected metallic states even in the absence of TRS and compatible with the negative MR depending on the field direction. Our experiment extends a materials basis represented by TaSb\textsubscript{2} as a new platform for future theoretical investigations and device applications of topological materials.

Diverging longitudinal resistivity with decreasing temperature is the most apparent transport property of a simple band insulator in distinction to any metals. However, such a distinct feature ceases to stand in various topological insulators due to the existing metallic surface states\textsuperscript{1, 2}. Nevertheless there are other transport phenomena, such as resistivity plateau and negative MR, which may distinguish at least ideal TIs and ideal TSMs from topologically trivial materials. In an ideal 3D TI where the bulk states are completely gapped out near the Fermi level, a resistivity plateau can be clearly established because the only participating surface states are TRS protected and thus robust to disorders, leading to a saturation of resistivity in the low temperature regime\textsuperscript{3–6}. In TSMs without coexisting bulk Fermi surfaces such as an ideal WSM, on the other hand, the bulk exci-
tations come from the two separated Weyl nodes in momentum space which are chiral in nature owing to the lack of TRS or inversion symmetry. When the applied magnetic field is parallel to the electric field direction, the density of the right/left chiral excitations increases/decreases accordingly as a consequence of the chiral anomaly, resulting in a non-dissipative current from the left to right nodes along the field direction, hence an unconventional negative MR appears.

Because the resistivity plateau and negative MR are opposite consequences for systems with or without TRS in the ideal situations mentioned above, coexistence of the both in a single material is unlikely or very difficult. Of course, these features should be more intriguing but much involved in realistic topological materials, and, TRS itself is not the only origin relevant to the resistivity plateau on a general ground. In two-dimensional electron gases or semiconducting films like graphite, for instance, the resistivity seemingly saturates after a field-induced metal-insulator transition while a clear resistivity plateau at lower temperatures was not reported. More recently, a field-induced plateau has been observed in LaSb, a potentially new candidate of TIs. While the interpretation of all these and related behaviors in topological materials remains a theoretical challenge, materials realizations of these effects are highly desirable not only in confronting this challenge but also in technique applications.

Here we report the discovery of a new TSM TaSb$_2$, which crystallizes in a monoclinic structure with centrosymmetric space group $C_{12/m1}$ (SI-Fig. S1). The longitudinal resistivity and Hall effect were measured for various magnetic fields applied along different directions. The semicon-
ducting behavior is associated with a low carrier density and a field-induced metal-to-insulator-like transition. High quality of the measured samples is indicated by ultrahigh mobility and XMR. The topological nature of the compound is evidenced by the Shubnikov de Haas (SdH) oscillation measurement as well as band structure calculations (SI-Fig. S6). Surprisingly, both the field-induced resistivity plateau and unsaturated negative MR can be clearly observed in this compound as illustrated in the following. Therefore, our experiment shows that the monoclinic TaSb$_2$ represents a potentially new class of topological materials exhibiting all these novel phenomena.

The magneto-transport properties of the sample 1 for TaSb$_2$ is summarized in Figure 1, where the applied magnetic field $\mathbf{B}$ is parallel to the c-axis, and normal to the current. Figure 1a describes the temperature dependent longitudinal resistivity $\rho(T)$ at various fields ($\mu_0H$ up to 9 T). At $\mathbf{B} = 0$, $\rho(T)$ exhibits highly metallic behavior down to 2 K. Its value at 300 K is $7.4 \times 10^{-2}$ mΩcm, about one order smaller than that of WTe$_2$, an electron-hole compensated semimetal with XMR$^{39}$. For nonzero field $\mathbf{B} (= \pm |\mathbf{B}|)$, $\rho(T)$ firstly decreases, then increases rapidly upon cooling, showing a crossover to insulating behavior. The insulating behavior becomes prominent when $\mathbf{B} > 1$ T. Remarkably, $\rho(T)$ saturates at the low temperature regime, developing a resistivity plateau which is clearly exhibited in Figure 1b with a logarithm scale.

This finding reminds us of the compound SmB$_6$, an important candidate of TIs exhibiting a very similar resistivity plateau at zero field$^{50,51}$. In that compound, the topological non-trivial surface states have been evidenced by both experiments$^{40,41}$ and band structure calculations$^{42}$, so the origin
of the plateau is best understood as due to the surface states. The similar plateau in $\rho(T)$ but under applied fields has been also observed in LaSb and other semimetal compounds, the former was predicted as a new kind of TIs. The plateau of TaSb$_2$ onsets at $T = 13$ K, almost three times larger than $T = 5$ K in SmB$_6$, and comparable to $T = 15$ K in LaSb.

Figure 1c shows temperature dependence of the derivative $\partial \rho(T)/\partial T$ at different fields. A clear drop is seen for $B \gtrsim 2$ T and becomes prominent for larger B. The temperature location of the drop peak, $T_i$, where $\partial^2 \rho(T)/\partial T^2 = 0$, is the inflection point where crossover from insulating behavior to plateau takes place. The metal-insulating-like transition takes place at an elevated temperature, $T_m$, where $\partial \rho(T)/\partial T = 0$. The inset of the Figure 1c shows the evolution of $T_m$ and $T_i$ vs. magnetic field B, indicating that $T_m$ increases monotonously and $T_i$ remains almost unchanged. This silent feature implies that the insulating behavior is of magnetic origin while the plateau may be of topological one. If this is true, the later could be there at zero field. Indeed, we find that the lines of $T_m$ and $T_i$ seem to merge at $B = 0$ T, suggesting a possible plateau there. In order to estimate the insulating gap, $E_g$, Figure 1d plots the $\log(\rho)$ as a function of $T^{-1}$ in the range of $T_i < T < T_m$ using $\rho(T) = \rho_0 \exp(E_g/K_BT)$ with constant $\rho_0$. The inset in Figure 1d shows the fitted activation energy gap $E_g \propto \sqrt{B}$. This is consistent with the gap opening in relativistic Dirac electrons by the magnetic field.

Figure 2 plots the field dependence of MR, where negative B means the opposite direction. As shown in the upper panel of Figure 2a, the XMR of TaSb$_2$ at low temperatures is exhibited
when the field is perpendicular to the current, reaching 15000% at $T = 2$ K and $B = 9$ T. This MR is quadratic for low field and almost linear for larger $B$ without saturation, similar to many known semimetallic materials including TaAs(P), NbAs(P) and WTe$_2$.[13, 23, 27, 29, 39] Upon heating, the MR decreases slowly below 10 K, but drops quickly at higher temperatures. The inset in the upper panel of Figure 2a shows a window for $7 \leq B \leq 9$ T where the SdH oscillations are clearly exhibited at $T = 2$ K and 5 K, respectively.

We also measured the MR by applying the magnetic field parallel to the current $B || I$, as shown in the low panel of Figure 2a. It is remarkable that (i) the MR in the whole temperature regime is less than 100%, much smaller than that in the case of $B \perp I$ shown previously; (ii) in the low temperature regime such as $T = 2$ K and 10 K, the MR is positive and parabolic for $B \lesssim 6.5$ T; (iii) while for $B \gtrsim 6.5$ T, it becomes negative and decreases with increasing magnetic field without saturation up to $B = 9$ T; (iv) when $T \gtrsim 50$ K, the MR is very small and positive, increasing monotonously with field.

Figure 2b describes the field dependence of MR at a fixed temperature $T = 2$ K with different angles $\theta$ between the magnetic field and current. The overall profile is nearly symmetric under $B \rightarrow - B$. By rotating $\theta = 90^\circ \rightarrow 0^\circ$, the MR drops quickly, in particular when approaching $\theta \approx 10^\circ$. The tendency of negative MR for smaller $\theta$ is further illustrated in the low panel of Figure 2b. At $B = 9$ T, the MR turns to negative for $\theta \lesssim 4^\circ$. The window for the negative MR is limited from this point to $B \gtrsim 6.5$ T at $\theta = 0^\circ$. The similar narrow $\theta$ window was observed in Na$_3$Bi and TaP compound.[9,13] The unsaturated negative MR, which decreases monotonically with fields
reaching about -74% at \( B = 9 \), is a remarkable feature compared to the prototype DSMs and WSMs such as \( \text{Na}_3\text{Bi} \)[12], \( \text{Cd}_3\text{As}_2 \)[14][15], \( \text{TaAs} \)[14], \( \text{NbP} \)[16], where the negative MR is limited not only in a narrow window of \( \theta \), but also in a window of \( B \), namely, the negative MR will saturate and return to positive for larger \( B \) even for \( \theta = 0^\circ \). In view of chiral anomaly, the MR should be always negative as long as \( B||I \) (or \( \theta = 0^\circ \)). If imperfect alignment of the magnetic field and the current in samples can be fully excluded, the limited window in \( B \) should be due to the disorder-induced weak localization. So the unsaturated negative MR observed here implies the consistency with the chiral anomaly interpretation and the high quality of the measured samples.

Figure 3 maps the Hall effect and the SdH oscillations. The magnetic field dependence of Hall resistivity at various temperatures is displayed in the main panel of Figure 3a. The negative slope for all of curves implies that electron-type carriers dominate the transport from 300 K to 2 K. The \( \rho_{xy} \) displays an overall linear dependence, and only slightly deviates from linear at around 9 T as \( T \leq 20 \) K. The Hall coefficient \( R_H \) versus temperature at 6 T and 9 T is plotted in the inset of Figure 3a. \( R_H \) is always negative and drops soon below 50 K without a sign change. Notice that the strong nonlinear behavior in \( \rho_{xy} \) and the sign changed \( R_H \) in several semimetals have been regarded as indications of the electron-hole compensation. Obviously, this interpretation cannot apply to the present \( \text{TaSb}_2 \) sample.

Accordingly, the calculated carrier concentration is \( n_e \sim 3.2 \times 10^{20} \text{ cm}^{-3} \) using \( n_e = 1/eR_H(T) \) and the estimated mobility \( \mu_e \sim 1.96 \times 10^4 \text{ cm}^2\text{V}^{-1}\text{S}^{-1} \) at 2 K. Hence, \( \text{TaSb}_2 \) has a low carrier
density of electron-type but with a high carrier mobility, similar to the results of LaSb$^{39}$. A direct consequence of these for the SdH oscillation is shown in Figure 3b for $\rho_{xx}(B)$ at 1.8 K, 4.2 K and 8 K, respectively. We extract the SdH oscillations using $\rho_H = \rho_0[1 + A(B, T)\cos2\pi(S_F/B - \gamma + \delta)]$, with $\rho_0$ being the non-oscillatory part, $A(B, T)$ the amplitude, $\gamma$ the Onsager phase, $F = \frac{h}{2\pi S_F}$ the frequency, and $S_F$ the cross-section area of the Fermi surface associated with the Landau level index $n$. The background is subtracted using a polynomial fitting. The obtained $\Delta \rho_{xx}$ as a function of $B^{-1}$ is then plotted in the inset of Figure 3b. Two oscillation sets can be extracted (SI-Fig. S5), corresponding to two frequencies: a small oscillation frequency at $F_\alpha = 220$ T, and a second frequency at $F_\beta = 465$ T with its harmonic $F_{2\beta} \approx 930$ T and $F_{3\beta} \approx 1377$ T, as shown in Figure 3c. The Berry phase $\Phi_B$ can be identified via the relation $\gamma = 1/2 - \Phi_B/2\pi$, so $\Phi_B$ is non-trivial when $\gamma \neq 1/2$. We thus plot the Landau fan diagram in Figure 3d, and count down to $n = 8$ and 16 for $F_\alpha$ and $F_\beta$, respectively, up to magnetic field 30 T. Linear fitting of $n$ versus $B^{-1}$ yields the Onsager phase of $\gamma_\alpha = 0.29$ and $\gamma_\beta = 0.2$, respectively, far away from one half, indicative of non-trivial $\pi$ Berry phases in TaSb$_2$. The slopes of the linear fitting are $F_{\alpha\text{fit}} = 228$ T and $F_{\beta\text{fit}} = 461$ T, respectively, consistent with the experimental values. The non-trivial Berry phases identified here indicate the topological origin of the resistivity plateau. This conclusion is also consistent with the first principle calculations for the electronic band structure of TaSb$_2$ as described in the Supplemental Information. The calculations indicate that TaSb$_2$ has a small bulk Fermi surface and a Dirac cone near the Fermi level, both of them are contributed by the partially occupied, topologically non-trivial electronic bands.
In summary, we report the discovery of a TMS TaSb$_2$ with a monoclinic crystal structure. It undergoes a metal-insulator-like transition induced by magnetic field upon cooling. Yet this compound shares a number of excellent transport properties including the positive XMR and high mobility. In the low temperature regime, it exhibits both the resistivity plateau and unsaturated MR when the applied field is perpendicular and parallel to the current, respectively. The topological property is manifested by the non-trivial Berry phases in the SdH oscillations as well as the band structure calculations.

Given the fact that the resistivity plateau and the negative MR are characteristic features of ideal TIs and TSMs with and without TRS, respectively, the coexistence of these two distinct phenomena in TaSb$_2$ is a rather remarkable observation. It implies that the resistivity plateau may be a generic feature of a wide class of topological materials possessing metallic surface states even in the absence of TRS. It also raises a challenge to understand the different fates of bulk Fermi surfaces, Dirac cone excitations, as well as metallic surface states in the presence of magnetic field. All these novel features, together with the field-induced metal-insulator transition, the XMR and the high carrier mobilities, suggest TaSb$_2$ as an interesting new platform of topological materials for future theoretical investigations and device applications.

**Methods**

High quality single crystals of TaSb$_2$ were grown via chemical vapor transport reaction using iodine as transport agent. Polycrystalline samples of TaSb$_2$ have been first synthesized by solid state reaction using high purified Tantalum powders and Antimony powders in a sealed quartz tube.
The final powders were ground thoroughly, and then were sealed in a quartz tube with a transport agent iodine concentration of 10 mg/cm³. The single crystals TaSb₂ were grown by a chemical vapor transport in a temperature gradient of 120 °C between 1120 °C - 1000 °C for 1-2 weeks. X-ray diffraction patterns were obtained using a D/Max-rA diffractometer with CuKα radiation and a graphite monochromator at the room temperature. The single crystal X-ray diffraction determines the crystal grown orientation. The composition of the crystals were obtained by energy dispersive X-ray (EDX) spectroscopy. No iodine impurity can be detected in these single crystals.

The (magneto)resistivity and Hall coefficient measurements were performed with a standard four-terminal method covering temperature range from 2 to 300 K in a commercial Quantum Design PPMS-9 system with a torque insert. The deviation of MR and Hall measurements associated with misalignment of the voltage leads could be corrected by reversing the direction of the magnetic field. The high magnetic field resistivity and Shubnikov-de Haas oscillations were measured up to 31 T at Hefei High Magnetic Field Laboratory. To check those experimental data, we performed resistivity and Hall effect measurements in both sample 1 and sample 2 with different RRR (See SI). The sample 2 with lower RRR also shows the same experimental results (See SI).

The electronic structure calculations were performed in the framework of density functional theory using the Vienna Abinitio Simulation Package (VASP) with projected augmented wave (PAW) approximation and Perdew Burke Ernzerhoff (PBE) flavor of the generalized gradient approximation (GGA). A 400 eV plane-wave energy cut-off and a 8 × 8 × 5 Γ-centered K-grid was chosen to ensure the convergence to 1 meV/atom. The Fermi surface was then obtained by extrapolating the DFT band structure to a dense K-grid of 100 × 100 × 100. The topological indices were
calculated following the parity-check method\textsuperscript{[2]} proposed by Fu et al..

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**Author Contributions**  Y. Li designed the research. L. Li synthesized the samples. L. Li and Y. Li performed most measurements. J. Wang and T. Wang performed part of measurements. C. Xi performed the SdH oscillations measurements at high magnetic fields. C. Cao performed the band structure calculations. C. Cao, J. Dai, Y. Li, and X. Xu discussed the data, interpreted the results, and wrote the paper.

**Competing Interests**  The authors declare no competing financial interests.

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Figure 1  Temperature dependence of resistivity in sample 1 for TaSb$_2$.  a, Resistivity of TaSb$_2$ as a function of temperature in several magnetic fields ($B = 1, 2, 3, 5, 7, 9$ T.) perpendicular to the current. The b shows a clear plateau resistivity at low temperature above 2 T.  c, Temperature dependence of $\partial\rho/\partial T$ at several magnetic fields. The peak in $\partial\rho/\partial T$ is defined as the inflection point $T_i$ at each field. The sign change in $\partial\rho/\partial T$ symbolizes the resistivity minimum at $T_m$. Inset shows $T_m$ and $T_i$ vs. magnetic fields. The dashed lines guide your eyes.  d, Log($\rho$) plotted as a function of the inverse temperature used to extract the activation gap ($E_g$) in TaSb$_2$. The cyan lines show the region of the linear fits. The inset shows field dependence of the activation gap values ($E_g$). Dashed line shows the linear behavior $E_g \propto B^{1/2}$.

Figure 2  Magnetic field dependence of MR in TaSb$_2$ single crystal.  a, the upper panel: Magnetoresistance (MR = $(\rho_{xx}(H) - \rho_{xx}(0))/\rho_{xx}(0)$%) versus magnetic fields along the c-axis at different temperatures as $B \perp I \parallel b$. The low panel: MR vs. fields for $B \parallel I \parallel b$.  b, The upper panel: MR plotted as a function of magnetic fields at different angles between $B$ and $I$. The low panel: the large and unsaturated negative MR emerges in a narrow window of angle around $\theta = 0^\circ$.

Figure 3  Hall effect and quantum oscillations in sample 1 for TaSb$_2$.  a, Magnetic field dependence of Hall resistivity at several different temperatures up to 9 T. The inset shows the Hall coefficient vs. temperatures.  b, The oscillation vs. high magnetic field up to 30 T at several temperatures. The inset shows the oscillatory $\Delta\rho$ vs. inverse field at 1.8,
4.2 and 8 K within the resistivity plateau regime.  

\( c \), The Fast Fourier Transform (FFT) spectrum of \( \Delta \rho \) at 1.8, 4.2 and 8 K, showing two oscillation frequencies \( F_\alpha = 220 \) T and \( F_\beta = 465 \) T.  

\( d \), Landau level index plots inverse B versus n. The peak position in \( \Delta \rho \) is assigned as the integer indices.
