

Quantum transport properties of β -Bi₄I₄ near and well beyond the extreme quantum limit

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We have investigated the magnetotransport properties of β -Bi₄I₄ bulk crystal, which was recently theoretically proposed and experimentally demonstrated to be a topological insulator. At low temperature T and magnetic field \mathbf{B} , a series of Shubnikov–De Haas (SdH) oscillations is observed on the magnetoresistivity (MR). Detailed analysis reveals a light cyclotron mass of $0.1 m_e$, and the field angle dependence of MR reveals that the SdH oscillations originate from a convex Fermi surface. In the extreme quantum limit (EQL) region, there is a metal-insulator transition occurring soon after the EQL. We perform scaling analysis, and all the isotherms fall onto a universal scaling with a fitted critical exponent $\zeta \approx 6.5$. The enormous value of critical exponent ζ implies this insulating quantum phase originated from strong electron-electron interactions in high fields. However, in the far end of EQL, both the longitudinal and Hall resistivity increase exponentially with \mathbf{B} , and the temperature dependence of the MR reveals an energy gap induced by the high magnetic field, signifying a magnetic freeze-out effect. Our findings indicate that bulk β -Bi₄I₄ is an excellent candidate for a three-dimensional topological system for exploring EQL physics and relevant exotic quantum phases.

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I. INTRODUCTION

Topological insulators (TIs) are quantum materials that have insulating bulk states and metallic surface states consisting of massless Dirac fermions [1–6]. The surface states are robust against time-reversal invariant perturbations and could lead to many interesting transport phenomena such as weak antilocalization [7–9, 1–3], quantum oscillations [10–12], and the Aharonov-Bohm effect [13–15]. By far, bismuth-based binary or ternary compounds such as three-dimensional (3D) Bi_{1-x}Sb_x, quasi-2D Bi₂Se₃, Bi₂Te₃, and Bi₂Te₂Se have become the essential class of TIs. These TIs have been studied extensively in past years and continue to attract research interest, due to promising both rich novel physics and potential applications in spintronics and quantum computation.

Recently, a new topological insulator β -Bi₄I₄ was theoretically predicted and experimentally confirmed [16–18]. However, angle-resolved photoemission spectroscopy (ARPES) experiments give different results about the topological classification of β -Bi₄I₄. Autès *et al.* performed the measurements on the (001) surface and revealed a single Dirac cone located at the M point of the surface Brillouin zone [17], confirming β -Bi₄I₄ as a strong TI. But in a recent work, Noguchi *et al.* reported that only the side surface [the (100) plane] hosts the Dirac surface state, indicate that β -Bi₄I₄ is a weak TI [18].

Despite the ambiguity in its classification, β -Bi₄I₄ has shown many fascinating transport properties. Hysteretic profile forms at around 300 K on the resistivity-temperature curve, indicating a structural phase transition between β -Bi₄I₄ and α -Bi₄I₄ [18]. The theoretical calculations and the experiments found a pressure-induced superconducting phase and multistructural or topological phase transitions in β -Bi₄I₄ [19–22]. The first-principles calculations found that β -Bi₄I₄ has a bulk electronic structure very similar to that of α -Bi₄I₄, with a 3D Fermi surface in the valence band and a two-dimensional (2D) Fermi surface in the conduction band [23].

In this work, we carried out detailed studies on the angular and temperature dependence of magnetotransport properties of β -Bi₄I₄ with the steady and extrahigh pulsed magnetic field. Through SdH oscillations, we obtained a small cyclotron mass of $0.1 m_e$, and a 3D angle mapping for magnetoresistivity (MR) reveals a convex Fermi surface. With the entrance of the extreme quantum limit (EQL), there occurs a metal-insulator transition. In the region far away from the EQL, the MR increases exponentially with regard to \mathbf{B} , and a field-induced effective gap is observed. Our findings indicate that β -Bi₄I₄ is possibly an ideal platform for exploring quantum limit physics and relevant exotic phases.

II. EXPERIMENTAL SETUP

Single crystals of β -Bi₄I₄ were grown using the methods described in Ref. [17]. Starting materials Bi and HgI₂ powder with mole ratio 1:2 were mixed and finely ground in a

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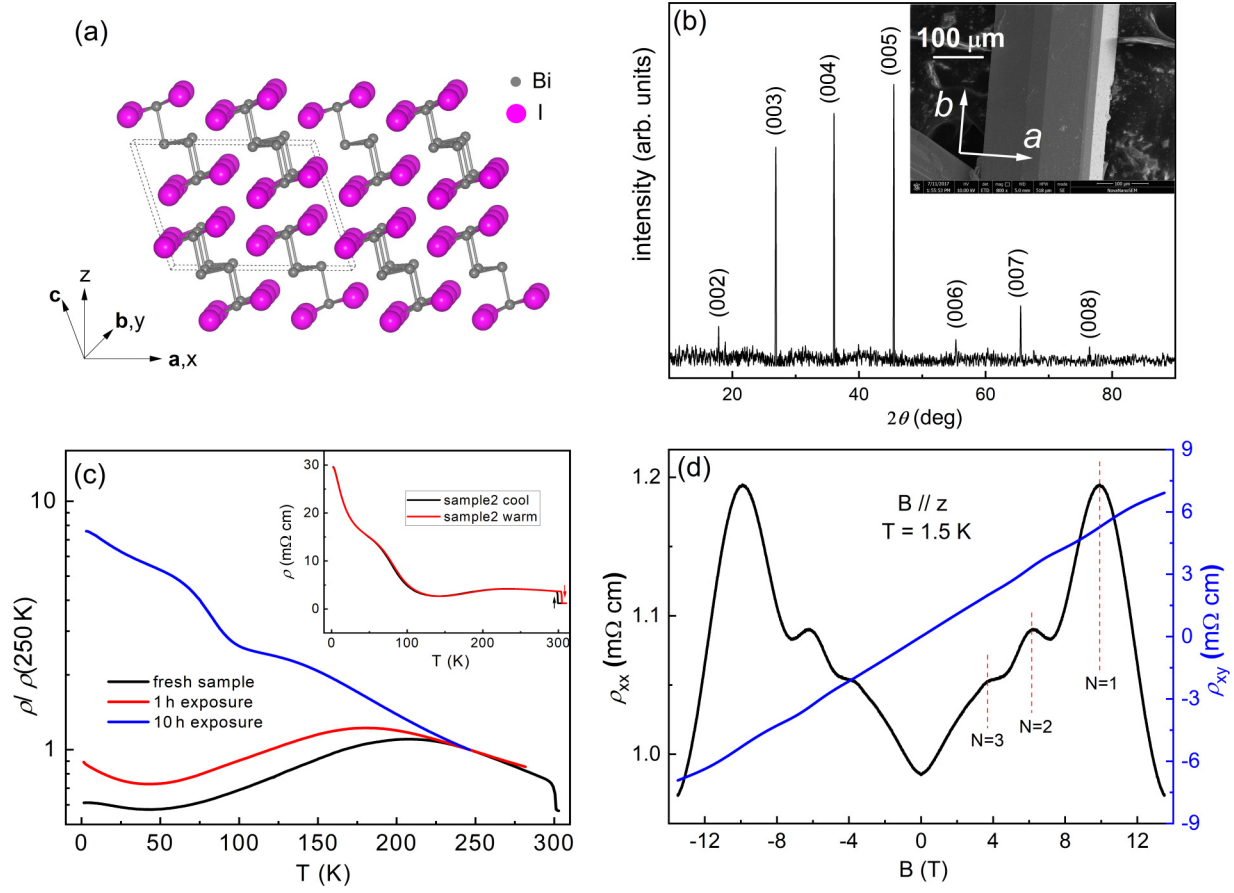


FIG. 1. (a) The β - Bi_4I_4 crystal in space group $C12/m1$ (No.12), with lattice parameters $a = 14.353 \text{ \AA}$, $b = 4.423 \text{ \AA}$, $c = 10.476 \text{ \AA}$, and $\beta = 107.8^\circ$ between the a and c axes. Pink and gray spheres represent the bismuth and iodine ions, respectively, and the gray dashed box shows the unit cell. (b) The XRD pattern of β - Bi_4I_4 single crystals with $(00l)$ reflections. Inset: A scanning electron microscopy (SEM) image of a ribbon-shaped sample usually has quite irregular facets. (c) Temperature dependence of ρ_{xx} curves normalized to ρ (250 K) for sample 1 with different exposure times in the air. The fresh sample refers to the sample with the least exposure time, which was measured immediately after the contacts were made. Upper inset: Room temperature curves of sample 2 show the insulating behavior of the α phase. A hysteresis profile can be seen at 300 K. (d) ρ_{xx} and ρ_{xy} of sample 1 taken at 1.5 K; the magnetic field is parallel to the z direction.

glove box, then loaded in a quartz tube 300 mm long and 16 mm in diameter. The quartz tube was flame sealed under high dynamic vacuum and placed into a two-zone furnace. A temperature gradient of 250°C – 210°C was applied. After 20–30 days, the quartz tube was taken out and cooled in the air. Needlelike crystals with typical size $5 \text{ mm} \times 0.3 \text{ mm} \times 0.2 \text{ mm}$ were obtained. The crystal structures and face index were examined by powder x-ray diffraction recorded by Rigaku SmartLab 9 KW with $\text{Cu } K\alpha$ radiation ($\lambda = 1.5406 \text{ \AA}$); average stoichiometry was determined by energy-dispersive x-ray spectroscopy. The transport properties under magnetic field were carried out in an Oxford TeslatronPT with temperature variable from 1.5 to 300 K and magnetic field up to 14 T. The 52 T pulsed magnetic field measurements were performed at Wuhan National High Magnetic Field Center. Before making the electrical connections, the crystal surface was cleaned by argon plasma followed by a deposition of 5/30 nm thick Cr/Au Hall bar pattern through a mask. Both the surface cleaning and Cr/Au deposition are crucial to the measurements, and it ensures the Ohmic contacts with a typical contact resistance of $\sim 1 \text{ } \Omega$. By contrast, directly using silver paste resulted in large contact resistance of 0.2–2 k Ω , which increases further

with time. This is possible because the surfaces of β - Bi_4I_4 are highly susceptible to the influence of moisture and organic solvent.

III. RESULTS AND DISCUSSION

β - Bi_4I_4 has a quasi-one-dimensional (1D) structure, see Fig. 1(a), with Bi-I molecular chains stacked along both the a and c axes through weak noncovalent interactions. The chain's direction is along axis b , as indicated in the inset of Fig. 1(b). The average atomic ratio determined from energy-dispersive x-ray spectroscopy is Bi:I = 0.51:0.49, and no traces of mercury were detected on the cleaved surfaces. The structure of β - Bi_4I_4 is very similar to that of α - Bi_4I_4 . One crucial feature which distinguishes the α phase is the presence of (005) and (007) reflection peaks located at $2\theta \approx 22.3^\circ$ and $2\theta \approx 31.5^\circ$ [18,23]. Figure 1(b) shows that none of the two peaks is observed, confirming that our crystals are β - Bi_4I_4 . Another feature that distinguishes these two phases is the temperature-dependent resistivity, with the α phase being much more resistive [18]. Figure 1(c) presents a typical curve of resistivity ρ as a function of temperature T for our β - Bi_4I_4 crystals.

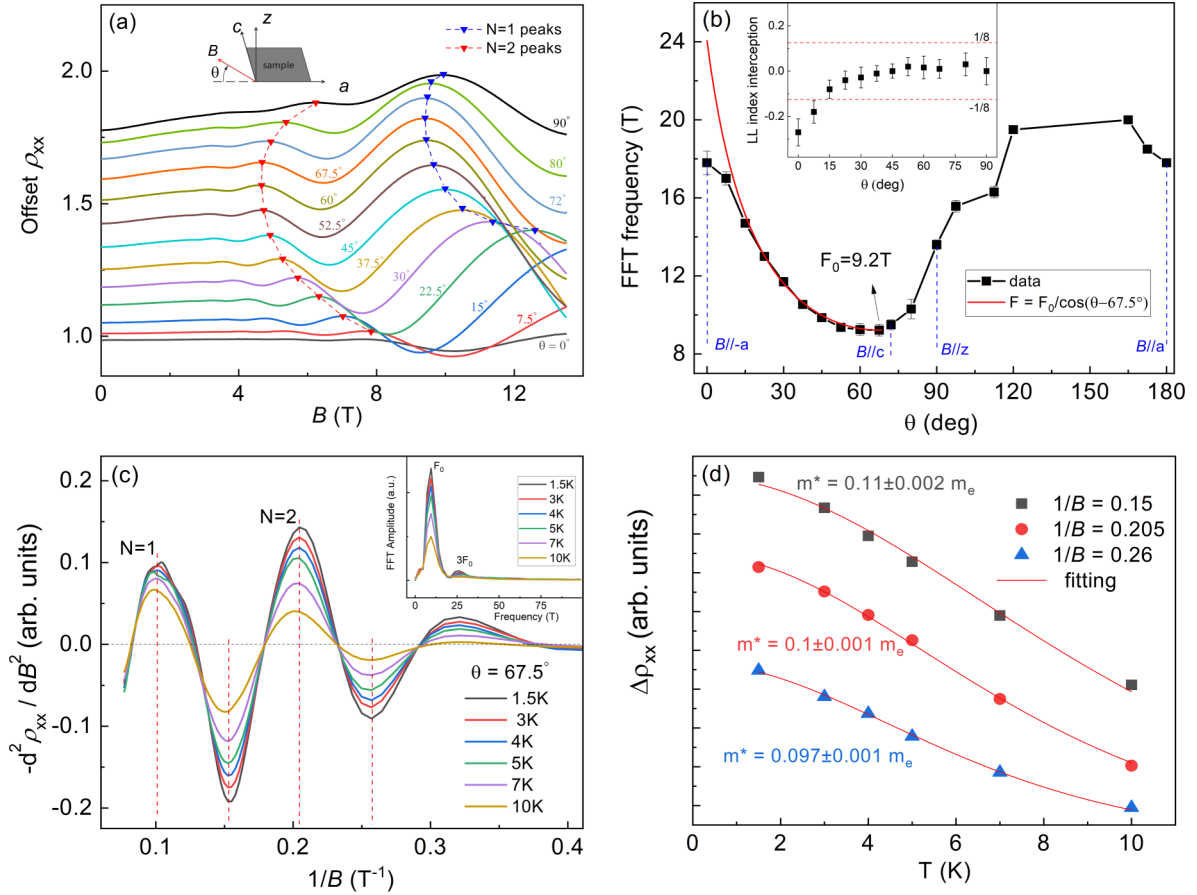


FIG. 2. (a) The angle dependence of the SdH oscillations from $\theta = 0^\circ$ to $\theta = 90^\circ$; the blue and red triangles mark the $N = 1$ and $N = 2$ Landau level peaks, respectively. (b) The obtained oscillation frequency as a function of θ from 0° to 180° . The red line draws the angle dependence of the frequency for a 2D Fermi surface; F_0 corresponds to the minimal cross-section area at $\theta = 67.5^\circ$. The inset shows the interception extracted from the Landau level (LL) fan diagram. (c) $-d^2 \rho_{xx} / dB^2$ as a function of $1/B$, at different temperatures ranging between 1.5 and 10 K. The inset shows the FFT transform of the data in the main panel. (d) The normalized oscillation amplitudes as a function of temperature, taken in different fields. The red line shows the fitting of the effective cyclotron mass following Lifshitz-Kosevich theory.

As T decreases from room temperature, the resistivity of the freshly prepared samples (black line) usually forms a wide peak centered at around 200–220 K, followed by a metallic behavior down to 50 K, then increases again until the lowest temperature. As a comparison, we also present a $\rho(T)$ curve of another sample in the inset, a clear hysteretic loop at 300 K, and the much more resistive behavior below 200 K indicates the formation of α - Bi_4I_4 after the phase transition at 300 K. We also found that resistivity is readily influenced by the environment. After warming up to 300 K and being exposed to the air for a period, the $\rho(T)$ changes with the peak shifts to lower temperature and the resistivity at low T increases. The total 10 h of exposure (blue line) resulted in a semiconductor behavior quite similar to the α -phase Bi_4I_4 . We speculate that the reaction of iodine with moisture may change the surface conductivity.

We further characterize the sample with magnetotransport measurements. As shown in Fig. 1(d), when the magnetic field is applied along the z direction, the longitudinal ρ_{xx} shows clear SdH oscillations. At least three peaks can be distinguished, corresponding to the Landau level index $N = 3, 2$, and 1. Another prominent feature is that the ρ_{xx} decrease sharply above $B = 10.4$ T, to a negative MR region. A

simple fast Fourier transform (FFT) analysis yields a single frequency, indicating that only one pocket is on the Fermi surface. The single-band nature is further confirmed by the Hall resistivity ρ_{xy} , which shows a linear behavior from -13.5 to 13.5 T. The bulk carrier density n and the mobility μ at 1.5 K are obtained from the linear fitting of $\rho_{xy} = B/ne$ and via $\sigma_0 = ne\mu$, which yield $n = 1.2 \times 10^{18}/\text{cm}^3$ (hole type) and $\mu = 5400 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$.

To understand the nature of the SdH oscillations, we performed magnetic field angle dependent measurements to tracked how the SdH oscillations shift with θ (the tilted angle between the magnetic field and the $-a$ axis). Figure 2(a) presents the ρ_{xx} at different angles θ ; here all the curves have been shifted for clarity. As θ increases from 0 (where $B \parallel -a$), the fields of $N = 1$ and $N = 2$ peaks decrease until $\theta = 67.5^\circ$, then began to increase. The minimal angle 67.5° is very close to 72° between the (001) and (100) planes of β - Bi_4I_4 . After an FFT analysis, the corresponding frequency as a function of θ is shown in Fig. 2(b). According to the Onsager relation $F = S_F \hbar / 2\pi e$, the oscillation frequency is proportional to the extremal cross-section area of the Fermi surface S_F . For a two-dimensional (2D) Fermi surface, the oscillation frequency depends only on the field component

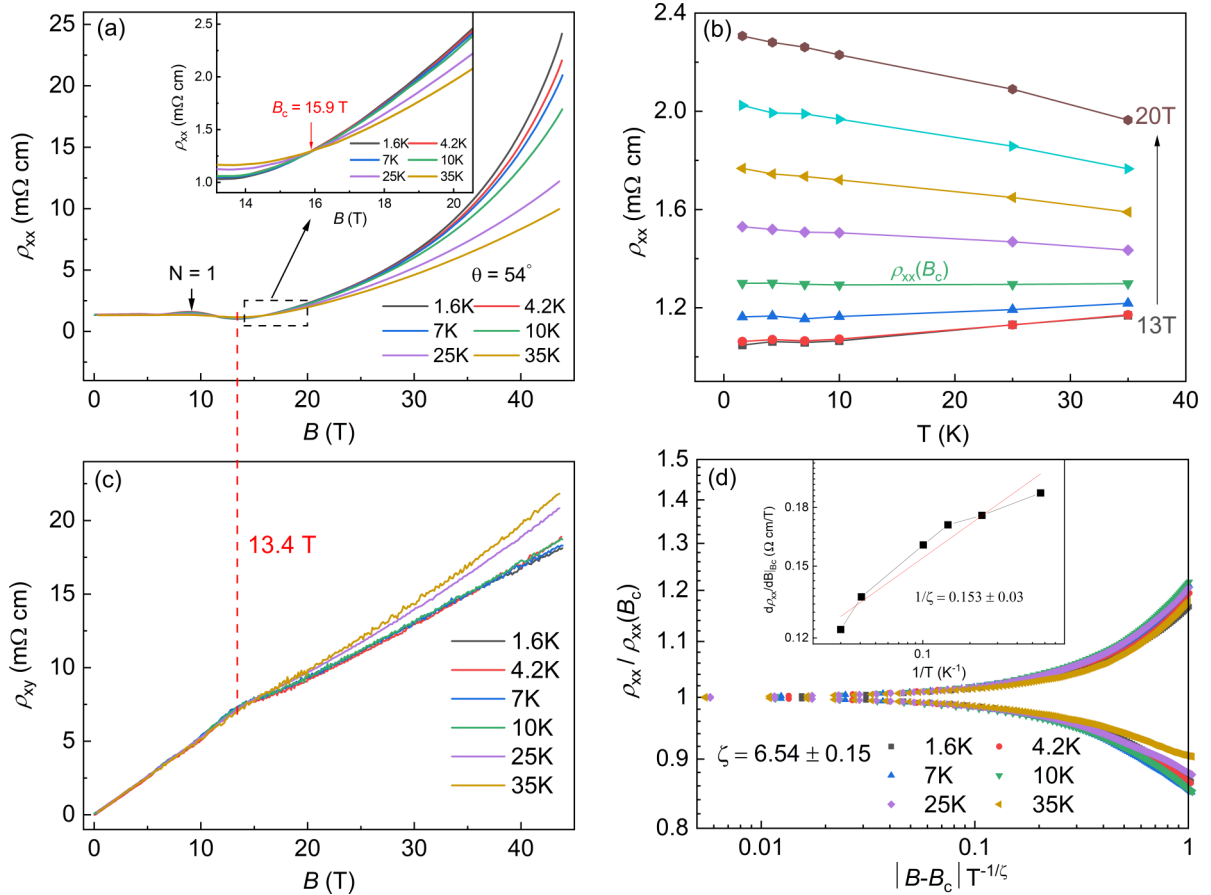


FIG. 3. (a) The field dependence of ρ_{xx} up to 43 T under different temperatures. The red dashed line marks the ending of the quantum oscillation, above which the system enters the EQL. The inset shows the metal to insulator transition happens at a critical field of 15.9 T. (b) Plot of the ρ_{xx} as a function of temperature at fixed magnetic field B . (c) The field dependence of ρ_{xy} up to 43 T. (d) Scaling plot of the normalized ρ_{xx} ; all the isotherms fall onto a nearly single curve as a function of $|B - B_c|T^{-1/\zeta}$ with $\zeta \approx 6.5$; the inset shows the linear fitting of the $\log(d\rho_{xx}/dB)$ at B_c vs $\log(1/T)$, which yields $1/\zeta$.

$B = B_0 \cos \theta$. As θ increase, the frequency will increase the following relation of $F(\theta) = F_0 / \cos \theta$. Choosing the minimal frequency as F_0 when $\theta = 67.5^\circ$ (which is 9.2 T), we have plotted the $F_0 / \cos(\theta - 67.5^\circ)$ in Fig. 2(b), as the red line shows. The measured data fit with the cosine relation only when $15^\circ \leq \theta \leq 67.5^\circ$, indicating that the oscillations originate from a 3D Fermi surface rather than a 2D, but with a convex shape. This is consistent with the first-principles calculation [23]. Fixing the angle at 67.5° , if we assume a circular Fermi surface cross section, the k_F can be calculated from $S_F = \pi k_F^2$, which is $k_F = 1.7 \times 10^{-2} \text{ \AA}^{-1}$. The inset in Figure 2(b) shows the interceptions obtained from the Landau level fan diagram, from which the Berry's phase can be determined. In a 3D system, an interception in the range of $\pm 1/8$ implies a nontrivial Berry's phase of π , and it is trivial otherwise. As the magnetic field tilted from $B//a$, the Berry's phase becomes nontrivial when $\theta \geq 15^\circ$, in accordance with the ARPES results in Ref. [18], where the surface Dirac cone is found at the side surface [(100) surface] of the crystal.

After subtracting the background of R_{xx} or ρ_{xx} by taking a second derivative, the SdH amplitudes taken at different temperatures as a function of reciprocal B are presented in Fig. 2(c), along with the FFT spectrum. The cyclotron mass of carrier can be obtained by fitting the temperature dependence

of the SdH amplitudes following Lifshitz-Kosevich (LK) theory:

$$A(T) = A_0 \frac{2\pi^2 k_B T / \hbar \omega_c}{\sinh(2\pi^2 k_B T / \hbar \omega_c)},$$

where k_B is the Boltzmann's constant, and ω_c is the cyclotron frequency $\omega_c = eB/m^*$. The effective cyclotron mass m^* is determined to be $0.1m_0$ (where m_0 is the free electron mass) in Fig. 2(d).

Such a small effective mass implies large cyclotron energy $\hbar \omega_c$ in magnetic fields. When the field B is high enough that the criterion $\hbar \omega_c \gg E_F \gg k_B T$ is satisfied, the system enters the EQL where all the electrons occupy only the lowest Landau level. Experimentally, the EQL can be identified by the last peak of quantum oscillations on resistivity or thermoelectric curves [24,25]. In Fig. 1(d), ρ_{xx} decreases sharply after the $N = 1$ peak at 10 T, suggesting that the system approaches the EQL. To investigate the quantum phases of $\beta\text{-Bi}_4\text{I}_4$ beyond the EQL, we further carried out the pulsed high magnetic field transport measurements up to 43 T, where the direction of magnetic field is pointed around the c axis, as shown in Figs. 3(a) and 3(c). The quantum oscillations end up at 13.4 T with the ρ_{xx} reaching the minima. Above 13.4 T, the ρ_{xx} begins to increase monotonically without saturate; meanwhile

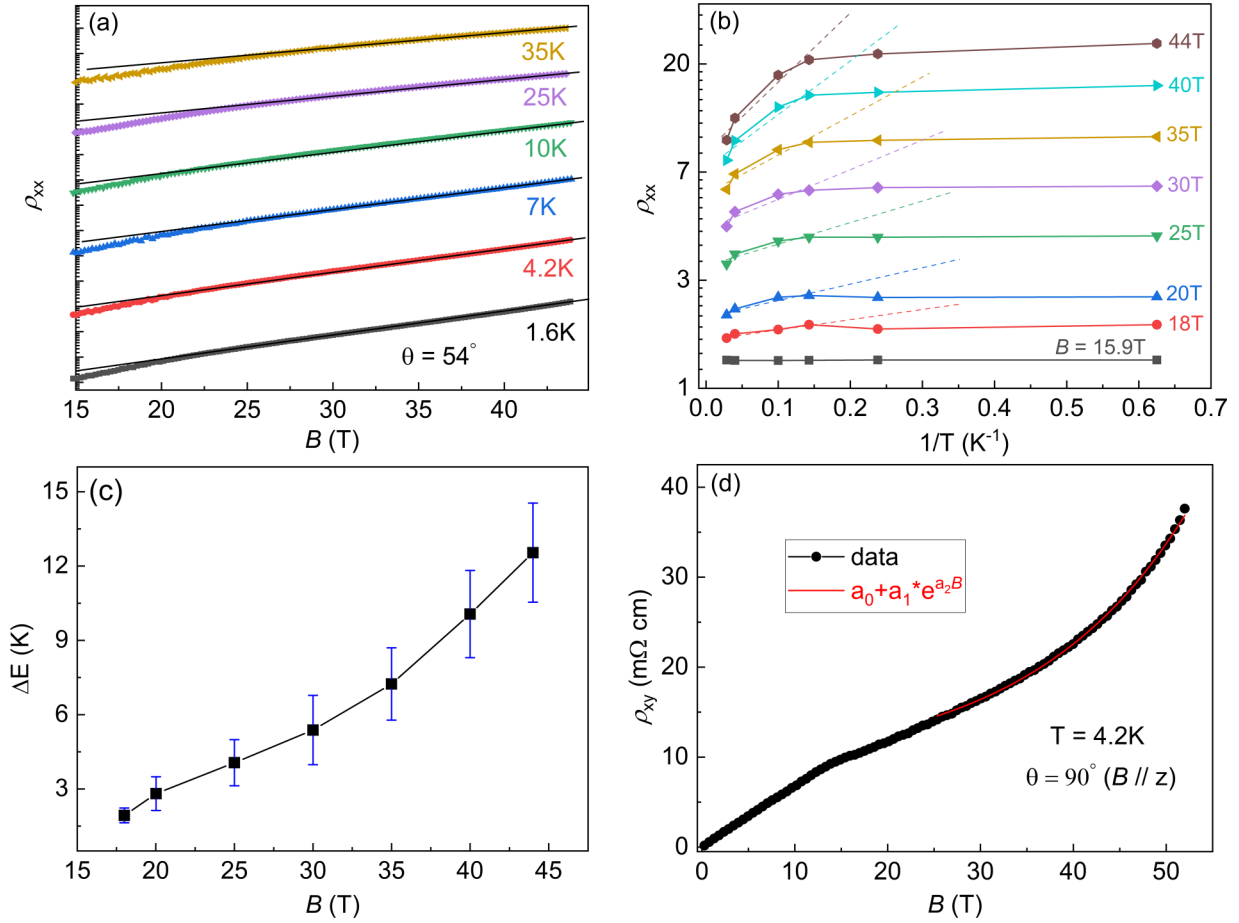


FIG. 4. (a) $\ln(\rho_{xx})$ as a function of field B up to 43 T; the curves have been shifted for clarity. The linear behavior above 20 T demonstrates the exponential increase of ρ_{xx} . (b) The Arrhenius plot of the temperature dependence of ρ_{xx} under fixed magnetic field ranging from 15.9 to 43 T. (c) The energy gap is relative to a function of B , and it increases almost linearly with the field. (d) The Hall resistivity up to 52 T at 4.2 K, where the field is parallel to the z direction. Under high field, ρ_{xx} also increase exponentially with B ; the red line draws a simple e^B fitting.

the Hall resistivity ρ_{xy} changes its behavior from temperature independent to temperature dependent. We believe that both the sign change in ρ_{xx} from negative to positive and the slope change in ρ_{xy} indicate the system enters the EQL at 13.4 T, where the Fermi energy crosses only the lowest Landau level band $N = 0$ and lies right below the bottom of the band $N = 1$ [26]. As the magnetic field increases further, there exists a critical field of $B_c = 15.9$ T where all the ρ_{xx} curves taken at different temperatures share a common crossing point [Fig. 3(a), inset]. We also plot ρ_{xx} as a function of temperature T with fixed B ranging from 13 to 20 T, as shown in Fig. 3(b). Below the critical field B_c , the ρ_{xx} decreases monotonically as T decreases, while it increases when the field is above B_c . This profile signals a metal-insulator transition at the critical field: The system is metallic below B_c and insulating above B_c . Typically, for a magnetic field induced metal-insulator transition, the scaling relation of the resistivity to temperature and magnetic field can be described by [27,28]

$$\rho(B, T) = \rho(B_c) f[|B - B_c| T^{-1/\zeta}],$$

where $f(x)$ is a scaling function with $f(0) = 1$, and ζ is the critical exponent which can be determined by evaluating the inverse slope of the log-log plot of $\frac{d\rho_{xx}}{dB}|_{B_c}$ vs $1/T$ [29].

With B_c and ζ in hand, the scaling function can be directly tested without applying another fitting parameter. In Fig. 3(d), we performed such a scaling analysis, and indeed all the isotherms fall onto a nearly single curve as a function of $|B - B_c| T^{-1/\zeta}$. In the inset of Fig. 3(d), we obtained $1/\zeta = 0.153 \pm 0.03$ from the linear fitting of the data; this yields the critical exponent of $\zeta \approx 6.5$. According to a recent theory which describes the field-induced metal-insulator transition in 3D systems [30], a large critical exponent $\zeta \approx 6$ implies that strong electron-electron interactions may be involved at high fields.

In Fig. 4(a), however, with a much higher magnetic field B (>25 T), the ρ_{xx} grows exponentially with increasing B . We have plotted the logarithm resistivity $\ln(\rho_{xx})$ as a function of B under various temperatures (the curves have been shifted for clarity). It can be clearly seen that $\ln(\rho_{xx})$ increases linearly with B from 20–25 T to 43 T, depending on the sample temperature. This exponential increasing ρ_{xx} is quite different from those B^2 , B , or \sqrt{B} relations as usually seen in topological semimetals under high fields [31]. This new insulator region can be attributed to the magnetic freeze-out effect that led to a reduced carrier concentration. Typically, the magnetic freeze-out arises from the electron localization

in the extrinsic hopping conductivity region [32,33]. At low temperatures, the electrons are localized at impurity or dopant ions, and the conduction mechanism in these materials is variable range hopping between the impurity or dopant. The high magnetic field will further compress the wave functions of the localized states and reduce the overlap of the electron orbitals between neighbor ions, resulting in an exponential MR. Another mechanism for the magnetic freeze-out effect is an increasing effective energy gap with the field in an intrinsic narrow band-gap semiconductor with small carrier masses [34]. Under such conditions, at high fields where $\hbar\omega_c \gg k_B T$ holds, the carrier concentration decreases exponentially with magnetic field, $\frac{n(B)}{n(0)} \propto \exp(-\hbar\omega_c/4k_B T)$, which results in the exponentially increase of both ρ_{xx} and ρ_{xy} with field B . Although both mechanisms exhibit an exponentially increasing magnetoresistivity, they are distinguishable in transport measurements. The distinctive evidence is that the argument of the exponential for the former is quadratic [35,32], while it is linear for the latter one [34]. In Fig. 4(b) we plot the $\ln(\rho_{xx})$ as a function of inverse temperature $1/T$ under fixed field B , then fit the high temperature data to extract the effective band gap following Arrhenius plotting, $\rho_{xx} \propto \exp(\frac{\Delta}{2k_B T})$. The obtained band gap with error bars are shown in Fig. 4(c); it increases almost linearly with magnetic field from 0 K at 15.9 T to 12 K at 43 T. As a result, the concentration of the thermal carriers decreases which leads to the exponential behavior as shown in Fig. 4(a). This is further confirmed by the Hall resistivity [Fig. 4(d)], where the magnetic field up to 52 T is parallel to the z axis. A clear nonlinear profile can be seen, and the high field data (>25 T) are proportional to e^B . Our data support the magnetic freeze-out effect of the carrier concentration mechanism well beyond the EQL. However, as the impurities and defects are inherently present in crystals, both an intrinsic

and extrinsic mechanism could be involved simultaneously in a particular sample. Further investigations are needed for better understanding in the near region of EQL.

IV. CONCLUSION

In summary, we have investigated the magnetotransport properties of a small band-gap topological insulator β -Bi₄I₄ in a magnetic field up to 43 T. There exists a critical field $B_c = 15.9$ T where metal to insulator transition happens. Below B_c the MR shows clear SdH oscillations, which reveals a 3D convex Fermi surface and a small effective hole mass of $0.1m_e$. In the near region of EQL, there is a metal-insulator transition around B_c . Furthermore, the result of scaling analysis suggests that it results in a strong electron-electron transition. Far away from the EQL, both the longitudinal and Hall resistivity increase exponentially with field B . The temperature dependence of the MR reveals an energy gap that increases linearly with field B , which signifies the magnetic freeze-out of carrier concentration, where the system has a narrow bulk energy gap. The extract mechanisms of the transitions observed here still require further investigation.

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