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# Quantized plateau in the thermoelectric Hall conductivity for Dirac electrons in the extreme quantum limit

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**When subjected to a sufficiently strong magnetic field, the properties of electronic quantum matter change dramatically. This is especially true in the “extreme quantum limit” (EQL), in which the spacing between quantized energy levels of the electron cyclotron motion becomes larger than any other relevant energy scale. Here we probe deeply into the EQL in the three-dimensional Dirac semimetal ZrTe<sub>5</sub>. We measure the bulk thermoelectric properties, and**

**we find them to be greatly enhanced over their zero-field values. Most strikingly, the thermoelectric Hall conductivity  $\alpha_{xy}$  acquires a universal, quantized value deep in the EQL, which is independent of magnetic field or carrier concentration. We explain this quantization theoretically and show how it is a unique signature of three-dimensional Dirac or Weyl electrons in the EQL.**

Strong magnetic fields can have a profound influence on quantum matter. In electronic systems, this influence arises primarily from the Lorentz force, which bends the trajectories of itinerant electrons and forces them to turn in tight cyclotron orbits. These orbits are quantized into discrete energy levels (Landau levels) in an analogous way to the quantization of electron orbitals within an atom. When the magnetic field is sufficiently strong, a many-electron system enters the “extreme quantum limit” (EQL), in which the spacing between Landau levels becomes larger than any other energy scale in the problem, such as the Fermi energy  $E_F$  or the thermal energy  $k_B T$ . In this limit all itinerant electrons occupy only the lowest Landau level, which means that their motion in the plane perpendicular to the magnetic field is locked into a single, highly degenerate quantum state, while only the electron motion parallel to the magnetic field remains free.

In typical metals, the Fermi energy is so large that achieving the EQL requires enormous magnetic fields on the order of  $\sim 10^5$  T. Such large fields seem to exist only in astrophysical scenarios such as neutron stars, [1] and are implausible for laboratory experiments. In doped semiconductors one can achieve low Fermi energy, but electrons in semiconductors are prone to localization effects, which arise from the disorder that is inevitably introduced by doping. [2] Since strong magnetic field tends to facilitate electron localization, the EQL can generally be probed in doped semiconductors only over a limited range of field (see, *e.g.*, [3, 4, 5, 6]). These limitations can be circumvented, however, by studying the EQL in three-dimensional *nodal semimetals*, such as the recently-discovered Dirac or Weyl semimetals. [7] In such materials

the Fermi energy can, in principle, be made arbitrarily small, while the absence of a band gap prevents electron localization.

It was recently suggested theoretically that the thermopower of nodal semimetals can be enormously enhanced in the EQL [8], which implies a potential new pathway for achieving efficient platforms for waste heat recovery or solid state refrigeration. [9] In a follow-up work, it was shown that this result can be understood in terms of a quantized value of the thermoelectric Hall conductivity [10]. Motivated by these predictions, in this work we study the thermoelectric effect in strong magnetic fields in the Dirac semimetal  $\text{ZrTe}_5$ .

In terms of its electronic properties, bulk  $\text{ZrTe}_5$  lies somewhere between a strong topological insulator and a weak one, sensitively depending on the crystal lattice constant. [11] At this phase boundary the band structure realizes a gapless Dirac dispersion around the  $\Gamma$  point. Many previous studies have shown that  $\text{ZrTe}_5$  is a Dirac semimetal with at most a small band gap (see, e.g., [12, 13, 14, 15, 16]). Since  $\text{ZrTe}_5$  possesses a single Dirac cone and can be grown with very low carrier density and very high mobility, it represents an ideal platform for studying three-dimensional (3D) Dirac electrons in the EQL.

Our samples are single crystals of  $\text{ZrTe}_5$  grown by the Tellurium flux method [17]. We should point out that our samples are n-type and have only a single (and small) electron pocket, which is significantly different from samples grown by the chemical vapor transport method. Relatively large crystals, with a typical size of  $3 \text{ mm} \times 0.4 \text{ mm} \times 0.3 \text{ mm}$ , were used for transport measurements. The longest dimension is along the  $a$  axis and the shortest dimension is along the  $b$  axis. In our measurements, either the electrical current or the temperature gradient was applied along the  $a$  axis, while the magnetic field was perpendicular to the  $ac$  plane. For the thermoelectric measurements, one end of the sample is thermally anchored to the sample stage, while the other end is attached to a resistive heater. The temperature difference between the two ends is measured by a type-E thermocouple. This difference is in the range of 100 to

160 mK, which is always much smaller than the sample temperature.

The electron concentration in our samples, as measured by the Hall effect, is  $n_{\text{Hall}} \approx 5 \times 10^{16} \text{ cm}^{-3}$  at low temperature. As we show below, at such low densities the EQL is reached already at fields  $\gtrsim 2 \text{ T}$ . The Hall effect remains linear in the magnetic field  $B$  up to 6 T at low temperatures, which is much higher than the quantum limit field. This linearity reflects a single band of carriers, indicating that we have a simple Dirac system. As in previous studies of  $\text{ZrTe}_5$  [18, 19], the Hall concentration  $n_{\text{Hall}}$  is seen to evolve with temperature, with the sample changing from  $n$ -type to  $p$ -type as  $T$  is increased above  $\approx 83 \text{ K}$ . Previous studies suggest that this change results from a temperature-dependent Lifshitz transition [17, 19]; a more thorough discussion of the electron and hole concentrations as a function of temperature is presented in the Supplementary Information. The shift from  $n$ - to  $p$ -type transport with increasing temperature is also reflected in the behavior of the zero-field resistivity  $\rho_{xx}$  and the zero-field thermopower  $S_{xx}$  as a function of temperature, which are shown in Fig. 1a.

As the magnetic field is increased from zero, the resistivity undergoes Shubnikov-de Haas (SdH) oscillations associated with depopulation of high Landau levels. These oscillations are plotted in Fig. 1b, which show that the EQL is achieved at all fields  $> 2 \text{ T}$ . The appearance of SdH oscillations at very low field ( $\approx 0.1 \text{ T}$ ) reflects the high mobility of our samples,  $\mu \approx 640,000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ . A previous study reported measurements of the SdH oscillations for the magnetic field oriented along the  $x$ ,  $y$  and  $z$  axes, from which the Fermi surface morphology is obtained [18]. The Fermi surface is an ellipsoid with the longest principal axis in the  $z$  direction. The carrier density estimated from SdH oscillations is in good agreement with  $n_{\text{Hall}}$ , confirming the dominance of a single band at the Fermi level at low temperature. The corresponding Fermi level is only 11 meV above the Dirac point at  $T = 1.5 \text{ K}$ .

Because of the low carrier density, the system enters the lowest ( $N = 0$ ) Landau level at  $B > 2 \text{ T}$  at low temperature. As the temperature is increased, the Fermi level shifts towards the

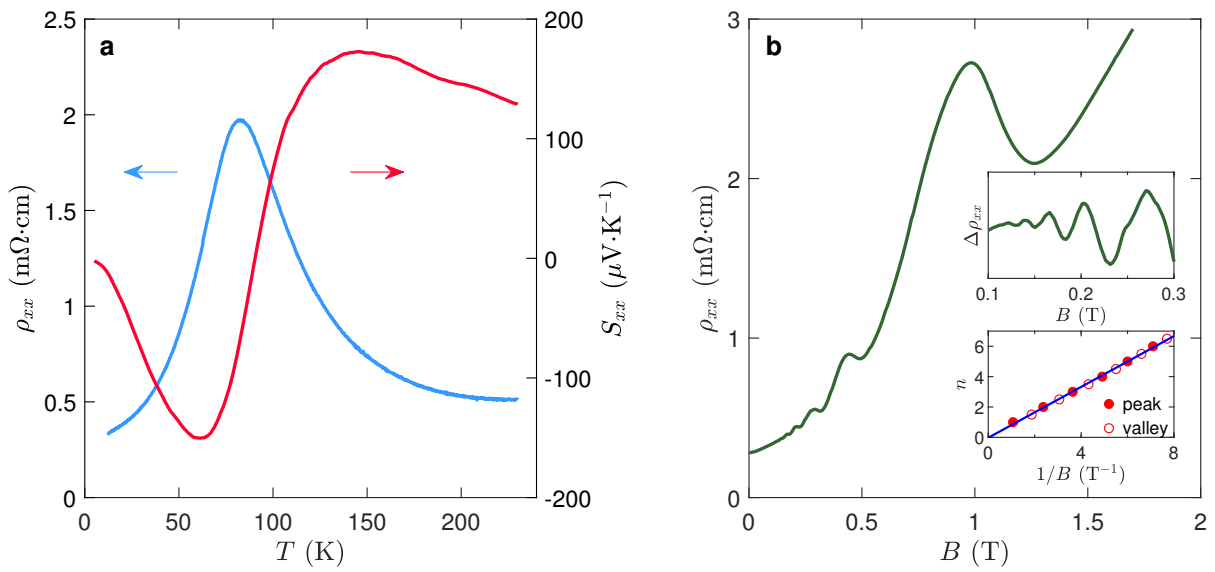


Figure 1: **a** Temperature dependence of the electrical resistivity and the thermopower. **b** Magnetoresistance at 1.5 K. On top of a strong positive magnetoresistance, SdH oscillations are evident (upper inset). The onset field of the oscillations is 0.13 T, indicating a high mobility. The system enters the EQL at  $\approx 2$  T. The lower inset shows the index  $n$  of the minima and maxima of each oscillation as a function of  $1/B$ .

Dirac point, implying that the quantum limit is reached at an even lower field. Therefore, the system is well within the EQL for a large range of magnetic field, which we sweep up to 14 T.

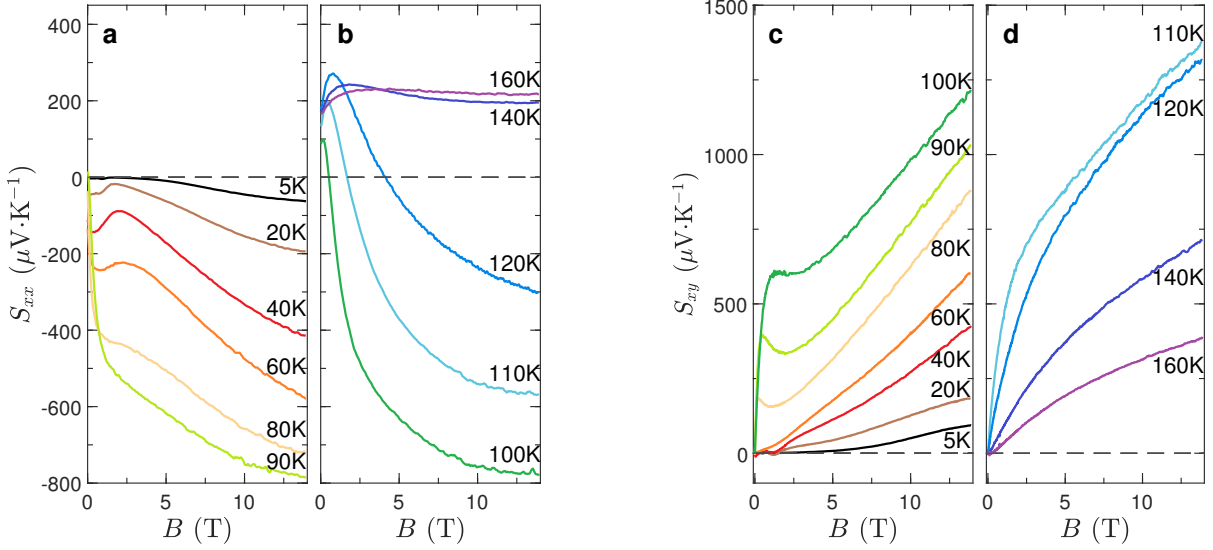


Figure 2: Longitudinal and transverse thermoelectric coefficients as a function of the magnetic field at different temperatures. **a b**  $S_{xx}$ . **c d**  $S_{xy}$ .

Our measurements of the longitudinal (Seebeck,  $S_{xx}$ ) and transverse (Nernst,  $S_{xy}$ ) thermoelectric coefficients are shown in Fig. 2 as a function of magnetic field. There is a general increase in the magnitude of both  $S_{xx}$  and  $S_{xy}$  with magnetic field in the EQL. Indeed, at  $T \approx 90$  K, where the carrier density is the lowest,  $S_{xx}$  becomes as large as  $800 \mu\text{V/K}$ , while  $S_{xy}$  becomes larger than  $1200 \mu\text{V/K}$ . The theoretical interpretation of  $S_{xx}$  and  $S_{xy}$ , however, is complicated by the variation of the carrier density with  $T$  and  $B$ . Indeed, the change in sign of  $S_{xx}$  with  $B$  at higher temperatures is likely related to the proximity of the system to a transition from  $n$ -type to  $p$ -type conduction, as is the sharp variation in  $S_{xx}$  with  $B$  at low fields and  $T \approx 90$  K. This variation of the carrier concentration with  $B$  seems to blunt the large, linear enhancement of  $S_{xx}$  with  $B$  predicted in Ref. [8] for higher temperatures.

These complications lead us to examine a more fundamental quantity, the thermoelectric

conductivity  $\hat{\alpha} = \hat{\rho}^{-1}\hat{S}$ . Here  $\hat{\rho}$  and  $\hat{S}$  denote the resistivity and thermoelectric tensors, respectively, so that both the longitudinal and transverse components of the tensor  $\hat{\alpha}$  can be deduced from our measurements. We focus, in particular, on the thermoelectric Hall conductivity  $\alpha_{xy}$ , which is plotted in Fig. 3 for the EQL. While  $\alpha_{xy}$  depends in general on both  $T$  and  $B$ , Fig. 3b shows that deep in the EQL  $\alpha_{xy}$  achieves a plateau that is independent of magnetic field. Furthermore, this plateau value of  $\alpha_{xy}$  is linear in temperature at temperatures  $T \lesssim 100$  K, which suggests that  $\alpha_{xy}/T$  is a constant in the EQL, independent of  $B$  or  $T$  (Fig. 3a).

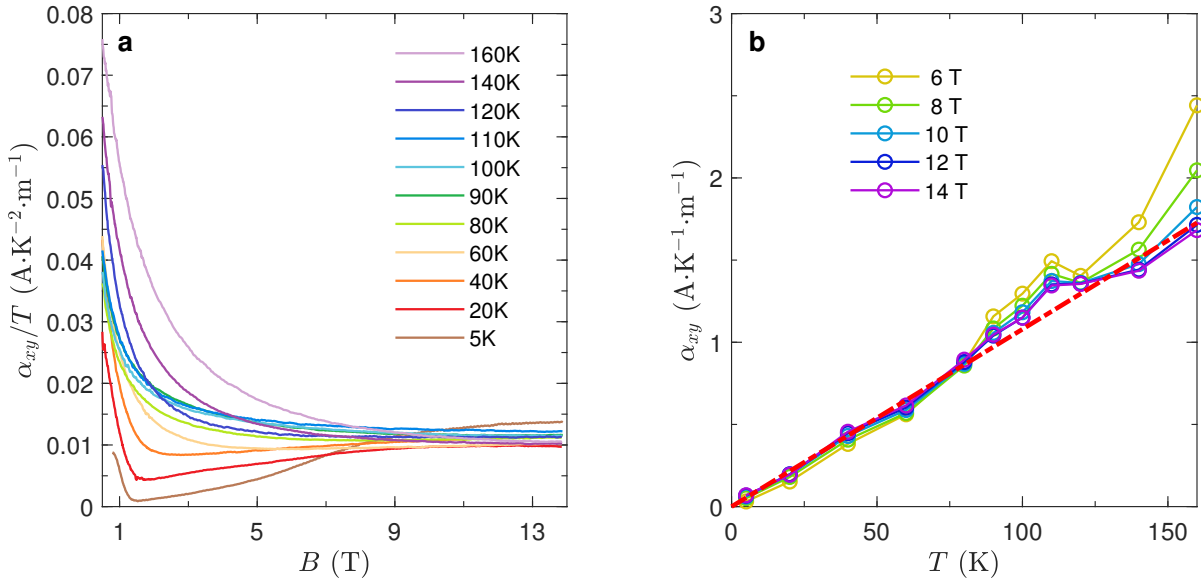


Figure 3: Transverse thermoelectric conductivity. **a**  $\alpha_{xy}/T$  as a function of  $B$  at different temperatures.  $\alpha_{xy}$  is independent of  $B$  at high fields. **b**  $\alpha_{xy}$  as a function of temperature at different fields. The red dashed line is a guide to the eye.

This strikingly universal value of  $\alpha_{xy}/T$  can be understood using the following argument. The thermoelectric Hall conductivity can be defined by  $\alpha_{xy} = J_y^Q/(TE_x)$ , where  $J_y^Q$  is the heat current density in the  $y$  direction under conditions where an electric field  $E_x$  is applied in the  $x$  direction and the temperature  $T$  is uniform. In the limit of large magnetic field (large Hall angle), electrons drift perpendicular to the electric field via the  $\vec{E} \times \vec{B}$  drift, and thus the heat



current density  $J_y^Q = v_d Q$ , where  $Q$  is the internal energy per unit volume of the electron system and  $v_d = E_y/B$  is the magnitude of the  $\vec{E} \times \vec{B}$  drift velocity. In a system with constant density of states  $\nu$ , the internal energy  $Q = (\pi^2/6)k_B^2 T^2 \nu$  [20]. Crucially, for a gapless Dirac system in the EQL, the density of states becomes an energy-independent constant,  $\nu = N_f e B / (2\pi^2 \hbar^2 v_F)$ , where  $\hbar$  is the reduced Planck constant,  $v_F$  is the Dirac velocity in the field direction, and  $N_f$  is an integer that counts the number of Dirac points multiplied by the degeneracy of each Dirac cone. (In our system  $N_f = 2$ .) Inserting this expression for  $\nu$  into the relations for  $Q$  and  $\alpha_{xy}$  gives

$$\alpha_{xy} = \frac{1}{12} \frac{T}{v_F} \frac{e k_B^2}{\hbar^2} N_f. \quad (1)$$

In other words, the value of  $\alpha_{xy}/T$  is determined only by the Dirac velocity, by the integer degeneracy factor  $N_f$ , and by fundamental constants of nature. In this sense one can say that  $\alpha_{xy} v_F / T$  is a quantized and universal quantity in the EQL of Dirac or Weyl materials. Equation (1) was predicted in Ref. [10], where it was derived in terms of quantum Hall-like edge states.

Notice, in particular, that Eq. (1) has no dependence on the carrier concentration. Thus, changes in the carrier concentration or even a transition from  $n$ -type to  $p$ -type conduction do not affect the value of  $\alpha_{xy}$ . This surprising independence apparently enables the universal plateau that we observe in  $\alpha_{xy}$ , even though the behavior of  $S_{xx}$  and  $S_{xy}$  in the EQL is more complicated. Figure 3a shows that the plateau in  $\alpha_{xy}/T \approx 0.01 \text{ AK}^{-2}\text{m}^{-2}$ . We can compare this to the theoretical prediction of Eq. (1) using the previously-measured Dirac velocity  $v_F \approx 3 \times 10^4 \text{ m/s}$  [18]. Inserting this value into Eq. (1) gives  $\alpha_{xy}/T \approx 0.015 \text{ AK}^{-2}\text{m}^{-2}$ , which is in excellent agreement with our measurement.

It is worth emphasizing that in conventional gapped systems, such as doped semiconductors,  $\alpha_{xy}$  varies with both the carrier concentration  $n$  and the magnetic field  $B$  in a nontrivial way [10]. In this sense the quantized plateau in  $\alpha_{xy}/T$  is a unique Hallmark of three-dimensional Dirac and Weyl semimetals, and it can potentially be used as a litmus test for detecting Dirac

and Weyl materials or as a method for measuring the Dirac velocity  $v_F$ .

In summary, in this Report we have demonstrated, for the first time, a universal, quantized plateau in the thermoelectric Hall conductivity of Dirac or Weyl semimetals in the EQL. This plateau is related to the large, field-enhanced thermoelectric response that we observe at strong magnetic field. Our findings imply that  $\text{ZrTe}_5$ , and three-dimensional nodal semimetals more generally, may serve as unprecedented platforms for achieving large thermopower and for exploring quantum matter in extreme magnetic field conditions.

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# Supporting Materials for “Quantized plateau in the thermoelectrical Hall conductivity for Dirac electrons in the extreme quantum limit”

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Fig. S1 shows the magnetoresistance and the Hall resistance of the sample presented in the main text. The resistivity strongly increases with field, e.g., by two to three orders of magnitude at  $B = 14$  T. Such a large magnetoresistance is typical for Dirac semimetals[S1]. At low temperatures, the Hall resistivity is rather linear below 6 T, indicating single band conduction. The linearity persists deep into the quantum limit. However, a strong deviation appears above 6 T. We believe that such a nonlinear Hall in the extreme quantum limit is the result of two-carrier contributions, as expected when the Fermi level shifts towards the Dirac point with increasing magnetic field. This shifting of the Fermi level is precisely the mechanism that gives rise to the non-saturating thermopower and transverse thermoelectric conductivity plateau, which are the main focus of this work. The low field slope of  $\rho_{xy}$  shows a sign-reversal at about 90 K, indicating the change of carrier from electron to hole, as expected by the Lifshitz transition in this material.

At temperatures that are much lower or much higher than 90 K, the Fermi level is not so close to the Dirac point and the transport is more or less one-carrier dominated at low fields. Correspondingly, the low field Hall is relatively linear and carrier density is simply calculated from the Hall slope. On the other hand, around 90 K, where the Fermi level is in the vicinity of the Dirac point, a significant amount of electrons and holes are thermally excited. The existence of two types of carriers with comparable densities causes  $\rho_{xy}$  to become non-linear even at relatively low fields. Consequently, a two-carrier model such that  $\hat{\rho}_{\text{tot}}^{-1} = \hat{\rho}_{\text{e}}^{-1} + \hat{\rho}_{\text{h}}^{-1}$ , is necessary to describe the magnetotransport and extract the carrier density. Here  $\hat{\rho}_{\text{tot}}$ ,  $\hat{\rho}_{\text{e}}$  and  $\hat{\rho}_{\text{h}}$  are the resistivity tensors of the total, electron contribution and hole contribution, respectively. For the longitudinal component of the resistivity tensor,  $\rho_{xx}$ , we need to take into account the strong magnetoresistance that a Dirac semimetal often exhibits. Since  $\rho_{xx}(B)$  at low temperatures displays a quadratic dependence when only one type of carriers dominate, it is reasonable to assume that it remains so for both electrons and holes at higher temperatures. Therefore, the following expressions are used to describe the electron and hole transport,

$$\begin{cases} \rho_{xx}(B) = \rho_0 + a|B| + bB^2 \\ \rho_{xy}(B) = R_{\text{H}}B \end{cases}, \hat{\rho} = \begin{pmatrix} \rho_{xx} & \rho_{xy} \\ -\rho_{xy} & \rho_{xx} \end{pmatrix} \quad (\text{S1})$$

By fitting data round 90 K, the carrier densities of holes and electrons,  $n_{\text{h}}$  and  $n_{\text{e}}$ , are obtained. Their difference,  $n_{\text{h}} - n_{\text{e}}$ , which reflects the Fermi level, is plotted in Fig. S1e.

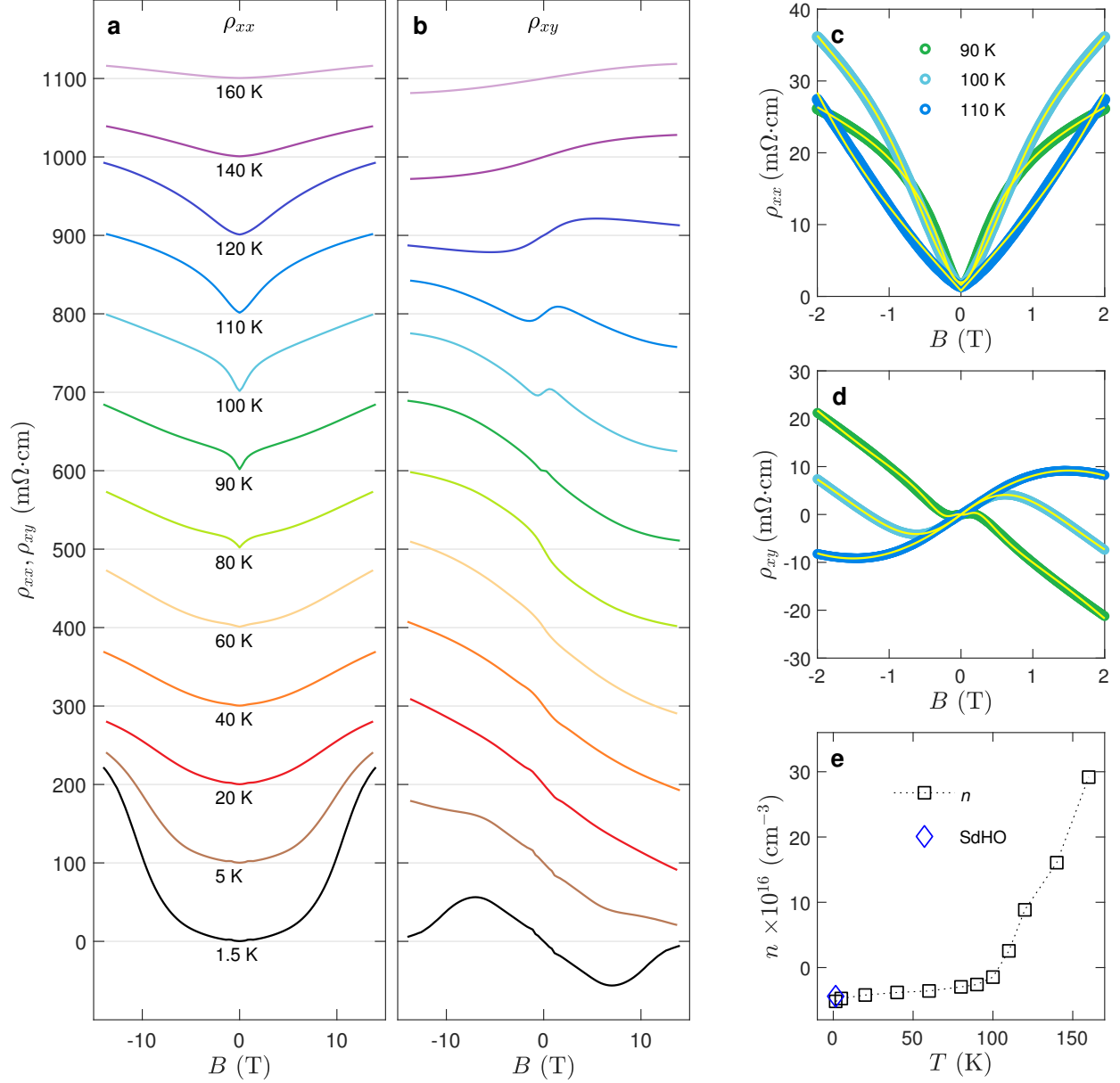


FIG. S1. Electrical transport. **a, b** Longitudinal electrical resistivity  $\rho_{xx}$  and transverse electrical (Hall) resistivity  $\rho_{xy}$  versus the magnetic field at different temperatures. Curves are shifted for clarity. **c, d**  $\rho_{xx}$  and  $\rho_{xy}$  at 90 K, 100 K, 110 K for  $B < 2$  T. Bright solid lines are fits to a two-band model. **e** Carrier densities as a function of temperature. Positive values represent holes, while negative values represent electrons. The diamond symbol represents the carrier density calculated from Shubnikov-de Haas oscillations at 1.5 K.



The temperature dependence of the carrier density is consistent with the Lifshitz transition observed in the angle resolve photo-emission spectroscopic study[S2].

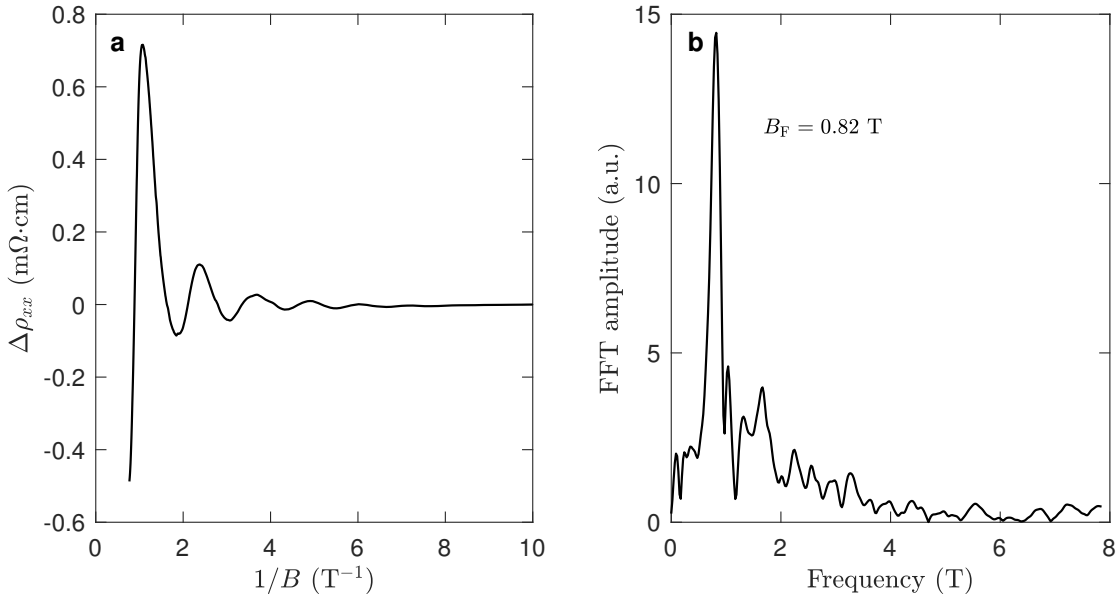


FIG. S2. Quantum oscillations of magnetoresistance at 1.5 K. **a** Oscillations of the magnetoresistance after subtraction of a background. **b** Fast Fourier transformation analysis of the quantum oscillation.

Fig. S2 shows the oscillatory part of the magnetoresistance at 1.5 K, which one can see is periodic in  $1/B$ . At least six periods of oscillations can be identified. The FFT spectrum displays a sharp peak at about 0.82 T, which agrees well with the slope of the Landau plot shown in Fig. 1 of the main text. From the oscillation frequency, we estimate the carrier density  $4.3 \times 10^{16} \text{ cm}^{-3}$ .

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