Signature of the Quantized Thermoelectric Hall Effect in a Topological Weyl Semimetal

Fei Han\textsuperscript{1*†}, Nina Andrejevic\textsuperscript{2†}, Thanh Nguyen\textsuperscript{1†}, Brian Skinner\textsuperscript{3†}, Quynh Nguyen\textsuperscript{1,3}, Zhiwei Ding\textsuperscript{4}, Ricardo Pablo-Pedro\textsuperscript{1}, Shreya Parjan\textsuperscript{5}, Vladyslav Kozii\textsuperscript{3}, Ahmet Alatas\textsuperscript{6}, Ercan Alp\textsuperscript{6}, Songxue Chi\textsuperscript{7}, Jaime Fernandez-Baca\textsuperscript{7}, Shengxi Huang\textsuperscript{8}, Liang Fu\textsuperscript{3*}, Mingda Li\textsuperscript{1*}

\textsuperscript{1}Department of Nuclear Science and Engineering, Massachusetts Institute of Technology, Cambridge, MA 02139, USA
\textsuperscript{2}Department of Materials Science and Engineering, Massachusetts Institute of Technology, Cambridge, MA 02139, USA
\textsuperscript{3}Department of Physics, Massachusetts Institute of Technology, Cambridge, MA 02139, USA
\textsuperscript{4}Department of Mechanical Engineering, Massachusetts Institute of Technology, Cambridge, MA 02139, USA
\textsuperscript{5}Department of Physics, Wellesley College, 106 Central St, Wellesley, MA 02481, USA
\textsuperscript{6}Advanced Photon Source, Argonne National Laboratory, Lemont, IL 60439, USA
\textsuperscript{7}High Flux Isotope Reactor, Oak Ridge National Laboratory, Oak Ridge, TN 37830, USA
\textsuperscript{8}Department of Electrical Engineering and Computer Science, The Pennsylvania State University, State College, PA 16801, USA

Thermoelectrics are promising by directly generating electricity from waste heat. However, (sub-) room-temperature thermoelectrics has posed a long-standing challenge, due to the entropic nature of thermopower that is suppressed at low temperature\textsuperscript{1}. In this light, topological materials\textsuperscript{2-4} offer a new avenue for energy harvest applications\textsuperscript{5}. Recent theories predicted that topological Weyl semimetals (WSMs) near quantum limit can lead to a non-saturating longitudinal thermopower\textsuperscript{6}, as well as a quantized thermoelectric Hall coefficient approaching to

*Corresponding authors: hanfei@mit.edu, liangfu@mit.edu, mingda@mit.edu.
†These authors contribute equally to this work.
a universal value. Here, we experimentally demonstrate the non-saturating thermopower and the signature of quantized thermoelectric Hall coefficients in WSM tantalum phosphide (TaP). An ultrahigh longitudinal thermopower $S_{xx} = 1.1 \times 10^3 \mu\text{V/K}$, along with a power factor $\sim 3 \Theta \mu\text{W/cm/K}^2$, are observed $\sim 50K$. Moreover, the thermoelectric Hall coefficients develop a plateau at high-fields and low temperatures, which further collapse into a single curve determined by universal constants. Our work highlights the unique electronic structure and topological protection of Weyl nodes toward thermoelectric harvest applications at low temperature.

Over two-thirds of the global energy production is rejected as waste heat. Thermoelectrics is attractive by directly converting waste heat into electricity without moving parts. The efficiency of the thermoelectric energy conversion is an increasing function of a dimensionless quantity $zT = \frac{\sigma S^2}{\kappa} T$, where $\sigma$, $S$, $\kappa$ denote the electrical conductivity, thermopower, and total thermal conductivity. Conventional thermoelectrics largely focus on tuning the thermal and electrical conductivities. Many efforts, such as lower dimension, microstructuring and nanostructuring, share the same driving force: by increasing the scattering of major heat carriers of long mean-free-path phonons without affecting the electrons with shorter mean-free-path, a level of independent control between electrical conductivity $\sigma$ and thermal conductivity $\kappa$ can be achieved, such as the phonon-glass electron-crystal state. However, less attention was paid to improve the thermopower $S$, even though the $S^2$ relation in $zT$ makes its appealing to improve. Moreover, current thermoelectrics mainly work at elevated temperatures, where room-temperature and sub-room-temperature thermoelectrics are highly challenging. In fact, these two main issues are deeply connected by the intrinsic entropic nature of thermopower $S$. To increase electrical conductivity $\sigma$, large carrier density-of-state is preferred. However, the large density-of-state indicates a large electronic entropy, which
is suppressed at reduced temperature\textsuperscript{14}. As a result, dramatic increase of electronic entropy without hampering electrical transport becomes a critical step to achieve high-performance thermoelectrics at sub-room-temperature regime.

In conventional thermoelectrics, bandstructure engineering has been applied to create partially-filled carrier pockets\textsuperscript{15}, which can lead to an increase of electronic entropy. This approach has also been applied to topological WSMs, where small Fermi level near the charge neutral point effectively increases the electronic entropy\textsuperscript{16-18}. However, this approach is intrinsically limited, due to the existence of an upper bound determined by the bandstructure topography. In this light, magnetic field has the potential to truly break the bound, since the density of state’s linear dependence allows the accommodation of unbounded, even macroscopic number of states in each Landau level. However, in conventional materials, the charge carriers are localized at high $B$-field due the cyclotron motion, limiting the possible thermoelectric applications.

The recent development of topological materials brings new hope to bypass the fundamental limitations met by conventional thermoelectrics, due to the topological protection of the electronic states. Compared to topological insulators where the topological protected states only emerges as boundary hence prohibits massive deployment, the topological WSMs\textsuperscript{19} with bulk topology offers new opportunities. Instead of utilizing the charge compensation effect in recent reports\textsuperscript{16-18}, it is worthy to note the WSM system has additional unique properties that make the WSM promising, including a special $n=0$ Landau level and the topological robustness of Weyl nodes. The $n=0$ Landau level in a WSM has a highly unique \textit{energy-independent} density of state $g_{\text{WSM}}(n=0) = N_f Be/4\pi^4\hbar^2v_F$ growing linear as $B$, which can create huge electronic entropy. Meanwhile, the topological protection of the Weyl node prohibits a gap opening hence is exempted from the localization. Based on this spirit, recent theories predicted a
non-saturating longitudinal thermopower and a quantized magnitude of thermoelectric Hall conductivity\textsuperscript{6,7}.

In this work, we carry out high-precision thermoelectric measurements using a centimeter-sized type-I WSM of TaP (Figure 1, a and b and Supplementary Information I). Fermi level is fine-tuned through the synthesis procedure to approach the \( n=0 \) Landau level near the W2 Weyl node (Figure 1g). As a result, on one hand, large, non-saturating longitudinal thermopower \( S_{xx} \) is observed, which grows linearly as a function of \( B \)-field, and reaches a peak value of 1100µV/K at 9T. On the other hand, the signature of the quantized thermoelectric Hall coefficients are observed, where at low-temperature and high fields, the thermoelectric Hall conductivity, also known as the transverse Peltier tensor \( \alpha_{xy} = [S/p]_y \) is independent of \( B \)-field, and collapse into a single curve determined by number of fermion flavors, Fermi velocity and universal constants. Moreover, the evidence of the breakdown of the Wiedemann-Franz law further indicates collective hydrodynamic electronic behaviours. Our work paves the way for a new class of high-performance thermoelectric materials that exploit the enhanced properties emerging from the unconventional topology found in nodal semimetals. These candidate materials of may fill the gap for low-temperature energy harvest.

Quantum oscillation. We first present our data of the longitudinal magnetoresistance up to 9T, where the magnetic field is parallel to the \( c \)-axis direction of TaP. A giant magnetoresistance was observed, where at \( T<25 \)K, the relative magnetoresistance \((R(9T) - R(0T))/R(0T) > 10^5\). Such large ratio is a signature behaviour for an electron-hole compensation system, which is further confirmed by the two-band model fitting, from which we obtain a \( n_e = 2.39 \times 10^{19}/\text{cm}^3 \) and \( n_h = 2.35 \times 10^{19}/\text{cm}^3 \) at the base temperature of 2.5K (Supplementary Information II). The quantum oscillation signature is observed from both the longitudinal and the transverse magnetotransport, which is preserved up to \( T=25 \)K (Figure 1d). This indicates an ultrahigh sample quality.
The Shubnikov–de Haas oscillations (SdH) are analysed by first subtracting a smooth background to obtain $MR$, which is plotted as a function of $1/B$ to determine the Landau level indices. Our analysis shows that the dominant charge carrier lies in the $n=2$ Landau level (Figure 1c). Through fast Fourier transform of $MR$, we can cross check that this $n=2$ Landau level corresponds to the oscillation frequency $F_B = 18T$. Since the dispersion for the linear-dispersive Weyl fermion at $k_z = 0$ of Landau level $n$ is given by $E_n = \text{sgn}(n)v_F \sqrt{2eB|n|}$, while the oscillation frequency $F$ is related to the Fermi-surface projection $S_F$ as $F = \frac{\hbar}{2\pi e} S_F = \frac{\hbar k_F^2}{2e} = \frac{E_F^2}{2e\hbar v_F^2}$, then we have $F = B|n|$, resulting in the $n=2$ Landau level, consistent with Landau-level index plot analysis. This $n=2$ Landau level corresponds to the electron pocket of the Weyl node W1 (Figure 1g). If we want to achieve the $n=0$ Landau level for W1, either large hole doping or a much higher magnetic field is required, which becomes unpractical. Fortunately, the TaP has two inequivalent Weyl nodes (Figure 1b). In fact, the extremely low frequency of $F_\alpha = 3T$ strongly suggests an extremely small Fermi surface very close to the Weyl node W2. Compared to a recent report with an $F_\alpha = 15T$ and a 13meV below the Fermi level$^{20}$, here, our result shows a Fermi level $E_F \sim 2.5\text{meV}$ below the W2 Weyl node. Since the spacing between $n=1$ and $n=0$ Landau levels is given by $E_1 - E_0 = v_F \sqrt{2eB} \sim 8\text{meV}$ at $B=9T$ using the reported value $v_F \sim 7 \times 10^4 \text{m/s}$ at W2 Weyl node$^{21}$, the fact that $E_F < E_1 - E_0$ confirms that our WSM sample lies in the $n=0$ Landau level of the W2 Weyl nodes.

**Non-saturating thermopower.** With the knowledge of charge carrier characteristics, we carried out the thermoelectric measurements using a diagonal offset geometry (Figure 2a), where both the electronic and thermal transport information along the longitudinal and transverse directions can be acquired by flipping the field polarity (Supplementary Information IV). In this section, we focus on the longitudinal thermoelectric properties. The longitudinal thermopower $S_{xx}$ is shown in Figure 2b, where a peak value $\max(S_{xx}) = 1.07 \times 10^3 \mu\text{V/K}$ is observed at $B = 9T$ and $T = 48\text{K}$. One prominent
feature is that $S_{xx}$ has developed an exotic double-peak behaviour, where a second peak emerges at slightly lower temperature, which can be attributed to the two types of the Weyl nodes: at low fields, the W1 will dominate and lead to the left peak given its higher carrier density from electron pocket, while at higher fields, the $n=0$ Landau level at W2 will take over. This is so since at high fields approaching to the quantum limit, $S_{xx} \sim \rho_{xy} \alpha_{xy}$, thus for $n=0$ Landau level, $\alpha_{xy}$ is a $B$-independent constant, while $\rho_{xy} \propto B$, indicating a dominant contribution of $S_{xx}$ from the carriers near W2 Weyl node. In particular, it has been predicted that the $S_{xx}$ has a simple formula:

$$S_{xx} = \frac{k^2}{\hbar^2} \frac{N_f}{12} \frac{TB}{v_F(n_e-n_h)}$$  \hspace{1cm} (1)

where $N_f$ is number of Weyl nodes, for TaP, we have $N_f = 24$, $v_F$ denotes the Fermi velocity and $n_e - n_h$ is the net carrier density. The linearity with $T$ and $B$ are shown in Figures 2c and 2d, respectively. Despite the simplicity of Eq. (1), it is worthwhile mentioning that Eq. (1) is in quantitative agreement with our result. For instance, at the peak value of $S_{xx}$ at $T = 48$K and $B = 9$T, our experimental results show a $n_e$ $n_h \sim 8 \times 10^{17}/\text{cm}^3$ and a $v_F \sim 2.2 \times 10^4 \text{m/s}$ obtained from the fitting discussed later, while the theory from Eq. (1) gives a $S_{xx,\text{theory}} \sim 9 \times 10^2 \mu \text{V/K}$, showing a generally good agreement with our data at $S_{xx,\text{exp}} \sim 1.07 \times 10^3 \mu \text{V/K}$. Moreover, such quantitative agreement is valid across all magnetic field range and up to ~50K (Figure 2e). Given the high thermopower, a giant longitudinal power factor up to $PF \equiv \sigma_{xx} S_{xx}^2 \sim 310 \mu \text{W/cm/K}^2$ is further achieved (Figure 2f). This is one order of magnitude higher than the peak values of practical thermoelectrics (e.g. SnSe, $PF(\text{max}) \sim 10 \mu \text{W/cm/K}^2$).

**Quantized thermoelectric Hall effect.** As to the transverse thermoelectric properties, we can see that the transverse thermopower $S_{xy}$ also reaches a peak value $\sim 10^3 \mu \text{V/K}$ at $T = 50$K (Figure 3a), moreover with a plateau. This plateau behaviour is consistent with the recent report arisen from the constant k-space volume while varying
the Weyl-cone separation as a function of field\textsuperscript{16}, and shall not be confused with the thermoelectric hall conductivity $\alpha_{xy}$. $\alpha_{xy} \equiv (S_{xy}\rho_{xx} + S_{xx}\rho_{xy})/(\rho_{xx}^2 + \rho_{xy}^2)$ is shown in Figure 3b, where at low-temperature range, the flatness against $B$-field starts to emerge. In particular, under the low-temperature $k_B T \ll E_F$ and high-field $B \gg E_F^2/\hbar v_F^2$ limit, the $\alpha_{xy}$ is predicted to approach to the following universal value that is dependent on neither field nor carrier density\textsuperscript{7}:

$$\alpha_{xy, \text{ideal}} = \frac{\pi^2 e^2 k_B T}{3 (2\pi \hbar)^2} \frac{N_f}{v_F}$$  \hspace{1cm} (2)

The linearity of $\alpha_{xy}$ as a function of $T$ is shown in Figure 3c, where we see that the linearity holds up to $T\sim 10$K. As a direct consequence, the $\alpha_{xy}/T$ curve collapses into one single curve, which can be seen in Figure 3d, where the ideal value of $\alpha_{xy, \text{ideal}}/T = 0.6$ for a $v_F \sim 1.2 \times 10^4 m/s$ obtained from full fitting with Eq. (3) and (4). This is at the same order of magnitude as our data (Figure 3d), yet the quantitative difference can be traced back to the W1 Weyl node with higher carried density, where a much higher field is need to bring both the electron pocket and hole pocket to quantum limit (Figure 3e). To take into account other Landau levels’ contribution, or take into account the finite scattering time which leads to the observed peak behaviour at , we used the multi-Landau level formula in dissipationless limit\textsuperscript{7}:

$$\alpha_{xy} = \frac{eN_f}{2\pi\hbar} \sum_{n=0}^\infty \int \frac{dk_z}{\pi} \left[ s \left( \frac{\varepsilon_n^0(k_z) - \mu}{k_B T} \right) + s \left( \frac{\varepsilon_n^0(k_z) + \mu}{k_B T} \right) \right]$$  \hspace{1cm} (3)

and a finite scattering formula for $n=0$ Landau level formula\textsuperscript{7}:

$$\alpha_{xy} = \frac{N_f}{18} \pi^2 k_B^2 T v_F \pi^2 B \left( 1 + 3\omega_F^2(E_F)\tau^2 \right)$$  \hspace{1cm} (4)

to extract a more reliable chemical potential and Fermi velocity. The fitting results using Eq. (3) and (4) are shown in Figure 3e and f, respectively, which are in very good
agreement. The acquired chemical potential $\mu$ is consistent with our independent electrical transport measurements (Figure1), while the Fermi velocity $v_F$ is slightly lower since all formula are for one fermion flavors, while for Weyl semimetal TaP, it has two distinct Weyl node, and the $n=2$ Landau level at W1 Weyl node away from linear dispersion will reduce the averaged $v_F$, which is reasonable.

**Breakdown of the Wiedemann-Franz Law.** Wiedemann-Franz (WF) law is a robust empirical law in metals stating that the ratio between the electronic thermal conductivity $k_e$ and electrical conductivity $\sigma$ is related to a universal value of Lorenz number:

$$ L_0 \equiv \frac{k_e}{\sigma T} = \frac{\pi^2}{3} \left( \frac{k_B \hbar}{e} \right)^2 = 2.44 \times 10^{-8} \text{W}\Omega^{-1}K^2. \quad (5) $$

Recently, it has been reported the strong violation of WF law in 2D Dirac fluid of graphene\textsuperscript{23} and Weyl semimetal WP\textsubscript{2}\textsuperscript{24} due to the collective hydrodynamic behaviors of the electron fluid. Since magnetic field also has an effect on the WF law\textsuperscript{25}, it is worthwhile to examine the validity of WF law in the type-I WSM of TaP. To do so, it is of paramount importance to properly separate the electronic and lattice thermal conductivity contribution. Here we use two independent methods to carry out the separation. On the one hand, we computed the lattice thermal conductivity $k_{ph}$ using *ab initio* density functional perturbation theories. The corresponding experimental phonon dispersion from inelastic scattering (Figure 4a) shows excellent agreement with the *ab initio* calculations, indicating a level of validity to use the theoretical $k_{ph}$ to perform the subtraction. On the other hand, we adopt the following empirical relation by taking advantage of the field-dependence\textsuperscript{26}:

$$ k_{xx}(T, B) = k_{xx}^{ph} + \frac{k_{xx}^e(T)}{1 + \beta_e(T)B^n}. \quad (6) $$
where $\beta_e(T)$ is a measure of zero-field electron mean free path. Using this method, the extracted lattice thermal conductivity $k_{ph}$ agrees well with the computed $k_{ph}$ from first-principles calculations (Figure 4b), making the extracted $k_{ph}$ serve as a reliable background to be subtracted. The corresponding electronic thermal conductivity $k_e$ and the $\frac{k_e}{\sigma T}$ ratio are shown in Figure 4c and Figure 4d, respectively. It can be seen that at $B = 0T$, the agreement with the WF law is good. However, as field increases up to $B = 9T$, as much as four-fold violation of WF law is observed (Figure 4d). The origin of the breakdown might be related to probable quasiparticle breakdown, or collective electron hydrodynamics, which is subject to further investigation.

In this work, we demonstrated a giant, non-saturating longitudinal thermopower and a quantized thermoelectric Hall conductivity, both showing quantitative agreement with recent theoretical proposals. In addition, a field-driven breakdown of the WF law is also observed. This opens up a new avenue to fill the gap of the low-temperature thermoelectric energy harvest: instead of focusing on thermal conductivity reduction, here the giant electronic entropy created by the topological protection of Weyl nodes provide a new arena where the giant thermopower is robust. This finding enables great potential application of topological materials on leading the breakthrough of the figure of merit of thermoelectric materials.

When we were finalizing this manuscript, we became aware that a similar work on Dirac semimetal was posted on arXiv. The related work mutually strengthened the reliability of the quantized thermoelectric Hall effect with our work together.
Figure 1. Quantum oscillation of TaP. (a) The crystal structure and (b) the Brillouin zone of TaP, highlighting the locations of the Weyl nodes W1 and W2. (c) Longitudinal magneto-resistivity (MR) as a function of temperature from 2K to 300K. A high (>10^5) MR ratio is observed. (d) Temperature-dependent transverse MR, where the quantum oscillation feature can sustain up to 25K. (e) Landau-level index plot showing an n=2 Landau level for the dominant charge carriers. (f) The Fourier transform of the MR showing a low oscillation frequency $F_\alpha=3T$. This is a signature that in addition to the main electron pocket from W1 Weyl node, we are very close to the W2 Weyl node. (g) The schematic bandstructure of our TaP sample.
Figure 2. Non-saturating thermopower at high fields. (a) The schematics of the diagonal offset thermoelectric measurement geometry. (b) Longitudinal thermopower $S_{xx}$ as a function of temperature at various fields. (c) $S_{xx}$ at low-temperature range, showing the quasi-linearity growth as a function of temperature. (d) $S_{xx}$ replotted as a function of $B$, showing the unbounded linear growth with field. The oscillatory behavior ~20K at $B=6T$ is from the quantum oscillation effect. (e) $S_{xx}$ as a function of $B$ at a few representative temperatures, fitted using Eq. (1). (f) The power factor as a function of temperature. The black-dashed line is a reference value.
Figure 3. The Quantized thermoelectric Hall effect. (a) Transverse thermopower $S_{xy}$ as a function of magnetic field at different temperatures. (b) Thermoelectric Hall coefficient $\alpha_{xy}$ as a function of magnetic field at different temperatures. (c) Thermoelectric Hall coefficient $\alpha_{xy}$ as a function of magnetic field at low temperatures. The insert shows a linear behavior of the $\alpha_{xy}$ versus $T$ curves at low temperatures. (d) $\alpha_{xy}/T$ as a function of magnetic field collapses to a constant plateau at quantum limit. (e) Enlarged view of $\alpha_{xy}/T$ at low temperatures indicating a convergence to the quantum limit. The gray dashed line gives the universal value obtained via fit of $\alpha_{xy}/T$. (f) Values of the band structure parameters obtained by fitting $\alpha_{xy}$ versus $T$. Details of the fitting procedure are given in Figure S7.
Figure 4. The Wiedemann-Franz Law. (a) Experimentally measured values of phonon modes of TaP along high-symmetry line Z-Γ-Σ taken by inelastic x-ray scattering with accompanying DFPT calculation (line) displaying good agreement between ab initio calculations and experiment. (b) Separation of phonon and electronic contributions to the longitudinal thermal conductivity with inset displaying a computation of the longitudinal thermal conductivity from first principles. (c) Plot of the electronic contribution of the thermal conductivity versus temperature and (d) of the Lorenz number versus temperature with black line indicating the theoretical value of the Wiedemann-Franz law.
References

1. Rowe, D. M. CRC handbook of thermoelectrics. (CRC Press, 1995).
2. Hasan, M. Z. & Kane, C. L. Colloquium: Topological insulators. Rev Mod Phys 82, 3045-3067, doi:10.1103/RevModPhys.82.3045 (2010).
3. Qi, X.-L. & Zhang, S.-C. Topological insulators and superconductors. Rev Mod Phys 83, 1057-1110, doi:10.1103/RevModPhys.83.1057 (2011).
4. Armitage, N. P., Mele, E. J. & Vishwanath, A. Weyl and Dirac semimetals in three-dimensional solids. Rev Mod Phys 90, 015001, doi:10.1103/RevModPhys.90.015001 (2018).
5. Heremans, J. P., Cava, R. J. & Samarth, N. Tetradymites as thermoelectrics and topological insulators. Nature Reviews Materials 2, 17049, doi:10.1038/natrevmats.2017.49 (2017).
6. Skinner, B. & Fu, L. Large, nonsaturating thermopower in a quantizing magnetic field. Science Advances 4, eaat2621, doi:ARTN eaat2621 10.1126/sciadv.aat2621 (2018).
7. Kozii, V., Skinner, B. & Fu, L. Thermoelectric Hall conductivity and figure of merit in Dirac/Weyl materials. eprint arXiv:1902.10123, arXiv:1902.10123 (2019).
8. Dresselhaus, M. S. et al. New Directions for Low-Dimensional Thermoelectric Materials. Advanced Materials 19, 1043-1053, doi:10.1002/adma.200600527 (2007).
9. Biswas, K. et al. High-performance bulk thermoelectrics with all-scale hierarchical architectures. Nature 489, 414-418, doi:10.1038/nature11439 (2012).
10. Il Kim, S. et al. Dense dislocation arrays embedded in grain boundaries for high-performance bulk thermoelectrics. Science 348, 109-114 (2015).
11. Minnich, A. J., Dresselhaus, M. S., Ren, Z. F. & Chen, G. Bulk nanostructured thermoelectric materials: current research and future prospects. Energy & Environmental Science 2, 466, doi:10.1039/b822664b (2009).
12. Vineis, C. J., Shakouri, A., Majumdar, A. & Kanatzidis, M. G. Nanostructured thermoelectrics: big efficiency gains from small features. Adv Mater 22, 3970-3980, doi:10.1002/adma.201000839 (2010).
13. Snyder, G. J. & Toberer, E. S. Complex thermoelectric materials. Nat Mater 7, 105, doi:10.1038/nmat2090 (2008).
14. Heremans, J. P. et al. Enhancement of Thermoelectric Efficiency in PbTe by Distortion of the Electronic Density of States. Science 321, 554, doi:10.1126/science.1159725 (2008).
15. Pei, Y. et al. Convergence of electronic bands for high performance bulk thermoelectrics. Nature 473, 66-69, doi:10.1038/nature09996 (2011).
16. Caglieris, F. et al. Anomalous Nernst effect and field-induced Lifshitz transition in the Weyl semimetals TaP and TaAs. Physical Review B 98, doi:10.1103/PhysRevB.98.201107 (2018).
17. Watzman, S. J. et al. Dirac dispersion generates unusually large Nernst effect in Weyl semimetals. Physical Review B 97, doi:10.1103/PhysRevB.97.161404 (2018).
18 Sharma, G., Moore, C., Saha, S. & Tewari, S. Nernst effect in Dirac and inversion-asymmetric Weyl semimetals. *Physical Review B* 96, doi:10.1103/PhysRevB.96.195119 (2017).

19 Xu, S.-Y. *et al.* Discovery of a Weyl fermion semimetal and topological Fermi arcs. *Science* 349, 613 (2015).

20 Arnold, F. *et al.* Negative magnetoresistance without well-defined chirality in the Weyl semimetal TaP. *Nat Commun* 7, 11615, doi:10.1038/ncomms11615 (2016).

21 Xu, N. *et al.* Observation of Weyl nodes and Fermi arcs in tantalum phosphide. *Nat Commun* 7, 11006, doi:10.1038/ncomms11006 (2016).

22 Zhao, L. D. *et al.* Ultralow thermal conductivity and high thermoelectric figure of merit in SnSe crystals. *Nature* 508, 373-377, doi:10.1038/nature13184 (2014).

23 Crossno, J. *et al.* Observation of the Dirac fluid and the breakdown of the Wiedemann-Franz law in graphene. *Science* 351, 1058-1061, doi:10.1126/science.aad0343 (2016).

24 Gooth, J. *et al.* Thermal and electrical signatures of a hydrodynamic electron fluid in tungsten diphosphide. *Nature Communications* 9, doi:ARTN 4093 10.1038/s41467-018-06688-y (2018).

25 Tanatar, M. A., Paglione, J., Petrovic, C. & Taillefer, L. Anisotropic violation of the Wiedemann-Franz law at a quantum critical point. *Science* 316, 1320-1322, doi:10.1126/science.1140762 (2007).

26 Ocana, R. & Esquinazi, P. Thermal conductivity tensor in YBa2Cu3O7-x: Effects of a planar magnetic field. *Physical Review B* 66, doi:ARTN 064525 10.1103/PhysRevB.66.064525 (2002).

27 Hartnoll, S. A. Theory of universal incoherent metallic transport. *Nat Phys* 11, 54-61, doi:10.1038/nphys3174 (2014).

28 Zhang, W. *et al.* Quantized plateau in the thermoelectric Hall conductivity for Dirac electrons in the extreme quantum limit. *eprint arXiv:1904.02157*, arXiv:1904.02157 (2019).
I. High-quality Single-crystal Growth

The single crystals of TaP were prepared by the vapor transport method. 3 grams of Ta (Beantown Chemical, 99.95%) and P (Beantown Chemical, 99.999%) powders were weighted, mixed and ground in a glovebox. The mixed powders were flame-sealed in a quartz tube which was subsequently heated to 700°C and dwelled for 20 hours for a pre-reaction. The obtained TaP powders were sealed in another quartz tube with 0.4 gram of I₂ (Sigma Aldrich, >=99.8%) added. The tube containing TaP and I₂ was then
horizontally put in a two-zone furnace. To improve the crystal size and quality, instead of setting a 100°C temperature difference which was reported previously,\textsuperscript{1,2,3} we gradually increased the temperature difference from zero until the I\textsubscript{2} transport agent started to flow. This process seems to be furnace- and distance- specific. In our case, the optimal temperatures for the two zones are 900°C and 950°C, respectively. With the help of the transport agent I\textsubscript{2}, the TaP source materials transferred from the cold end of the tube to the hot end and condensed at the hot end in a single-crystalline form in 14 days. The resultant products of TaP single crystals are centimeter size and have a metallic luster. Figure S1 exhibits a typical sample of TaP crystals.

**Figure S1.** Single crystals of TaP grown by the vapor transport method.

**II. Experimental Details and Data Analysis for High-quality Thermoelectric Measurement**

Due to the very high electronic and thermal conductivities of TaP, it is difficult to do precision electronic and thermal transport measurements on the as-grown crystals. To magnify the electronic resistance and the temperature gradient in the electronic and thermal transport measurements, one piece of crystal was polished to thin down along the c-axis. Figure S2(b) shows a side-view of a thinned-down crystal whose thickness is only 0.17 mm.
Figure S2. (a) Top view and (b) side view of the thinned-down sample we used for the thermoelectric measurement.

To measure the longitudinal and transverse resistivities $\rho_{xx}$ and $\rho_{xy}$ and Seebeck coefficients $S_{xx}$ and $S_{xy}$ simultaneously, we used a diagonal offset probe geometry for the electronic and thermal transport measurements, as shown in Figure S2(a).

Figure S3(a) shows a schematic diagram for the principle of the thermal transport measurement. The thermal transport option (TTO) system of PPMS heated the left end of the crystal and sunk the heat from right to generate a continuous heat flow along the $a$ or $b$ axis ($a$ and $b$ are equivalent for this tetragonal system). With detecting the temperature difference between the two thermometers, the TTO of PPMS can then calculate thermal conductivity directly from the applied heater power, resulting $\Delta T$, and sample dimension. The TTO also monitored the voltage drop between the two thermometers simultaneously, which yields the Seebeck signals by calculating $-\Delta V/\Delta T$. Magnetic fields were applied along the $c$ axis for detecting the proposed quantized thermoelectric Hall effect. Figure S3(b) exhibits the temperature dependences of thermal conductivity of TaP at 9 T and -9 T. From this plot, we can see the data at positive and negative magnetic fields have a very slight shift. This indicates the thermoelectric Hall effect (the transverse movement of thermal electrons in the presence of magnetic field) provides a slight but observable heat flow along the transverse
direction. The magnitude of the thermal conductivity of TaP is very large compared with most of materials. This explains the thinning-down requirement of the sample. The Seebeck signals at 0 T, 9 T and -9 T are plotted in Figure S3(c) from which giant magnetic-field-induced Seebeck signals can be observed at 9 T and -9 T and the data for 9 T and -9 T is asymmetrical due to the mutual presence of longitudinal and transverse Seebeck signals. To separate the longitudinal and transverse Seebeck signals, we use the following equations to calculate the longitudinal and transverse Seebeck coefficients $S_{xx}$ and $S_{xy}$:

$$S_{xx} = \frac{S_{meas}(+H) + S_{meas}(-H)}{2}, \quad S_{xy} = \frac{S_{meas}(+H) - S_{meas}(-H)}{2} \frac{L}{W'}$$ (1)

here $L$ and $W$ represent the length and the width between the two thermometers. The temperature dependences of $S_{xx}$ and $S_{xy}$ collected at different magnetic fields are presented in Figure S3(d) and (e). It is obvious for audiences that the applied magnetic fields induce giant Seebeck coefficients along both longitudinal and transverse directions. The longitudinal Seebeck coefficient $S_{xx}$ shows a linear behavior being proportional to the magnitude of the magnetic fields, and it does not show a trend to saturate up to 9 T. Differently, $S_{xy}$ tends to saturate at high magnetic fields. It should be noted such kind of giant magnetic-field-induced Seebeck coefficients cannot be observed in the case of $B//a//heat$ current, revealed by the comparison of two geometries in Figure S3(f). This illustrates the giant magnetic-field-induced longitudinal and transverse Seebeck coefficients in the case of $B//c//heat$ current originate from the quantized protection of thermoelectric Hall Effect. Another novel behavior in $S_{xx}$ and $S_{xy}$ is the presence of a two-peak feature at around 40 K. In the main text we give a clear explanation to this feature.
Figure S3. (a) Schematic diagram of the thermal transport measurement. (b) Thermal conductivities of TaP at 9 T and -9 T. (c) Measured Seebeck signals at 0 T, 9 T and -9 T for the diagonal offset probe geometry. (d) Longitudinal and (e) transverse Seebeck coefficients $S_{xx}$ and $S_{xy}$ as a function of temperature at different magnetic fields. (f) Comparison of the $B//a//heat current and B//c//heat current geometries. The giant Seebeck coefficients were not observed in the $B//a//heat current case.

After the TTO did the thermal transport measurement at a certain temperature, it would subsequently switch its mode to carry out the electronic transport measurement at the same temperature. Figure S4(a) is the schematic diagram for the principle of the electronic transport measurement. In the presence of magnetic fields, the system applied an electronic current along the $a$ or $b$ axis, and the voltage meter between the diagonal offset probes detected the voltage drop which contains both longitudinal and transverse components. The longitudinal resistivity $\rho_{xx}$ and the transverse resistivity (also called Hall resistivity) $\rho_{xy}$ are separated with the following equations:

$$
\rho_{xx} = \frac{\rho_{\text{meas}}(+H) + \rho_{\text{meas}}(-H)}{2}, \quad \rho_{xy} = \frac{\rho_{\text{meas}}(+H) - \rho_{\text{meas}}(-H)}{2} \frac{L}{W}.
$$

(2)
Figure S4(b) exhibits the measured resistivities of 0 T, 9 T and -9 T. The disagreement of the 9 T and -9 T data gives the evidence for the mutual presence of the longitudinal and transverse resistivities $\rho_{xx}$ and $\rho_{xy}$. After $\rho_{xx}$ and $\rho_{xy}$ are separated with the equation (2), we can observe a giant magnetoresistance (MR) in $\rho_{xx}$. The longitudinal and transverse conductivities $\sigma_{xx}$ and $\sigma_{xy}$ plotted in Figure S4(e) and (f) are obtained by the tensor operation:

$$
\sigma_{xx} = \frac{\rho_{xx}}{\rho_{xx}^2 + \rho_{xy}^2}, \quad \sigma_{xy} = -\frac{\rho_{xy}}{\rho_{xx}^2 + \rho_{xy}^2}.
$$

(3)

**Figure S4.** (a) Schematic diagram of the electronic transport measurement. (b) Measured resistivities of TaP at 0 T, 9 T and -9 T for the diagonal offset probe geometry. Longitudinal and transverse resistivities and conductivities (c) $\rho_{xx}$, (d) $\rho_{xy}$, (e) $\sigma_{xx}$ and (f) $\sigma_{xy}$ as a function of temperature at different magnetic fields.

**III. Magnetotransport and Carrier Concentration**

To extract the information about the Fermi surface and the carriers, other than the electronic transport measurement measured with the TTO we talked about in the last part, we also carried out a more delicate electronic transport measurement with the
electronic transport option (ETO) of PPMS. For this measurement, we adopted the symmetric six-probe geometry, with its schematic diagram shown in Figure S5(a). With the symmetric probes, the symmetry of the longitudinal and transverse resistivities $\rho_{xx}$ and $\rho_{xy}$ at positive and negative magnetic fields can be observed, as shown in Figure S5(b) and (c). At low temperatures, a signature giant magnetoresistance for an electron-hole compensation system is observed, and in both $\rho_{xx}$ and $\rho_{xy}$ strong Shubnikov-de Haas oscillations exist from 2 K to a relatively high temperature, 25 K, at which in most of cases the thermal noise can kill the quantum oscillations. This indicates a high quality of crystallization in our sample. The longitudinal and transverse conductivities $\sigma_{xx}$ and $\sigma_{xy}$ are calculated with the equation (3). At each measured temperature, we simultaneously fit our transverse and longitudinal conductivity data as a function of $B$ using a two-band model defined by:

\[
\sigma_{xy} = \left[ n_h \mu_h^2 \frac{1}{1 + (\mu_h B)^2} - n_e \mu_e^2 \frac{1}{1 + (\mu_e B)^2} \right] eB,
\]

\[
\sigma_{xx} = \frac{n_e \mu_e e}{1 + (\mu_e B)^2} + \frac{n_h \mu_h e}{1 + (\mu_h B)^2}, \quad (4)
\]

where $n_e$ and $n_h$ denote the electron and hole carrier densities, $\mu_e$ and $\mu_h$ are the corresponding mobilities, and $e$ is the elementary charge. We thereby extract the electron and hole carrier densities and mobilities as functions of temperature, as shown in Figure S5(f) and (g). The $n_e$ and $n_h$ are almost compensated at low temperatures. This proves the origin of the giant magnetoresistance.
**Figure S5.** (a) Schematic diagram of the electronic transport measurement in the symmetric six-probe geometry. Longitudinal and transverse resistivities and conductivities (b) $\rho_{xx}$, (c) $\rho_{xy}$, (d) $\sigma_{xx}$ and (e) $\sigma_{xy}$ as a function of magnetic field at different temperatures. (f) Carrier concentrations and (g) mobilities of electron and hole resulted from the two-band model fitting.

**IV. Thermoelectric Hall Coefficient**

To validate the quantized thermoelectric Hall effect, especially the quantized plateau of the thermoelectric Hall coefficient $\alpha_{xy}$ at high magnetic field limit, we calculated $\alpha_{xy}$ with the following equation:

$$\alpha_{xy} = \frac{\rho_{xy} S_{xx} + \rho_{xx} S_{xy}}{\rho_{xx}^2 + \rho_{xy}^2}.$$  \hspace{1cm} (5)

Before the calculation, we took the points of $S_{xx}$ and $S_{xy}$ from Figure S3 and $\rho_{xx}$ and $\rho_{xy}$ from Figure S4, and replotted them as a function of magnetic field, as shown in Figure 6(a), (b), (c) and (d). The resultant $\alpha_{xy}$ calculated with the equation (5) is displayed in Figure 6(e). A detailed analysis on the quantized thermoelectric Hall effect can be found in the main text.
Figure S6. Longitudinal and transverse Seebeck coefficients and resistivities (a) $S_{xx}$, (b) $S_{xy}$, (c) $\rho_{xx}$ and (d) $\rho_{xy}$ as a function of magnetic field at different temperatures. (e) Thermoelectric Hall coefficient $\alpha_{xy}$ as a function of magnetic field at different temperatures.

Figure S7. Fitted Thermoelectric Hall coefficient and extracted band structure parameters in the low-temperature dissipationless limit (a-c); low-temperature and low-
magnetic field limit with weak scattering (d-f); and high-temperature limit with weak scattering (g-i).

To both extract the value of Fermi velocity $v_F$ and chemical potential $\mu$, as well as identify the quantized value of $\alpha_{xy}/T$ approached at very large fields, we fit our low-temperature data up to $T=10$K using the general expression of $\alpha_{xy}$ in the dissipationless limit [7]:

$$\alpha_{xy} = \frac{e N_f}{2\pi \hbar} \sum_{n=0}^{\infty} \int_0^{\infty} \frac{dk_z}{\pi} \left[ s \left( \frac{\epsilon_n^0(k_z) - \mu}{k_B T} \right) + s \left( \frac{\epsilon_n^0(k_z) + \mu}{k_B T} \right) \right]$$

where $N_f$ equals the number of Weyl points, and $\epsilon_n^0(k_z)$ denote the Landau level energies

$$\epsilon_n^0(k_z) = \text{sign}(n)v_F\sqrt{2e\hbar B|n| + \hbar^2 k_z^2}$$

The function $s(x)$ is given in [7]. The fit is shown in S7 (a), and we extrapolate the fitted function to even larger magnetic fields, revealing we are near the onset of the quantized limit, shown in the inset. The value of $\alpha_{xy}/T$ approached in this limit is $\sim 0.5 \text{ AK}^{-2} \text{ m}^{-1}$. The corresponding fitted parameters are given in S7 (b) and (c).

To verify this fit, we additionally fit our low-temperature data using the expression for $\alpha_{xy}/T$ valid in the low-temperature and low magnetic field limit, but which includes a scattering time $\tau$ and is thus a more expressive form for data with weak scattering present:

$$\alpha_{xy} = \frac{N_f e^2 k_B^2 T v_F \tau^2 B}{18} \frac{1 + 3 \omega_c^2 (E_F) \tau^2}{(1 + \omega_c^2 (E_F) \tau^2)^2}$$

where the cyclotron frequency $\omega_c$ is given by

$$\omega_c(\epsilon) = \frac{e B v_F^2}{\epsilon}$$
This fit is shown in Figure S7 (d) with the corresponding fitted parameters shown in S7 (e) and (f), which are in fairly good agreement with those of the previous fit.

Similarly, we fit our high-temperature data in the limit of weak scattering using

\[
\alpha_{xy} = \frac{N_f e^2 k_B^2 T \nu_F \tau^2 B}{6 \pi^2 \hbar^3} \int_{-\infty}^{\infty} \frac{x^4 e^x}{(1 + e^x)^2 x^2 + \omega_c^2(k_B T) \tau^2} \, dx
\]

which is shown in Figure S7 (g) with corresponding fitted parameters in S7 (g) and (i).

V. Separation of Phonon and Electron Contributions to Thermal Conductivity

To check the obeying or violation of Wiedemann-Franz law, the phonon and electron contributions to thermal conductivity need to be separated. Figure S3(b) shows a slight shift of thermal conductivity at positive and negative magnetic fields resulted from the transverse heat flow provided by the thermoelectric Hall effect. We use the follow equations to extract the longitudinal thermal conductivity from the measured thermal conductivity:

\[
\rho_{thermal,xx} = \frac{\kappa(+H) + \kappa(-H)}{2\kappa(+H)\kappa(-H)}, \quad \rho_{thermal,xy} = \frac{L}{W} \frac{\kappa(+H) - \kappa(-H)}{2\kappa(+H)\kappa(-H)},
\]

\[
\kappa_{xx} = \frac{\rho_{thermal,xx}}{\rho_{thermal,xx}^2 + \rho_{thermal,xy}^2}.
\]

Figure S8(a) displays the obtained longitudinal thermal conductivity \(\kappa_{xx}\) as a function of temperature at different magnetic fields. From the insert of Figure S8(a) we can see the applied magnetic field gradually suppresses the longitudinal thermal conductivity. This phenomenon is coherent with the giant magnetoresistance. To separate the phonon and electron contribution, we fit the \(\kappa_{xx}\) versus \(B\) curves with the following empirical equation:

\[
\kappa_{xx}(T, B) = \kappa_{xx}^{phonon}(T) + \frac{\kappa_{xx}^{electron}(T, B = 0T)}{1 + \beta_e(T)B^n},
\]
here $\beta_e(T)$ is proportional to the zero-field electronic mean free path of quasiparticles and $n$ is related to the nature of the quasiparticle scattering.\textsuperscript{4,5,6}

Figure S8(b) and (c) reveals that the $\kappa_{xx}$ versus $B$ curves at typical high and low temperatures 300 K and 100 K can be well fitted with the equation (7). The fitting process for the $\kappa_{xx}$ versus $B$ curves at different temperatures successfully achieves the separation of the phonon and electron contributions to thermal conductivity. The yielded phonon and electron thermal conductivities have been talked in the main text. Here we stress that the $\beta_e(T)$ yielded from the fitting shows a typical behavior of thermally elevated electron-phonon scattering, as shown in Figure S8(d). This indicates that our fitting process is reasonable.
**Figure S8.** (a) Longitudinal thermal conductivity \( \kappa_{xx} \) as a function of temperature at different magnetic fields. The curve fitting for the \( \kappa_{xx} \) versus \( B \) curves at (b) 300 K and (c) 100 K. (d) The fitting parameter \( \beta_e(T) \) as a function of temperature.

**VI. Consistency of Charge Neutral**

Our results from different measurements show high self-consistency. Taking charge neutral as an example, we can observe a high agreement of the temperatures of the charge neutral in carrier concentration and in Seebeck coefficient. In the plot of the carrier concentrations of electron and hole as a function of temperature, the top panel of Figure S9, the electron and hole get neutralized at around 100 K while the Seebeck coefficient at 0 T changes its sign at around 75 K, as shown in the bottom panel of Figure S9.

![Graphs showing carrier concentrations and Seebeck coefficient](image)

**Figure S9.** Consistency of charge neutral in carrier concentration and Seebeck coefficient.
References

1. Arnold, F. et al. Negative magnetoresistance without well-defined chirality in the Weyl semimetal TaP. *Nat Commun* **7**, 11615, doi:10.1038/ncomms11615 (2016).

2. Hu, J. et al. π Berry phase and Zeeman splitting of Weyl semimetal TaP. *Sci. Rep.* **6**, 18674, doi: 10.1038/srep18674 (2016).

3. Zhang, C. et al. Large magnetoresistance over an extended temperature regime in monophosphides of tantalum and niobium. *Phys. Rev. B* **92**, 041203(R), doi: 10.1103/PhysRevB.92.041203 (2015).

4. Ocana, R. et al. Thermal conductivity tensor in YBa$_2$Cu$_3$O$_{7-x}$: Effects of a planar magnetic field. *Phys. Rev. B* **66**, 064525, doi: 10.1103/PhysRevB.66.064525 (2002).

5. Pogorelov, Y. et al. Mechanisms of heat conductivity in high-Tc superconductors. *Phys. Rev. B* **51**, 15474, doi: 10.1103/PhysRevB.51.15474 (1995).

6. Yu, F. et al. Tensor Magnetothermal Resistance in YBa$_2$Cu$_3$O$_{7-x}$ via Andreev Scattering of Quasiparticles. *Phys. Rev. Lett.* **74**, 5136, doi: 10.1103/PhysRevLett.74.5136 (1995).

7. Kozii, Vladyslav, Brian Skinner, and Liang Fu. "Thermoelectric Hall conductivity and figure of merit in Dirac/Weyl materials." arXiv preprint arXiv:1902.10123 (2019).