Quantized thermoelectric Hall effect induces giant power factor in a topological semimetal

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Thermoelectrics are promising by directly generating electricity from waste heat. However, (sub-)room-temperature thermoelectrics have been a long-standing challenge due to vanishing electronic entropy at low temperatures. Topological materials offer a new avenue for energy harvesting applications. Recent theories predicted that topological semimetals at the quantum limit can lead to a large, non-saturating thermopower and a quantized thermoelectric Hall conductivity approaching a universal value. Here, we experimentally demonstrate the non-saturating thermopower and quantized thermoelectric Hall effect in the topological Weyl semimetal (WSM) tantalum phosphide (TaP). An ultrahigh longitudinal thermopower $S_{xx} \sim 1.1 \times 10^3 \, \mu\text{V} \, \text{K}^{-1}$ and giant power factor $\sim 525 \, \mu\text{W} \, \text{cm}^{-1} \, \text{K}^{-2}$ are observed at ~40 K, which is largely attributed to the quantized thermoelectric Hall effect. Our work highlights the unique quantized thermoelectric Hall effect realized in a WSM toward low-temperature energy harvesting applications.

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"ver two-thirds of global energy production is rejected as waste heat. Thermoelectrics are attractive by directly converting waste heat into electricity without moving parts. The efficiency of thermolectric energy conversion is an increasing function of a dimensionless quantity $zT = \sigma S^2 T / \kappa$, where $\sigma$, $S$, and $\kappa$ denote the electrical conductivity, thermopower, and total thermal conductivity, respectively. Conventional thermoelectrics largely focus on tuning the thermal and electrical conductivities. Many efforts, such as lowering dimensionality, microstructuring, and nanostructuring, share the same principle: by increasing the scattering of major heat carriers of long mean free-path phonons without affecting the short mean free-path electrons, a level of independent tunability 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 $S^2$ dependence in $zT$ makes such improvement appealing. Moreover, thermopower $S$ is proportional to the entropy per carrier and is therefore suppressed at reduced temperature. For this reason, current thermoelectrics are generally effective only at elevated temperatures and there is a pressing need for thermoelectrics that work efficiently at room temperature and below. Filling this need requires new materials that can exhibit large electronic entropy at low temperatures while maintaining significant electrical conductivity.

One approach to creating large electronic entropy is band-structure engineering through low carrier density, partially filled carrier pockets; a similar principle has also been applied to semimetals, such as Bi$_2$Te$_3$, graphite, and most recently Weyl semimetals (WSMs), to explore large entropy at low carrier density. However, the electrical conductivity is thereby reduced. Magnetic field offers an additional incentive to dramatically increase the entropy, as the linear field dependence of the density of states (DOS) enables unbounded, macroscopic number of states in each Landau level (LL), yet in conventional thermoelectrics, charge carriers will be localized at high B-field due to the cyclotron motion, still resulting in low conductivity. Consequently, increasing power factor ($\equiv \sigma S^2$) creates a significant challenge as it requires optimization of both $\sigma$ and $S$ under conflicting conditions.

The recent development of topological materials, including topological WSMs, offers a new pathway to surpass conventional thermoelectrics that relies on the topological protection of electronic states. It is particularly worthy to note that the WSM system has a unique $n = 0$ LL, which has a highly unusual, energy-independent DOS $g(n = 0) = N_f Be / 4\pi^2 h v_f$ increasing linearly with $B$, and therefore can create huge electronic entropy. More importantly, the system remains gapless at high field, thanks to the topological nature of Weyl nodes. Consequently, recent theories predicted a non-saturating thermopower and quantized thermoelectric Hall conductivity at the quantum limit, where electrons and holes contribute additively to high thermoelectric performance without experiencing localization.

In this work, we carry out high-precision thermoelectric measurements using a centimeter-sized crystal WSM TaP (Fig. 1a, b and Supplementary Notes 1 and 2). The Fermi level is fine-tuned through the synthesis procedure to approach the $n = 0$ LL near the W2 Weyl node (Fig. 1g). In this system, giant, non-saturating longitudinal thermopower $S_{xx}$ is observed, which exhibits linear dependence with B-field without saturation. In addition, the signature of the quantized thermoelectric Hall conductivity is observed, where the low-temperature, high-field thermoelectric Hall conductivity $s_{xy} \equiv \rho_{xy} / \sigma_{xx}$ approaches a universal curve determined by number of fermion flavors, Fermi velocity, and universal constants. Moreover, evidence of Wiedemann–Franz (WF) law violation further indicates a breakdown of quasiparticle behaviors. Our work leverages the effects of topology to overcome challenges for low-temperature thermoelectric energy harvesting from a power factor perspective.

**Results**

**Quantum oscillations.** We first present the longitudinal magnetoresistance (MR) data, where Giant MR was observed. At $T < 25$ K, the $\text{MR} = (\rho_{xx}(B) - \rho_{xx}(0T)) / \rho_{xx}(0T)$ exceeds 10%, which is a signature of electron-hole compensation, which is further confirmed by the two-band model fitting of conductivity, with $n_e = 2.39 \times 10^{19} \text{cm}^{-3}$ and $n_h = 2.35 \times 10^{19} \text{cm}^{-3}$ at $T = 2.5$ K, along with a high mobility of $1 \times 10^5 \text{cm}^2 \text{V}^{-1}\text{s}^{-1}$ (Supplementary Note 3). The background-subtracted MR, denoted $\Delta MR$, exhibits Shubnikov-de Haas oscillations, which are plotted against $1/B$ to determine oscillation frequencies (Fig. 1d). The Fourier transform of $\Delta MR$ shows two small carrier pockets with low frequency $F_\alpha = 4$ T and $F_\beta = 18$ T among four pockets (Supplementary Note 4 and Fig. 1e). The LL fan diagram analysis indicates the two small pockets are at $n = 2$ LL and $n = 0$ LL, respectively (Supplementary Note 5 and Fig. 1f). The intersections of the linear LL index plots $(-0.037$ for $n = 0$ LL and $+0.065$ for $n = 2$) lying between $-1/8$ to $+1/8$ indicates that the two pockets are both topologically nontrivial, from which we attribute the $n = 2$ LL to the electron pocket of the W1 Weyl node, and the $n = 0$ LL to the hole pocket of the W2 Weyl node (Fig. 1g). Moreover, we see that the W2 and W1 pockets enter the quantum limit at $B = 3.8$ T and $16$ T, respectively. There is an alternate way to infer $n_L$. The Weyl fermion dispersion of the $\nu_{th}^n$ LL at is given by $E_n = \sgn(n)v_f/2\hbar n B|n|$, whereas the oscillation frequency $F$ satisfies $F = E_{p\nu}/2\hbar v_f$. When $E_{p\nu} \sim E_F$, we have $F \sim B / n$. This leads to an agreement between $n = 2$ LL and the measured $F_\beta = 18$ T at $B = 9$ T. For $F_\alpha$, the low frequency 4 T suggests an extremely small Fermi surface. Since the spacing between $n = 1$ and $n = 0$ LLs is given by $E_\alpha = 3v_f / 4\hbar B$, whereas the condition to reach the $n = 0$ LL quantum limit for W2 pocket is at $B > F_\alpha = 4$ T. This value agrees well with the above LL index analysis.

**Non-saturating thermopower and giant power factor.** Having determined the carrier characteristics, we carried out thermo-electric measurements using a diagonal offset geometry (Fig. 2a), where the electrical and thermal transport along both the longitudinal and transverse directions can be acquired together by flipping the field polarity (Supplementary Note 6, which also contains the phase relations between various thermoelectric quantities). The longitudinal thermopower $S_{xx}$ is shown in Fig. 2b, where $S_{xx}$ increases over 200-fold compared to its zero-field value, and reaches a giant magnitude of $1.07 \times 10^6 \mu\text{V}^{-1}$ without sign of saturation at $B = 9$ T and $T = 40$ K. One prominent feature is that $S_{xx}$ develops a double-peak behavior, which may be attributed to the two types of Weyl nodes: the higher carrier mobility and lower carrier density at the W2 node leads to reduced phonon scattering, and thus the high $S_{xx}$ can persist to higher temperatures. Quantitatively, it has been predicted that for the $n = 0$ chiral LL of Weyl electrons, $S_{xx}$ obeys a simple formula:

$$S_{xx} = \frac{k_B^2 N_f}{h^2} \frac{TB}{12 v_f^2 (n_h - n_q)} \tag{1}$$

where $N_f$ is the degeneracy of the Weyl nodes, $n_h - n_q$ is the net carrier density, and $v_f$ is an effective Fermi velocity. As TaP has two sets of Weyl nodes with different velocities and energies, in this work we introduce $v_f^\text{eff}$ as an effective parameter capturing an average Fermi velocity of the system.
The linearity of $S_{xx}$ with $T$ and $B$ is shown in Fig. 2c, d, respectively. It is noteworthy that Eq. (1) is in quantitative agreement with our result if we adopt the fitted value of the $v_{\text{eff}}^2$ using Eq. (3) and Eq. (S14), described in greater detail in the following section. Such quantitative agreement is valid across all fields and up to $\sim 40$ K and is a measure of the success of the effective model (Fig. 2e). Moreover, a giant longitudinal power factor $\equiv S_{xx}/\rho_{xx}$ up to $525 \mu W \text{cm}^{-1} \text{K}^{-2}$ is achieved due to the large entropy generated by the linearly-dispersive bands at quantizing magnetic fields, while a low $\rho_{xx}$ is maintained due to the protection of the gapless $n = 0$ LL, evading the typical fate of carrier cyclotron motion and localization under fields\textsuperscript{10,11}, which can achieve high thermopower at high magnetic fields with linear-dispersive bands, but cannot simultaneously maintain a low magneto-electrical resistivity.

**Quantized thermoelectric Hall effect.** Regarding the transverse properties, we see that the transverse thermopower $S_{yx}$ exhibits a plateau with increasing $B$-field, which is known to originate from the constant $k$-space volume as thermopower is a measure of occupational entropy in state space\textsuperscript{12}. The thermoelectric Hall conductivity $\alpha_{xy} \equiv (S_{yx} P_{yx} - S_{xx} P_{xx}) / (\rho_{xx}^2 + \rho_{yx}^2)$ is shown in Fig. 3b, where in the low-temperature range, the flatness with respect to $B$-field starts to emerge. In particular, under the low-temperature $k_B T \ll E_F$ and high-field $B \gg E_F / h v_F$ limit, $\alpha_{xy}$ is predicted to approach the following universal value that is independent of $B$-field, disorder, carrier density, and even carrier type\textsuperscript{21}:

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

The temperature dependence of $\alpha_{xy}$ is shown in Fig. 3c, where we see that the linearity with $T$ holds up to $T \sim 10$ K. As a direct consequence, the $\alpha_{xy}/T$ curves converge to a single curve at high fields (Fig. 3d, e), where an ideal value $\alpha_{xy, \text{ideal}}/T = 0.4 \text{A} \text{K}^{-2} \text{m}^{-1}$ is determined by evaluating Eq. (2) using $v_F^2 = 1.4 \times 10^4 \text{m s}^{-1}$, which is extracted by fitting a more general Eq. (3) at base temperature:

$$\alpha_{xy} = \frac{e N_f}{2 \pi \hbar} \sum_{n=0}^{\infty} \int_0^{\infty} \frac{dk_z}{\pi} \left[ (\phi_n(k_z) - \mu) \frac{k_y T}{k_B T} + (\phi_n^*(k_z) + \mu) \frac{k_y^* T}{k_B T} \right]$$  \hspace{1cm} (3)

in which $s$ is the electronic entropy function (Eq. S13). The magnitude $v_F^2$ is comparable to the single weighted average of projected Fermi velocity $v_{F1}^{W1} = 0.77 \times 10^5 \text{m s}^{-1}$, $v_{F2}^{W2} = 1.88 \times 10^5 \text{m s}^{-1}$, which gives $v_F^2 = 1.5 \times 10^5 \text{m s}^{-1}$, where the $z$-direction was chosen to coincide with the magnetic field direction. The fitted chemical potential $\mu$ is consistent with the electrical transport measurements, whereas the $v_F^2$ is lower than the $v_F$ at $225$. This can be understood, as carriers at W1 Weyl nodes at $n = 2$ LL have yet to reach extreme quantum limit (Figs. 1g and 3f, and Supplementary Notes 6 and 7). For temperatures above 10 K, scattering effects are significant and the dissipationless limit assumed in Eq. (3) is no longer valid; thus, for fits at $T \geq 10$ K, approximate forms of $\alpha_{xy}$, which include a
finite scattering time were used (Eq. (S14) and Eq. (S16)). To corroborate the universal quantization behavior of \( \alpha_{xy}/T \), we performed separate thermoelectric measurements up to \( B = 14 \, \text{T} \) at \( T = 2 \, \text{K} \) and \( 4 \, \text{K} \) and \( 6 \, \text{K} \), where the collapse onto a single curve and a clearer plateau are observed (Supplementary Note 8), in addition to giving \( \alpha_{xy, \text{ideal}}/T = 0.37 \, \text{A} \, \text{K}^{-2} \, \text{m}^{-1} \), in quantitative agreement with the \( 9 \, \text{T} \) data. Finally, to show that the quantized thermoelectric Hall coefficient \( \alpha_{xy} \) drives the ultrahigh thermopower and giant power factor, we decompose \( S_{xx} \) into its transverse \( ( -\alpha_{xy} \rho_{xy} ) \) and longitudinal \( ( \alpha_{xx} \rho_{xx} ) \) components, where we see that the transverse term \( \alpha_{xy} \rho_{xy} \) contributes to \( \sim 90\% \) of the longitudinal \( S_{xx} \) (Fig. 4a and Supplementary Note 9). The corresponding decomposed contributions to power factor \( S_{xx}^2/\rho_{xx} \) is shown in Fig. 4b.

In a nutshell, the quantized \( \alpha_{xy} \) large non-saturating \( S_{xx} \), and ultrahigh power factor \( S_{xx}^2/\rho_{xx} \) all originate from the topological Weyl node, but with increasingly stringent manifestation conditions: the quantized \( \alpha_{xy} \) comes directly from the gapless \( n = 0 \) LL states of Weyl fermions; as \( S_{xx} = -\alpha_{xy} \rho_{xy} + \alpha_{xx} \rho_{xx} \); \( \rho_{xy} \) should increase at high fields to obtain non-saturating \( S_{xx} \) with the field-independent \( \alpha_{xx} \); only when the transverse components \( -\alpha_{xy} \rho_{xy} \) dominate the \( S_{xx} \) with moderate \( \rho_{xx} \) can the power factor \( S_{xx}^2/\rho_{xx} \) be enhanced; the gapless \( n = 0 \) LL states can also contribute to reduce the \( \rho_{xx} \).

**Breakdown of the Wiedemann-Franz law.** The Wiedemann-Franz law is a robust empirical law stating that the ratio between the electronic thermal conductivity \( \kappa_e \) and electrical conductivity \( \sigma \) are related by a universal Lorenz number:

\[
L_0 \equiv \frac{\kappa_e}{\sigma T} = \frac{\pi^2}{3} \left( \frac{k_B}{e} \right)^2 = 2.44 \times 10^{-8} \, \text{W} \, \text{Ω}^{-1} \, \text{K}^{-2}
\]

Recently, it has been reported that there is strong violation of the WF law in the 2D Dirac fluid of graphene\(^{26}\) and WSM WP\(^{27}\) due to collective electron hydrodynamics. Other behaviors of electrons, like quantum criticality\(^{28}\) or quasiparticle breakdown\(^{29,30}\), can also lead to the WF law violation. It is thus worthwhile to examine the validity of the WF law in the field-induced high-entropy state of TaP. To do so, it is crucial to properly separate \( \kappa_e \) from the lattice thermal conductivity \( \kappa_{\text{ph}} \). We adopt the following empirical relation by using the field-dependence of \( \kappa_{\text{ph}} \)\(^{31}\):

\[
\kappa_{xx}(T, B) = \kappa_{\text{ph}}(T) + \frac{\kappa_{xx}^0(T)}{1 + \beta(T)B_m}
\]

where \( \beta(T) \) is a measure of zero-field electron mean free path and \( m \) is a factor related to the nature of scattering. Figure 5a demonstrates an example for such a separation procedure (Supplementary Note 11). Using this method, we see that the extracted \( \kappa_{\text{ph}} \) agrees well with the computed value from ab initio calculations (Fig. 5b and Supplementary Note 12), from which the phonon dispersions are also computed, and agree well with measured dispersion from inelastic scattering (Fig. 5c and Supplementary Note 10). All these agreements indicate the reliability of the separation process. The corresponding \( \kappa_e \) and the \( L_0 \) is shown in Fig. 5d, e, respectively. At \( B = 0 \, \text{T} \), the agreement with the WF law is good. However, as field increases to \( B = 9 \, \text{T} \), a fourfold violation of WF law is observed (Fig. 5d). This happens...
Fig. 3 The quantized thermoelectric effect. a Transverse thermopower $S_{xy}$ as a function of magnetic field at different temperatures. b Thermoelectric Hall conductivity $\alpha_{xy}$ as a function of magnetic field at different temperatures. The peak value is caused by the finite scattering effect. c Thermoelectric Hall conductivity $\alpha_{xy}$ as a function of temperature at various fields. The inset shows a linear behavior of $\alpha_{xy}$ versus $T$ at low temperatures. d $\alpha_{xy}/T$ as a function of magnetic field at various temperatures. e An extrapolation of d showing a convergence to the quantized value at low temperatures. f The density of states (DOS) of each Landau level (LL), highlighting the unique $n = 0$ LL in a WSM. At high-enough $B$, $n = 0$ LL drives the DOS $\propto B$.

Fig. 4 Dominant contribution from the transverse thermoelectric Hall conductivity to longitudinal thermopower and power factor. a Total $S_{xy}$ (left-hand side, LHS) at various temperatures as a function of magnetic field and its transverse component $-\alpha_{xy}\rho_{xy}$ (first term on the right-hand side, RHS) and longitudinal contribution $+\alpha_{xy}\rho_{xx}$ (second term on the RHS). b Total $S^{2}_{xy}/\rho_{xx}$ (LHS) at various temperatures as a function of magnetic field and its transverse component $+\alpha_{xy}^{2}\rho_{xy}$ (first term on RHS), longitudinal contribution $+\alpha_{xy}\rho_{xx}$ (second term on the RHS), and the cross-term $-2\alpha_{xy}\alpha_{y}\rho_{xy}$ (third term on the RHS). The dominant contribution of transverse component can be seen at all temperatures.
across a wide temperature range but not at low temperatures, indicating the link of scattering (Supplementary Note 11). The observed strong violation of the WF law hints at the possibility of field-driven, scattering-enhanced collective behaviors in a large entropic system and is subject to further investigation.

Discussion

Pathway toward room-temperature topical thermoelectrics. In this work, we report high thermopower and giant thermoelectric power factor in the WSM TaP, induced by the quantized thermoelectric Hall effect originating from topologically protected Weyl nodes at the quantum limit. These features are linked as follows: in a strong magnetic field, $S_{xx}$ is $\sim a_{xx}p_{xx}$, the quantizing behavior of $a_{xx}$ combined with the continual increase of $p_{xx}$ with magnetic field leads to the growth of $S_{xx}$, while suppression of longitudinal portion $a_{xx}p_{xx}$ to $S_{xx}$ further contributes to high power factor $S_{xx}^2/p_{xx}$. The choice of TaP is due to its simpler Fermi surface compared to other members in the TaAs family.25 On the other hand, the huge mass difference between Ta and P atoms reduces the three-phoron process and results in a high thermal conductivity, making it not directly applicable as a thermoelectric material. Even so, our work sheds light on a systematic pathway to seek promising topological thermoelectrics: To increase $S_{xx}$, large carrier compensation is desired (Eq. (1)). To simultaneously maintain low $p_{xx}$, simultaneously high carrier densities $n_\parallel$ and $n_\perp$ are required but not sufficient. In a topologically trivial semimetal such as Bi, although high thermopower can be achieved at the quantum limit ($S_{xx}(Bi) \sim 3 \times 10^3 \mu V K^{-1}$ vs $S_{xx}(TaP) \sim 1 \times 10^3 \mu V K^{-1}$), the electrical resistivity is significantly enhanced at high magnetic field ($\rho_{xx}(Bi) \sim 2 \times 10^{-2} \Omega m$ vs $\rho_{xx}(TaP) \sim 1 \times 10^{-5} \Omega m$)10, indicating the crucial contribution of the gapless $n = 0$ LL states from the topologically protected Weyl nodes. To tune the working temperature toward room temperature, long relaxation time is favored, along with preservation of the quantum limit, where thermal energy is smaller than the LL level spacing, $k_B T \ll v_F \sqrt{\hbar eB}$26. Finally, intrinsic magnetism can be used to replace the external $B$-field. Overall, we foresee that magnetic topological WSMs and related topological nodal line semimetals32,33 with protected gapless states are promising candidate materials for thermoelectrics when charge carriers are largely compensated and the Fermi level is tuned to the gapless nodes to unlock the quantized thermoelectric Hall effect. To summarize, we demonstrated non-saturating longitudinal thermopower, giant power factor, and a signature of quantized thermoelectric Hall conductivity in a WSM in quantitative agreement with recent theoretical proposals. Furthermore, a field-driven breakdown of the WF law is observed at intermediate temperatures. Given the promising magnitudes of thermopower and power factor, our work sheds light on a few essential requirements that high-performance room-temperature thermoelectrics should meet. These include a way to create giant electronic entropy and reduce carrier density, and a way of evading localization while maintaining high electrical conductivity. Interestingly, the $n = 0$ LL state with topologically protected Weyl nodes in a WSM satisfies all these requirements. Our work thus demonstrates the possibility of topological materials to lead the breakthrough of thermoelectric materials working below room temperature.
Note added. When we were finalizing this manuscript, we became aware of a work on Dirac semimetals. The related work and our work mutually strengthened each other on the part of the quantized thermoelectric Hall effect.

Data availability

The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

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Author contributions

M.L. and F.H. designed the experiments. V.K., B.S., and L.F. formulated the theory. F.H. synthesized the samples and performed the transport measurements, with the help of T.N., Q.T.N., and S.P. N.A., T.N., and F.H. analyzed the data, with contribution from all the authors. T.H. and F.H. carried out additional 14T measurements. Z.D. carried out ab initio calculations. N.A., T.N., F.H., R.P.-P., and M.L. along with A.A., E.A., S.C., J.F.-R., and S.H. performed inelastic scattering measurements. M.L., N.A., F.H., and T.N. wrote the paper with input from all authors.

Competing interests

The authors declare no competing interests.

Additional information

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