Giant Magnetic Quantum Oscillations and Chiral Anomaly in the Thermal Conductivity of a Weyl Semimetal

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(Dated: January 26, 2018)
Abstract

Giant quantum oscillations of magneto-thermal conductivity amounting to two orders of magnitude of the estimation based on the Wiedemann-Franz law have been observed in the prototypical Weyl semimetal TaAs. The characteristic oscillation frequency \( F \approx 7 \, \text{T} \) agrees well with that confirmed for a small hole-type Fermi pocket enclosing a Weyl node. A comparative analysis of various potential scenarios suggests a significant electron-phonon coupling that strongly modulates the phonon mean free path through Landau quantization of the electronic density of states to be at the heart. Resembling the chiral-anomaly induced positive magneto-electrical conductivity, an increase of the thermal conductivity in parallel magnetic field has also been observed. Our findings pose the question whether these are characteristic also for other recently discovered topological electronic materials, calling for more intensive investigations along this line.

PACS numbers: Valid PACS appear here

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In comparison to the electrical transport that reveals an abundance of novel features like chiral anomaly [1–4], thermal transport in particular thermal conductivity of Weyl semimetals, remains largely unexplored. This is partially ascribable to the technical difficulties in precise thermal management at low temperatures. Also, such work is seemingly insignificant because, for most electrical conductors, the Wiedemann-Franz (WF) law has already provided an easy approach to the electronic thermal conductivity from the electrical conductivity $\sigma$ in the elastic scattering limit of Fermi liquid picture. At least in the zero-temperature limit, the WF law is theoretically expected to be fulfilled for various topological materials [5–7]. Recent experimental results, however, suggest its violation by Dirac/Weyl fermions in particular circumstances, e.g., on the surface of a topological insulator [8], in the hydrodynamic transport regime [9], and in the presence of magnetic field [10], attracting renewed attention.

On the other hand, magnetic quantum oscillations (MQOs) of thermal transport like thermopower and Nernst effect have turned out to be useful complements to the conventional techniques like the Shubnikov-de Haas (SdH) effect of $\sigma(B)$ [11–14]. However, MQOs in the isothermal magneto-thermal conductivity $\kappa(B)$ have been rarely reported thus far. For, an estimate of the oscillation amplitude based on both the SdH effect and the WF law (denoted as $\kappa_{e,WF}(B)$ in Fig. 1b) is generally too small to be experimentally detected, as is the case of TaAs ($10^{-3}$–$10^{-1}$ W/Km), cf. Fig. 1 and Supplementary Information (SI). The few reported examples of MQOs in $\kappa(B)$ include GaAs/AlGaAs heterostructures containing two-dimensional electron system (2DES) [15] and the Weyl semimetal NbP [16]. An enhanced electron-phonon (e-p) interaction that modulates the lattice thermal conductance, or a large electronic bipolar-diffusion effect, were discussed as possible origin, respectively.

In this Letter, we report on an experimental observation of surprisingly large MQOs in magneto-thermal conductivity of the prototypical Weyl semimetal TaAs. The oscillation amplitude is the largest so far found for this quantity, exceeding the expectation of the WF law [$\kappa_{e,WF}(B)$] by two orders of magnitude. A comparative analysis involving different possible scenarios indicates that the lattice thermal conductivity, through significant e-p coupling, plays a dominant role in the observed MQOs. In addition, an unprecedented enhancement of $\kappa(B)$ was observed only when the field was aligned parallel to heat current ($B||dT$), reminiscent of the chiral-anomaly induced, positive magneto-electrical conductivity. Our results provide a new avenue for studying quantum transport of chiral fermions.
FIG. 1: Isothermal $\rho_{\parallel}(B)$ (a) and $\kappa_{\parallel}(B)$ (b) measured at 2 K for sample TAc. Also shown in (b) are the calculated $\kappa_{e,WF}(B)$ from the measured $\rho_{\parallel}(B)$ by the WF law. Note that the oscillation amplitude of $\kappa_{e,WF}(B)$ is two order of magnitude smaller than that of the measured $\kappa_{\parallel}(B)$. This is also the case of $\kappa_{e,HC}(B)$ whose values were calculated from the quantum oscillating magneto-heat capacity (SI) and the kinetic formula. The dashed line in (b) marks the position of the last extremum, where $\kappa_{\parallel}(B)$ minimizes, but $\kappa_{e,WF}(B)$ and $\kappa_{e,HC}(B)$ maximize, with contradictory phases. Insets are a sketch of the sample configuration for transport measurements shown in this plot (left) and the Fourier analysis of MQOs in $\rho_{\parallel}(B)$ (right).

We employed two TaAs samples cut from one single crystal grown by chemical vapor transport [17]. They were rectangular shaped, with the long axis either along the $c$ axis (denoted as TAc, dimension $0.3 \times 0.8 \times 2.2$ mm$^3$, cf. Figs. 1 left inset & S1) or along the $a$ axis (TAA, $0.4 \times 1.0 \times 2.1$ mm$^3$). The thermal conductivity was measured utilizing a home-designed sample holder with a chip resistor of 2000 $\Omega$ as heater and a thin ($\phi = 25 \mu m$) chromel-AuFe$_{0.07%}$ thermocouple for detecting the temperature difference $dT$, see SI. Prior
to our measurements, the thermocouple had been carefully calibrated in magnetic fields by using two Cernox CX1050 thermometers; a similar field dependence as previously reported [18] has been observed, cf. Fig. S2 in SI. The employment of a thin thermocouple ensures a rapid thermal relaxation when scanning the field, which is of uttermost importance to detect the MQOs in $\kappa(B)$. This can be confirmed from the recorded $dT(B)$ curve at $T = 2$ K, cf. Fig. S3, which is almost field-symmetric with reproducible MQOs.

Figure 1 presents the isothermal $\rho_\parallel(B)$ (a) and $\kappa_\parallel(B)$ (b) measured at $T = 2$ K for sample TAc. The subscript $\parallel$ denotes parallel electric/thermal current and magnetic field. A decrease of $\rho_\parallel(B)$ can be observed at $B > 1$ T, characteristic of the positive magneto-electrical conductivity arising from charge pumping between Weyl nodes of different chirality, in agreement to previous report [17]. Further increasing $B$ leads to significant MQOs in $\rho_\parallel(B)$, with several characteristic frequencies of $F \approx 7$, 15, and 23 T, cf. Fig. 1 right inset for the fast Fourier transform (FFT) spectrum. These observations are consistent with current knowledge on this material [17, 19–21]; the MQO of the lowest frequency is due to a tiny hole-type Fermi pocket enclosing a Weyl node.

Surprisingly, $\kappa_\parallel(B)$ reveals not only visible MQOs, but much more pronounced oscillation amplitude than that of $\rho_\parallel(B)$ (note the logarithmic $y$ axis of Fig. 1b). Such contrast of the two quantities is in particular marked when $B < 4$ T. Reliability of the observed MQOs in $\kappa_\parallel(B)$ can be confirmed by the oscillating $dT(B)$ in both positive and negative fields (Fig. S3). The maximum change of $\kappa_\parallel(B)$ in the last oscillation period ($\Delta \kappa$ in Fig. 1b) amounts to 12 W/Km, being 3.4 times of zero-field value of $\kappa_\parallel$. Unlike $\rho_\parallel(B)$, the MQOs of 7 T frequency are overwhelmingly dominant in $\kappa_\parallel(B)$, cf. $\kappa_\parallel/T$ vs $B^{-1}$ shown in Fig. 2 for different temperatures, as well as the corresponding FFT analysis (SI). The excellent agreement of the characteristic frequency to that probed other techniques underlines that the MQOs observed in thermal conductivity are a direct consequence of the Landau quantization of the electronic density of states (DOSs).

MQOs of this frequency, with 5–6 times smaller amplitude, have also been observed for sample TAa in $\kappa_\perp(B)$ ($dT||a, B||c$), see SI. Nevertheless, they are still huge, as becomes clear when comparing the estimated MQOs in $\kappa_{e,WF}(B)$ with the measured ones (1:10$^3$), cf. Fig. S5. The giant MQOs are, therefore, not a specific feature of the parallel field configuration, but rather determined by the anisotropic electronic structure. Likewise, the recently observed MQOs in $\kappa_\perp(B)$ of NbP ($\kappa_\parallel(B)$ not measured) reveals a smaller but
FIG. 2: Isothermal $\kappa_{\parallel}/T$ as a function of $B^{-1}$ measured at selected temperatures between $T = 1.8$ and 6 K. A characteristic frequency $F \approx 7$ T can be readily observed from the $T$-independent MQOs in period of $\sim 0.14$ T$^{-1}$. Refer to the corresponding FFT analysis in Fig. S6 (SI), too. MQOs with higher frequencies are too small to be reliably derived. For clarity, the $\kappa_{\parallel}/T$ curves have been shifted upward by 7 (6 K), 6 (5 K), 5 (4 K), 5.5 (3.5 K), 4.5 (3 K), 3.5 (2.6 K), 2 (2.3 K), 1 (2 K) units of W/K$^2$m, respectively.

comparable oscillation magnitude ($d\kappa_0/T \approx 30\%$), with however a more complicated FFT spectrum compared to that of TaAs [16].

The common description of $\kappa$ for nonmagnetic conductors considers an electronic ($\kappa_e$) and a lattice contribution ($\kappa_l$). For semimetals and semiconductors a bipolar contribution $\kappa_{bi}$ has to be taken into account, too. This is due to thermally excited electron-hole pairs which diffuse to the cold end where they recombine, releasing their excitation energies [22]. For a Fermi liquid, $\kappa_e$ can be obtained from $\sigma$ via the WF law,

$$\kappa_e/T = \sigma L_0,$$

with the Sommerfeld value of Lorenz number $L_0 \equiv \frac{\pi^2}{3}(\frac{k_B}{2})^2 = 2.44 \times 10^{-8}$ W·Ω·K$^{-2}$. The WF law is valid for a wide variety of materials, as long as the dominating charge scattering process is elastic. This is often realized at low enough temperatures under dominating impurity scattering. On the other hand, a potential bipolar term $\kappa_{bi}$ will increase with $T$ due to the enhanced thermal excitation of electron-hole pairs, see ref. [23].
FIG. 3: Temperature dependence of the normalized FFT amplitude of the characteristic MQOs with $F \approx 7$ T in $\kappa_{\parallel}(B)$, compared to those of magnetization (extracted from ref. [21]) and electrical resistivity with similar frequency.

To identify possible origins of the giant MQOs observed in $\kappa_{\parallel}(B)$ and $\kappa_{\perp}(B)$ (Figs. 1b, 2 and S4), we first consider an electronic contribution $\kappa_{e}(B)$ which is well enhanced over its WF counterpart viz. $L(B) \gg L_{0}(B)$. In order for this scenario to work, $L(B)$ has to exceed $L_{0}$ by a factor of $\sim 100$. For nonmagnetic conductors, application of magnetic field hardly affects $L$ because the charge-relaxation mechanisms involved remain unchanged. $L$ in Cu, for example, changes by only $\sim 10\%$ in $B < 5$ T at 4.8 K [24]. Recently, a moderately enhanced $L (< 3L_{0})$, which however decreases with $B$, was discussed for Cd$_{3}$As$_{2}$ [10]. Compelling evidence against this scenario stems from the $T$ dependence of the oscillations (see also the FFT spectrum at 7 T in Fig. S6), cf. Fig. 3. While the oscillation amplitude of both the magnetization $M$ and the $\rho$ follows the Lifshitz-Kosevich function anticipated for thermally damped Landau quantization [21], that of $\kappa_{\parallel}(B)$ does not: It becomes maximal at $T \approx 2.6$ K and diminishes upon further cooling, hinting at a non-electronic contribution.

Except for the factor-of-100 smaller amplitude, the MQOs in the calculated $\kappa_{e,WF}(B)$ reveal another important feature when compared to those of the measured $\kappa_{\parallel}(B)$. They have opposite phases, as demonstrated by the oscillation at $B \approx 7.3$ T corresponding to the first Landau level, see the dashed in Fig. 1b. This observation implies that in the field where the Landau level intersects with the Fermi level, TaAs is electrically conductive but thermally
insulated, contrary to the general expectation. The same conclusion is achieved from the heat capacity $C(B)$ (SI). The estimated $\kappa_{e,HC}(B)$ from $C(B)$ through the kinetic formula reveals MQOs that are two orders of magnitude smaller and opposite in phase compared to the observations in $\kappa_\parallel(B)$, resembling the aforementioned behavior of $\kappa_{e,WF}(B)$, cf. Fig. 1b.

In comparison, a violation of the WF law by a factor up to 22 was recently observed in the Dirac fluid regime of graphene in the range $T = 50-80$ K [9], i.e., an interacting quasi-relativistic electron-hole plasma in reduced dimensions that obeys hydrodynamics. This scenario does not apply to TaAs, because (i) this is a bulk system, and (ii) the transport in the low-$T$ range of interest is characterized by electron-phonon scattering, incompatible with electron hydrodynamics. At low temperatures, the WF law in graphene is indeed expected to be violated upon applying field due to the linear band dispersions [25]. However, the resulting oscillation amplitude of $\kappa_e(B)$ is very small, i.e., less than 1% of the zero-field value up to $B = 6$ T, in striking contrast to that of TaAs, which amounts to more than 300%, cf. Figs. 1-2.

An alternative explanation for the giant MQOs in $\kappa(B)$ relies on the bipolar-diffusion term $\kappa_{bi}(B)$. This effect has been intensively investigated for graphene which fulfills all the requirements for a significant $\kappa_{bi}(B)$, e.g., a zero band gap and charge neutrality [23]. There, bipolar-diffusion enhances $L$ by a factor 2–4 at room temperature; this factor diminishes with lowering $T$ due to decreasing thermal excitations. The enhancement was experimentally found to be only 35% at subkelvin region [26]. For TaAs, the estimated $\kappa_{e,WF}(T)$ amounts to 1/4 (TAc) and 70% (TAa) of the measured $\kappa(T)$ at room temperature [27]. There, the approximate $T^{-1}$ profile of $\kappa(T)$ indicates a sizable contribution due to the lattice contribution $\kappa_l$. An enhancement of $L$ due to the bipolar diffusion, if any, should be no larger than 4 fold and decreases upon cooling. Therefore, this effect appears to be negligible in current discussion on TaAs. In addition, the dominant one-frequency ($F = 7$ T) oscillation also argues against the bipolar scenario for the oscillating $\kappa(B)$ in this material.

Given that neither $\kappa_e(B)$ nor $\kappa_{bi}(B)$ can provide a satisfactory explanation for the observed MQOs in $\kappa(B)$ of TaAs, whether the Landau quantized electronic DOSs can trigger a quantum oscillating phonon contribution $\kappa_l(B)$ appears to be interesting. Indeed, the oscillating $\kappa(B)$ observed in GaAs/AlGaAs heterostructures was explained by considering the e-p coupling [15]. There, the heat-carrying phonons couple to the Landau-quantized 2D-DOSs confined to the interface and the phonon mean free path is consequently modulated.
by quantizing field. The non-monotonic $T$-dependence of the oscillation amplitude shown in Fig. 3 strongly supports such a scenario for TaAs: The MQOs tend to disappear at absolute zero because there $\kappa_1$ vanishes. We note the great difference of the oscillations between the two cases. In the 2D heterostructures, $dT$ oscillates within only 2% of its zero-field value, which is two orders of magnitude smaller than that of TaAs.

As acoustic phonons dominate $\kappa(T)$ of TaAs \[28\], our findings suggest a significant interaction between Weyl fermions and acoustic phonons. This is likely for TaAs with a Weyl node of extremely low energy ($\sim 2$ meV, cf. ref. \[20\]), allowing for acoustic phonons to be scattered by low-energy Weyl fermions of similar wavevectors. A different but intimately related consequence of e-p interaction in a quantizing field is known as ’magnetophonon oscillation’ \[29\], i.e., resonance scattering of quantized electrons by phonon of a distinct frequency. First-principles calculations of phonon dispersion for TaAs reveal a negligible field dependence of the phonon DOSs (see SI). This suggests that the modulated phonon mean free path, rather than the phonon frequency, is at the heart of the giant MQOs in $\kappa(B)$ which, therefore, occur in the lattice heat conductivity $\kappa_l(B)$ rather than its electronic counterpart $\kappa_e(B)$.

Apart from the giant MQOs, yet another unprecedented feature stands out, namely, the apparently different trends of $\kappa(B)$ measured in different field configurations. Figure 4 shows the isothermal $\kappa_\parallel(B)$ and $\kappa_\perp(B)$ of TAc measured at relatively higher temperatures $T = 8$ and 15 K, to avoid the giant oscillations. At low fields, $\kappa_\perp(B)$ and $\kappa_\parallel(B)$ match well to each other. Upon increasing field, they bifurcate, followed by a decrease of $\kappa_\perp(B)$ and an increase of $\kappa_\parallel(B)$. This difference is obviously not a consequence of thermally damped MQOs. The field of bifurcation shifts from 2 T ($T = 8$ K) up to 5 T ($T = 15$ K), cf. arrows in Fig. 4. When $T$ is sufficiently increased, the difference shrinks and finally the two curves fall almost on top of each other (Fig. 4 inset).

Generally, the isothermal $\kappa(B)$ of a nonmagnetic conductor decreases with $B$ because of the classical, positive orbital magnetoresistance and the consequently diminishing $\kappa_e(B)$, cf. Eq. 1 and ref. \[30\]. This term, however, is far too insufficient to explain the decreasing $\kappa_\perp(B)$, cf. Fig. 4 \[31\]. Instead, a field-induced, local diamagnetism that yields an enhanced anharmonic magnetic force on the lattice vibrations is probably involved in the decrease of $\kappa_\perp(B)$, as has been reported for InSb \[32\].

A much more intriguing observation, as shown in Fig. 4, is an increasing $\kappa_\parallel(B)$ in a para-
FIG. 4: Isothermal $\kappa(B)$ measured at $T = 8$ and $15$ K in both parallel ($\kappa_{\parallel}$) and perpendicular ($\kappa_{\perp}$) field configurations for sample TAc. Unlike $\kappa_{\perp}(B)$ that deceases and tends to saturate with increasing field, $\kappa_{\parallel}(B)$ bifurcate from $\kappa_{\perp}(B)$ at certain field (indicated by arrows) and reveals upward tendency in higher fields. Inset: $\kappa_{\parallel}(B)$ and $\kappa_{\perp}(B)$ measured at $T = 150$ K both show decreasing tendency, with almost equivalent behavior.

This provides a qualitative explanation to the different trends between $\kappa_{\perp}(B)$ and $\kappa_{\parallel}(B)$. A quantitative comparison requires the values of the prefactors $\alpha$ and $\beta$ of the chiral anomaly terms, which depend on the band structure and can be obtained by $k$-space integrals of Boltzmann transport coefficients with Berry curvature and chiral anomaly effects taken into account [6].

In conclusion, we have observed giant MQOs in the thermal conductivity $\kappa(B)$ of the prototypical Weyl semimetal TaAs. Electronically triggered magnetic quantum oscillations of the lattice thermal conductivity $\kappa_l(B)$ through corresponding oscillations of the electron-phonon coupling can explain our observations, at least qualitatively. In addition, the clear difference between $\kappa_{\parallel}(B)$ and $\kappa_{\perp}(B)$ very likely manifests the, as yet never found, evidence of...
chiral-anomaly in the electronic thermal conductivity. Our observations open a novel route to approach the interaction between charge and heat transport in a topological electronic material. Further insight into the thermal transport of Weyl fermions is badly required.

We would like to acknowledge fruitful discussions with K. Behina, X. Dai, G. Li, H. Weng and Z. Fang. This work was supported by the National Science Foundation of China (Grant Nos:11474332, 11474015, 11774018 and 61227902), the MOST of China (Grant Nos: 2015CB921303 and 2017YFA0303103) and the Chinese Academy of Sciences through the strategic priority research program (XDB07020200).

[1] H. B. Nielsen and M. Ninomiya, Phys. Lett. B 130, 389396 (1983).
[2] P. Hosur and X. L. Qi, C. R. Phys. 14, 857 (2013).
[3] A.A. Burkov, J. Phys.: Condens. Matter 27, 113201 (2015).
[4] S. Wang et al., Advances in Physics X 2, 518 (2017).
[5] K.S. Kim, Phys. Rev. B 90, 121108 (2014).
[6] G. Sharma, P. Goswami, and S. Tewari, Phys. Rev. B 93, 035116 (2015).
[7] R. Lundgren et al., Phys. Rev. B 90, 165115 (2014).
[8] Z. Luo, J. Tian, M. Srinivasan, Y.P. Chen and X. Xu, ArXiv. 1702.01716.
[9] J. Crossno et al., Science 351, 1058 (2016).
[10] A. Pariari, N. Khan and P. Mandal, arXiv: 1508.02286.
[11] J. G. Checkelsky and N. P. Ong, Phys. Rev. B 80, 081413R (2009).
[12] T. Liang et al., Nat. Commun. 4, 2696 (2013).
[13] Z. Zhu et al., Phys. Rev. Lett. 114, 176601 (2015).
[14] M. Matusiak, J.R. Cooper, and D. Kaczorowski, Nature Commun. 8, 15219 (2017).
[15] J.P. Eisenstein, A.C. Gossard, and V. Narayanamurti, Phys. Rev. Lett. 59, 1341 (1987).
[16] U. Stockert et al., J. Phys.: Condens. Matter 29, 325701 (2017).
[17] X.C. Huang et al., Phys. Rev. X 5, 031023 (2015).
[18] U. Stockert and N. Oeschler, Cryogenics 51, 154 (2011).
[19] C.L. Zhang et al., Nat. Commun. 7, 10735 (2016).
[20] H. Weng, C. Fang, Z. Fang, B.A.Bernevig and X. Dai, Phys. Rev. X 5, 011029 (2015).
[21] F. Arnold et al., Phys. Rev. Lett. 117, 146401 (2016).
Considering $\rho_\perp (B)$ of TAc at $T = 8$ and 15 K, the estimated $\kappa_{e,WF} (B)$ decreases by only less than 0.5 W/Km in low field then saturates. A gross violation of the WF law by a factor of 100 is indispensable in order for this term to account for the decrease of $\kappa_\perp (B)$ shown in Fig. 4. As discussed in the paper, this is unlikely.