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Gate-tunable negative longitudinal magnetoresistance in the predicted type-II Weyl semimetal WTe_2

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The progress in exploiting new electronic materials has been a major driving force in solid-state physics. As a new state of matter, a Weyl semimetal (WSM), in particular a type-II WSM, hosts Weyl fermions as emergent quasiparticles and may harbour novel electrical transport properties. Nevertheless, such a type-II WSM material has not been experimentally observed. In this work, by performing systematic magneto-transport studies on thin films of a predicted material candidate WTe_2 , we observe notable negative longitudinal magnetoresistance, which can be attributed to the chiral anomaly in WSM. This phenomenon also exhibits strong planar orientation dependence with the absence along the tungsten chains, consistent with the distinctive feature of a type-II WSM. By applying a gate voltage, we demonstrate that the Fermi energy can be *in-situ* tuned through the Weyl points via the electric field effect. Our results may open opportunities for implementing new electronic applications, such as field-effect chiral devices.

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Since the discovery of topological insulators, which significantly enriched band theory^{1,2}, the possibility of realizing new topological states in materials other than insulators, such as semimetals or metals, has attracted substantial attention^{3–7}. Weyl semimetals (WSMs), which host Weyl fermions⁸ as emergent quasiparticles, have recently sparked intense research interest in condensed matter physics^{3,9–16}. In WSMs, the conduction and valence bands linearly disperse across pairs of unremovable discrete points (Weyl points) along all three momentum directions^{3,17}, with the existence of Fermi Arc surface states as a consequence of separated Weyl points with opposite chirality³. Since the first theoretical prediction in pyrochlore iridates³, several materials that break either the time-reversal or spatial-inversion symmetry have been proposed as WSMs, including a series of transition metal monophosphides^{12,13}. These theoretical predictions have been experimentally confirmed by the observation of bulk Weyl points and surface Fermi Arcs^{18–21}, or the signature of chiral anomaly^{15,22–30} via electric transport studies. Many other new properties, such as the topological Hall effect¹⁴ and non-local quantum oscillations³¹, have also been proposed.

The type-II WSM was recently proposed as a new type of WSM with Weyl points appearing at the boundary of electron and hole pockets^{32–36}. Its distinctive feature of an open Fermi surface (in sharp contrast with a closed point-like Fermi surface in type-I WSMs) can induce exotic properties, such as planar orientation-dependent chiral anomaly. However, such type-II WSM materials have not been experimentally observed. As a unique layered transition-metal dichalcogenide that exhibits large and unsaturated (perpendicular) magnetoresistance (MR)³⁷, tungsten ditelluride (WTe_2) has been reported as a major material candidate for type-II WSM. While angle-resolved photoemission spectroscopy measurements encounter certain challenges in observing the Weyl points because of the limited experimental spectroscopic resolution^{32,38}, exploring the potential unique transport properties and realizing their tunability for future device applications are highly desirable.

In this report, low-temperature transport studies on thin WTe_2 samples are performed, revealing a clear negative longitudinal MR when the electric and magnetic fields are parallel. This phenomenon is highly angle sensitive and is suppressed by a small angle between the electric and magnetic fields, and this behaviour can probably be attributed to the chiral anomaly in the WSM. A unique property of type-II WSM, the planar orientation dependence, is also confirmed by the observed absence of negative longitudinal MR for all studied devices along the tungsten chains (a axis). We further demonstrate that by applying a gate voltage, the Fermi energy of such a material can be effectively tuned through the Weyl points; thus, the unique transport properties can be controlled, suggesting possible applications in future chiral electronics.

Results

The selection and fabrication of thin-film devices. WTe_2 is a T_d type of transition-metal dichalcogenide (space group $Pnm2_1$) with a tungsten chain along the a axis, as shown in Fig. 1a. The other principle axis, the b axis, is perpendicular to the a axis^{37,39}. This T_d phase breaks the inversion symmetry and was predicted to support the existence of type-II Weyl points^{32,33}. We first focus on a key signature of the possibly existed Weyl points: the chiral-anomaly-induced negative longitudinal MR phenomenon. To make such observation feasible, thin flakes are required to sufficiently suppress the contribution of the strong positive longitudinal MR⁴⁰. However, these thin flakes must be sufficiently thick, with energy bands similar to those of bulk crystals

(see Supplementary Fig. 1) to allow the existence of Weyl points. Thus, we selected thin WTe_2 flakes with thicknesses of 7–15 nm, which were prepared using the standard mechanical exfoliation method on a SiO_2 substrate and measured using an atomic force microscope. The crystalline orientations were identified using polarized Raman spectra⁴¹ (see Supplementary Fig. 2).

Thin WTe_2 devices with metal electrodes were fabricated using a home-made shadow mask method⁴², which effectively avoided undesirable wet process-induced doping in the pristine WTe_2 flakes⁴³. A typical optical image of a four-probe device is shown in Fig. 1b, where the determined thickness of the thin flake was ~ 14 nm (inset of Fig. 1b). Figure 1c shows the schematic drawing of the device structure and four-probe MR measurement setup. Here, the angle between the applied magnetic field \mathbf{B} and current direction \mathbf{I} is θ .

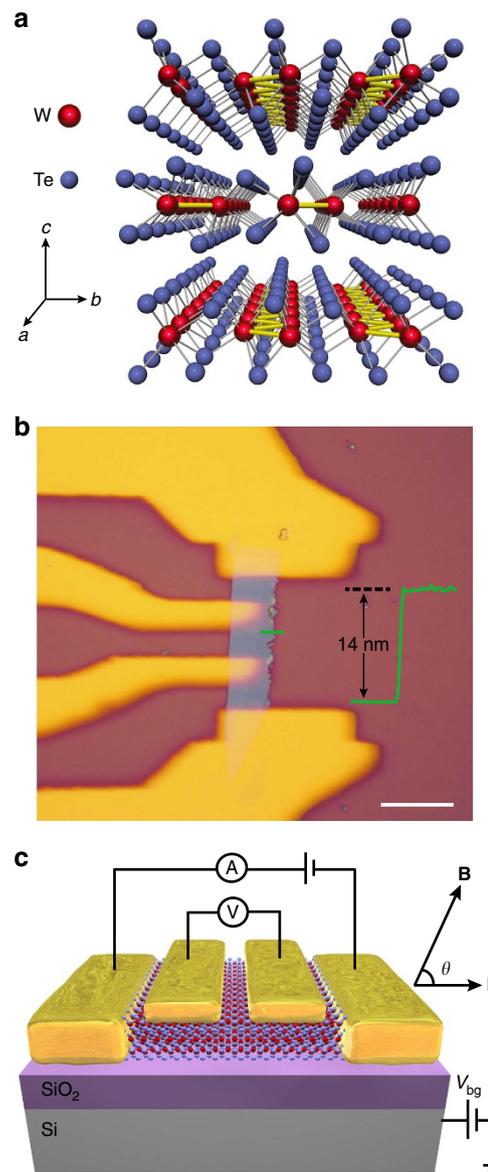


Figure 1 | Thin WTe_2 film devices. (a) The crystal structure of WTe_2 ; the yellow zigzag lines represent the tungsten chains along the a axis. (b) Optical image of a four-probe thin WTe_2 film device. Scale bar, $15\ \mu\text{m}$. Inset: atomic force microscope (AFM) height profile of the flake along the green line. (c) Schematic structure and measurement circuit of the gated four-probe devices. The angle between the magnetic field and current is defined as θ .

Angle-sensitive negative longitudinal MR. To examine the possible signal of the chiral anomaly, we performed longitudinal MR measurements on the devices by applying a magnetic field (from -12 to 12 T) parallel or at small angles to the current direction at 1.6 K. We observe two types of negative longitudinal MR phenomena when \mathbf{B}/\mathbf{I} ($\theta = 0^\circ$), with typical data shown in Fig. 2a (sample #1) and Fig. 2b (sample #2). Both types of negative longitudinal MR exhibit strong angle sensitivity with the strongest signal at $\theta = 0^\circ$ and an apparently suppressed signal at small θ when the magnetic field was slightly rotated (pronounced suppression at $\sim 3.05^\circ$ and -1.75° for samples #1 and #2, respectively). Within a relatively small range of the magnetic field, weak anti-localization (WAL) effect was observed and could be induced by the spin-orbit coupling in WTe_2 (ref. 44). Sample #1 shows only negative longitudinal MR at high magnetic field and the MR begins to decrease at approximately ± 3.5 T and continues over the entire studied magnetic field range (until ± 12 T). Sample #2 shows negative longitudinal MR with a positive MR signal at higher magnetic fields; the MR begins to decrease at approximately ± 1.1 T and subsequently increases from approximately ± 4.7 T. The observed positive longitudinal MR at higher magnetic fields is similar to what has been observed in TaAs ^{26,27} and TaP ^{45,46}. Its physical mechanism is still not clear, even though there are some theoretical proposals such as the Coulomb interactions among the electrons occupying the chiral states²⁶ or the anisotropy of the Fermi surface⁴⁷. In our thin-flake samples, the positive longitudinal MR is much suppressed compared with the reported value (1,200%) in bulk crystals⁴⁰, making the observation of the negative longitudinal MR feasible. To fully understand why the positive longitudinal MR gets suppressed for thinner samples is theoretically challenging at current stage and requires more future research efforts.

While the negative longitudinal MR is rare in non-ferromagnetic materials, it can serve as one of the key transport signatures in WSMs. As the coupled Weyl points have opposite chiralities, the electrons are pumped from one point to the other and lead to a non-zero potential among them if the dot product of the magnetic and electric fields is not 0, that is, $\mathbf{B} \cdot \mathbf{E} \neq 0$. This chiral imbalance-induced potential will induce positive contribution to the conductance. Under the semi-classical approximation, when \mathbf{B}/\mathbf{E} , the anomaly conductivity²³ is described by

$$\sigma = \frac{e^4 v_F^3 \tau B^2}{4\pi^2 \hbar \Delta E^2} \quad (1)$$

where e is the electron charge, v_F is the Fermi velocity near the Weyl points, ΔE is the measured chemical potential from the energy of the Weyl points and τ is the inter valley scattering time. The quadratic relation with a magnetic field leads to a negative MR effect with high sensitivity to the angle between \mathbf{B} and \mathbf{E} , which is consistent with our observations in thin WTe_2 samples.

There are few other origins other than the chiral anomaly, such as current jetting⁴⁸ and magnetic effects⁴⁹, which could induce the negative longitudinal MR effect under certain conditions. As WTe_2 is not a magnetic material, the possible origin of magnetic effects can be safely excluded. The current-jetting effect is usually induced by inhomogeneous currents generated when attaching point contact electrodes to a large bulk crystal. In our thin-film devices (rather than bulk crystals) with well-defined electrodes, it can be excluded as well⁵⁰. Several theoretic proposals related to defects or impurities are also not applicable in our systems. For example, the negative longitudinal MR observed in our samples is not as temperature sensitive as the WAL effect (see Supplementary Fig. 3), suggesting it is not related to the defect-induced weak localization effect. Another theoretical work^{51,52} predicting that certain impurities could induce negative longitudinal MR at small magnetic fields can be

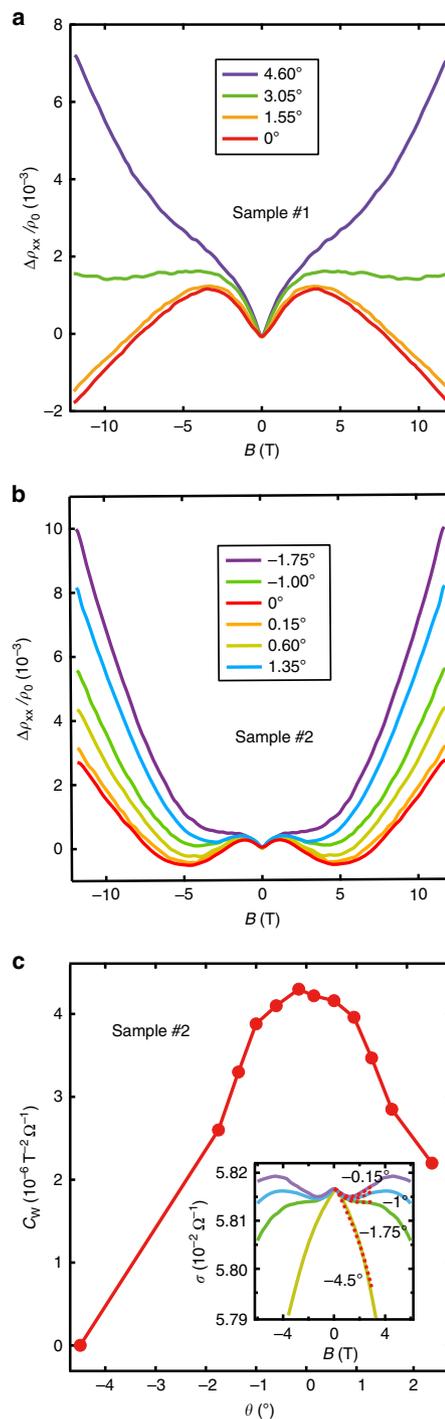


Figure 2 | Angle-dependent negative longitudinal MR of thin WTe_2 .

(a) Sample #1 exhibits only negative longitudinal MR at high magnetic fields, which is apparently suppressed at $\sim 3.05^\circ$. (b) Sample #2 exhibits a negative longitudinal MR and a positive MR signal at higher magnetic field, which is apparently suppressed at approximately -1.75° . (c) The extracted chiral anomaly coefficient C_W for sample #2 was obtained from fittings with the semi-classical formula. The results show strong angle θ sensitivity. Inset: fitting result (red dashed lines) of experimental magneto-conductivity curves (solid lines) at various angles. The MR data were collected at 1.6 K.

excluded, owing to the fact that our observations happen at much higher fields (up to 12 T). In the case of ultra-quantum limit, the impurities were also suggested to induce negative longitudinal MR in any three-dimensional metal, regardless of its

band structures⁵³. To investigate this prediction, by analysing the measured Shubnikov–de Haas oscillations (see Supplementary Fig. 4 and Supplementary Table 1), we carefully calculated the Landau level indexes of different samples exhibiting negative longitudinal MR. The results indicate that the samples remain in the semi-classical limit.

Thus, the negative longitudinal MR can be quantitatively analysed using the formula^{27,54} in the semi-classical limit, which includes the chiral anomaly contribution of the Weyl points:

$$\sigma_{xx}(B) = C_W B_{\parallel}^2 - C_{WAL} \left(\sqrt{B} \frac{B^2}{B^2 + B_c^2} + \gamma B^2 \frac{B_c^2}{B^2 + B_c^2} \right) + \sigma_0 \quad (2)$$

where C_W is the chiral coefficient, C_{WAL} is the WAL coefficient, B_c is the crossover critical field of two regions with different dependences (low field with B^2 dependence and higher field with \sqrt{B} dependence)⁵⁴ and σ_0 is the zero field conductivity when B/I . For small θ values, the term of σ_0 is replaced by $\sigma_0/(1 + \mu^2 B_{\perp}^2)$ to represent the contribution of transverse positive MR, where μ is the mobility. We analysed the angle-dependent longitudinal MR data of sample #2 when $0 \text{ T} < B < 3 \text{ T}$ and extracted the chiral coefficient C_W from the fitting

results. The inset of Fig. 2c shows the fitting results of magneto-conductivity curves at various angles. The extracted C_W versus θ data are plotted in Fig. 2c, revealing that C_W is an effective parameter characterizing the strength of the contribution from chiral anomaly, which exhibits strong longitudinal angle sensitivity.

The anisotropy of negative longitudinal MR. A unique feature of the chiral anomaly in a type-II WSM is the predicted planar orientation dependence of the negative longitudinal MR effect due to the tilted band structure and coexistence of electron and hole pockets. We further examined the crystalline orientation dependence of the longitudinal MR along two principle axes. When the current was applied parallel to the b axis (vertical to the tungsten chains), we observed negative longitudinal MR in all four measured samples (with aforementioned suitable thicknesses of 7–15 nm) (see Supplementary Fig. 5). In sharp contrast, for all four studied samples under similar conditions (exfoliated from the same batch of single crystals and in the identical thickness range) but different current orientation parallel to the a axis (the tungsten chains), only positive longitudinal MR was observed (see Supplementary Fig. 6). These findings support the predicted

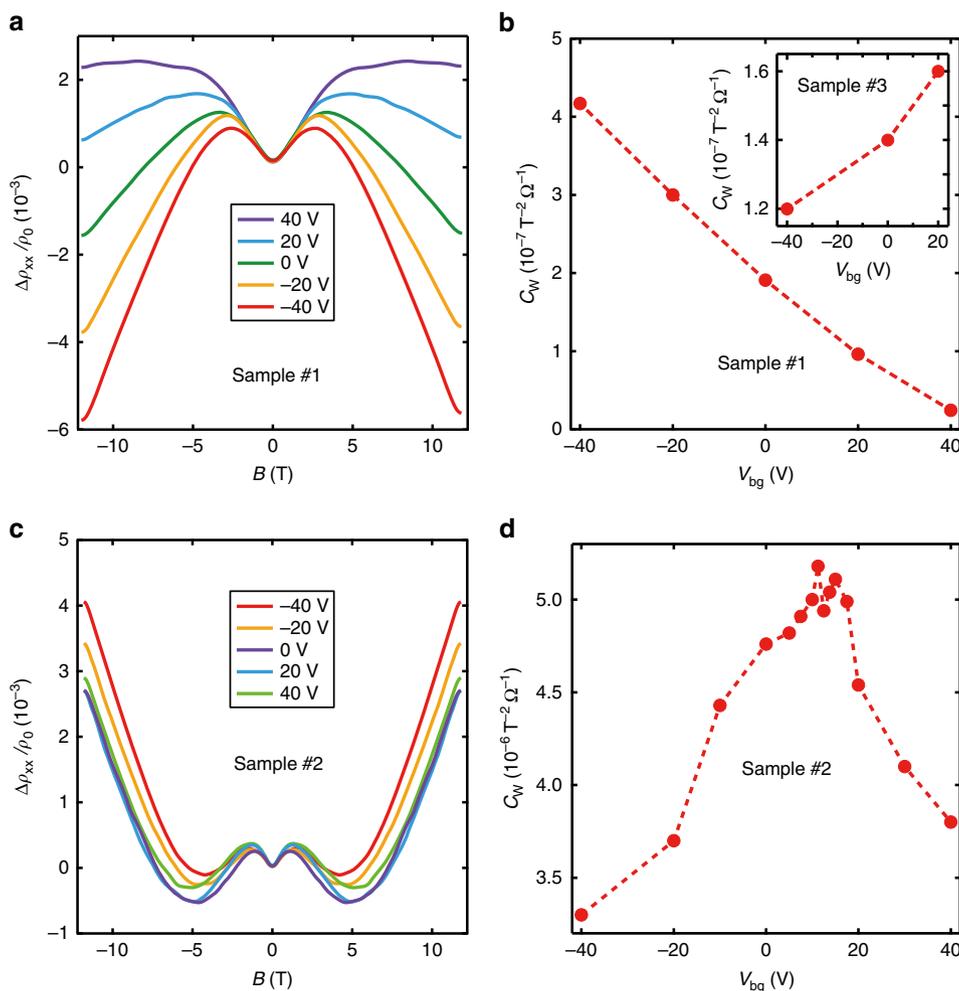


Figure 3 | Gate-tunable negative longitudinal MR of thin WTe₂. (a) The negative longitudinal MR of sample #1 for various V_{bg} , which shows a suppressed negative longitudinal MR effect with increasing V_{bg} from -40 to $+40$ V. (b) Plot of the extracted chiral anomaly coefficient C_W of samples #1 (main) and #3 (inset), showing monotonous decreased/increased C_W with increasing V_{bg} . (c) The negative longitudinal MR of sample #2 for various V_{bg} shows a non-monotonous $C_W - V_{bg}$ dependence with a maximum C_W at certain V_{bg} . (d) The C_W data extracted from the dataset, where the maximum value of C_W occurs at 10–17.5 V.

signature of the type-II Weyl fermion chiral anomaly in thin WTe_2 films (theoretical calculation in Methods).

In-situ tuning of the Fermi energy through the Weyl points. Compared with other experimentally studied WSMs (all bulk materials with fixed doping), another enormous advantage of a thin-layered type-II WSM is the potential for realizing gate tunability, which lies at the heart of modern electronics, and, more importantly, is crucial to verify the negative longitudinal MR as a signature of topological semimetal. The negative longitudinal MR in topological semimetals arises from their ‘monopoles’ in momentum space, which generate a non-trivial Berry curvature that couples an external magnetic field to the velocity of electrons. As a result, an extra chiral current can be induced in parallel magnetic fields, leading to the negative MR. As the Berry curvature diverges at the Weyl nodes^{23,25}, the negative longitudinal MR is expected to be maximized at the Weyl nodes. Therefore, to verify the negative longitudinal MR as a signature of topological semimetal, it is crucial to measure its dependence on the carrier density with a tunable gate voltage *in situ*. So far, no such experiment has been reported in WSMs.

Gate-tunable negative longitudinal MR effect in WTe_2 thin films has been observed in most studied devices. Figure 3a shows the longitudinal MR of sample #1 for various back gate voltages V_{bg} from -40 to 40 V. The negative longitudinal MR was pronounced at -40 V, it was gradually suppressed as V_{bg} increased and was nearly completely suppressed at 40 V, as indicated by the extracted C_W plot in Fig. 3b (see fitting results in Supplementary Fig. 7). This result implies that as V_{bg} increases, the Fermi energy increases and moves away from the Weyl points from above. In contrast, an opposite trend (monotonously increasing C_W with increasing V_{bg}) was observed in sample #3 (as shown in the inset of Fig. 3b and see Supplementary Fig. 8), suggesting that the Fermi energy approaches the Weyl points from below.

More interestingly, a non-monotonous $C_W - V_{\text{bg}}$ curve is observed in sample #2 with C_W maximized at certain V_{bg} . As shown in Fig. 3c, as V_{bg} increases from -40 to 0 V, the native longitudinal MR is gradually enhanced until reaching a maximum between 0 and 20 V. When higher V_{bg} is applied, the native longitudinal MR is apparently suppressed. The C_W data extracted from the complete data set are plotted in Fig. 3d, showing the maximum value of C_W in the range of 10 – 17.5 V. As the anomaly conductivity reaches the maximum, while crossing the Weyl points, these results indicate that we can successfully access the Weyl points via gate tuning. While modulating other bulk WSMs is mostly achieved through chemical/physical doping approaches and the material properties are fixed by the selected composition and doping level during material processing, the *in-situ* tuning of the Fermi energy in layered type-II WSMs could provide an important platform to explore Chiral physics of type-II Weyl fermions.

Discussion

In conclusion, our observations of the angle-sensitive negative longitudinal MR and the strong planar orientation dependence in thin WTe_2 samples reveal important signatures of chiral anomaly in such a predicted type-II WSM. Taking advantage of the thin-film geometry, we successfully demonstrated the *in-situ* tuning of the Fermi energy through the Weyl points, resolving the tunability of unique transport properties and verifying the negative longitudinal MR as a signature of topological semimetal. Our results suggest that gated thin WTe_2 films may constitute a new and ideal platform to control and exploit the unique properties of type-II Weyl fermions (around the Weyl points) using numerous experimental

techniques and pave the way for the implementation of future chiral electronics.

Methods

Materials and devices. The WTe_2 thin films were mechanical exfoliated from single crystals (HQ-graphene, Inc.) onto the silicon substrate covered by 285 nm SiO_2 . The thickness of the samples was confirmed by using a Bruker Multimode 8 atomic force microscopy. The electrodes (5 nm $\text{Ag}/40$ nm Au) were patterned using home-made shadow mask method and deposited by standard electron beam evaporation.

Experimental setup. The devices were measured in an Oxford cryostat with a magnetic field of up to 12 T and based temperature of about 1.6 K. The MR signals were collected by using a low-frequency Lock-in amplifier. A rotary insert (Oxford Instruments) was used to tilt the angle between the magnetic field and current, θ . As the magnitude and direction of the magnetic field is fixed, rotating a device placed on the rotation unit is equivalent to rotating the magnetic field with a fixed device current direction. The rotary insert has precise control on the tilted angle, with error about $\pm 0.05^\circ$.

Details on theoretical calculation of anisotropic chiral anomaly in WTe_2 .

Owing to the C_{2T} symmetry, we can get the general form of the Hamiltonian around a Weyl point, while keeping only terms linear with \mathbf{k}

$$H(\mathbf{k}) = Ak_x + Bk_y + (ak_x + bk_y)\sigma_y + (ck_x + dk_y)\sigma_z + ek_z\sigma_x$$

The energy spectrum of $H(\mathbf{k})$ can be expressed as

$$e_{\pm}(\mathbf{k}) = Ak_x + Bk_y \pm \sqrt{(ak_x + bk_y)^2 + (ck_x + dk_y)^2 + (ek_z)^2}$$

Hence, the kinetic and potential components can be expressed as

$$T(\mathbf{k}) = Ak_x + Bk_y, \quad U(\mathbf{k}) = \sqrt{(ak_x + bk_y)^2 + (ck_x + dk_y)^2 + (ek_z)^2}$$

We can thus define the ratio around the Weyl point

$$R = (T(\mathbf{k}))^2 / (U(\mathbf{k}))^2 = \frac{(Ak_x + Bk_y)^2}{(ak_x + bk_y)^2 + (ck_x + dk_y)^2 + (ek_z)^2}$$

As the direction of $R > 1$ permits the existence of chiral anomaly³², we calculated the values of R along a and b axes. According to the band structure (see Supplementary Fig. 9), for the Weyl points at $E = 52$ meV with respect to the Fermi level, we can get $R = 0.57$ along a direction, whereas $R = 143.68$ along b direction. For the other four Weyl points at $E = 58$ meV, $R = 0.63$ along a direction, whereas $R = 9.3$ along b direction. The calculated results predict the absence of the chiral anomaly along the direction of a axis and the existence of chiral anomaly along the direction of b axis for all Weyl points, which agree well with our observations in experiments.

Data availability. The data that support the findings of this study are available from the corresponding author on request.

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Author contributions

F.M. and Y.W. conceived the project and designed the experiments. Y.W., E.L., J.Z., Y.F., M.W. and K.X. performed the device fabrication and electrical measurements. Y.W., F.M., X.W., H. Lu, B.W., E.L., H.Liu, Y.P. and L.Z. conducted the data analysis and interpretation. Y.W., E.L., Z.H. and Z.W. carried out the Raman spectroscopy measurements and analysis. F.M., X.W., Y.W. and H.Liu co-wrote the paper and all authors contributed to discussions about and the preparation of the manuscript.

Additional information

Supplementary Information accompanies this paper at <http://www.nature.com/naturecommunications>

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