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Temperature Dependent In-Plane Anisotropic Magnetoresistance in HfTe₅ Thin Layers

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We report the observation of in-plane anisotropic magnetoresistance and planar Hall effect in non-magnetic HfTe₅ thin layers. The observed anisotropic magnetoresistance as well as its sign is strongly dependent on the critical resistivity anomaly temperature $T_{\rm p}$. Below $T_{\rm p}$, the anisotropic magnetoresistance is negative with large negative magnetoresistance. When the in-plane magnetic field is perpendicular to the current, the negative longitudinal magnetoresistance reaches its maximum. The negative longitudinal magnetoresistance effect in HfTe₅ thin layers is dramatically different from that induced by the chiral anomaly as observed in Weyl and Dirac semimetals. One potential underlying origin may be attributed to the reduced spin scattering, which arises from the in-plane magnetic field driven coupling between the top and bottom surface states. Our findings provide valuable insights for the anisotropic magnetoresistance effect in topological electronic systems and the device potential of HfTe₅ in spintronics and quantum sensing.

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Negative longitudinal magnetoresistance (NLMR) has been intensively observed in ferromagnetic metals^[1-3] with structural anisotropy.^[4-7] For this</sup> type of NLMR, one origin is the external-field-induced magnetization that reduces the carrier spin scattering and thereby increases the conductivity in electric transport. Nevertheless, in conventional metallic ferromagnets, the NLMR effect is relatively weak and there are too many impurities in material systems, limiting their practical applications in memory or sensing. Recently, large NLMR effect resulted from chiral anomaly was verified in topological electronic systems, e.g., Dirac and Weyl semimetals.^[8–12] However, due to the required chiral charge pumping between separated Weyl nodes, it requests subtle direction alignment of the applied external magnetic field and electric field. The experimental realization of noticeable NLMR in topological materials is still challenging. Therefore, seeking other NLMR effect is of fundamental importance for the integration of topological materials in electronics.

In this Letter, we report our observation of anisotropic magnetoresistance (AMR) in HfTe₅ thin layers with the sign controlled by temperature, which allows to generate large and magnetic-field direction insensitive NLMR effect. HfTe₅ is a layered topological material with a unique lattice structure:^[13] the one-dimensional atomic chain HfTe₃ extends along the a direction together with two tellurium atoms connecting the chains into a plane in c direction, and then stacking in b direction to form the bulk, where a, b, and c directions are set to be x, z, and y axes as shown in Fig. 1(a). Because of the weak bonding strength along the b and c directions, the lattice constants respond differently to temperature. Therefore, HfTe₅ is considered as either a strong or weak topological insulator,^[14] depending on the specific lattice constants. In contrast to bulk systems, where the NLMR

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effect is induced by chiral anomaly,^[12,15] the origin of in-plane NLMR effect in the system of HfTe₅ thin layers is completely different. At cryogenic temperature, the NLMR is observed in most of the in-plane field directions. The magnitude of this NLMR effect can reach up to 30%, which is much higher than that in the conventional ferromagnets.

In experiment, the electronic transport measurements were performed in HfTe₅ thin layers with tens of nanometers. We observed temperature-dependent in-plane AMR. The anisotropy reverses around the critical resistivity anomaly temperature $T_{\rm p}$. Below $T_{\rm p}$, the positive magnetoresistance turns to be negative at most of the in-plane field directions. The NLMR reaches the maximum when the in-plane magnetic field is perpendicular to current, which rules out the origin of chiral anomaly in our study. By measuring the angle and temperature dependence of magnetoresistance, we found that the formation of the Landau levels inhibits the NLMR effect at low temperature and strong magnetic field. In addition, the correlation between the sign reverse temperatures of AMR and T_p suggests that the appearance of the AMR and the NLMR is related to the band structure near the Fermi energy. According to these evidences, we found one origin of these phenomena, i.e., the in-plane magnetic field can shift the Dirac point in k space, and the hybrid coupling between top and bottom surface states can generate angle-dependent net spin polarization that can induce anisotropy and reduce the spin scattering.^[16,17]



Fig. 1. In-plane anisotropic longitudinal and Hall resistance of HfTe₅ thin layers. (a) Crystal structure of HfTe₅. (b) Temperature-dependent longitudinal resistance of HfTe₅. The peak temperature is marked by the dashed line at about 140 K. The inset is schematic of the transport measurements. The current is always along a direction and the magnetic field rotates in ab and ac planes labeled by α and β . (c) In-plane anisotropic magnetoresistance R_{xx} at different temperatures from 20 K to 230 K under 9 T. The black lines are the fitting according to $R_{xx} = R_{\perp} - \Delta R \times \cos^2\beta$, $\Delta R = R_{\perp} - R_{\parallel}$. (d) High (230 K) and low (20 K) temperature anisotropic magnetoresistance under different magnetic fields: 1 T, 3 T, 5 T, 7 T, and 9 T. (e) and (f) Planar Hall effect at 50 K and 180 K under different magnetic fields. Due to the misalignment between the sample and magnetic field, the base line is not zero after symmetrization.

In the sample preparation, we took 99.999% Hf powder and Te powder to grow single crystal of HfTe₅ using the chemical vapor transport method, then mechanically dissociated the bulk with scotch tape and poly dimethyl siloxane (PDMS). There will be some thin-layer samples on the PDMS in about dozens of nanometers. We transferred them to a silicon wafer with 280 nm SiO_2 surface and made a six-electrode pattern by electron beam lithography. The mobility of the HfTe₅ thin layers is about 10000, which is an order of magnitude less than the bulk sample, and the carrier density is about $1.0 \times 10^{18} \text{ cm}^{-3}$, which is about an order of magnitude higher than the bulk sample. We find that the properties of the samples from different batches are slightly different, whereas the NLMR effect always exists.

The in-plane AMR measurement was performed in a physical property measurement system. The temperature range was 2–300 K, the magnetic field range was 0–14 T, and the rotation angle range was 0–360°. The remaining experimental items were measured in the TeslatronPT Dewar of Oxford Instrument of SUST, where the temperature range was 1.5–300 K, and the magnetic field range was 0–13 T. During the measurement, the current was always along *a* direction, and the magnetic field rotated along the *ab* and *ac* planes, which are denoted by α and β , respectively, as shown in the inset of Fig. 1(b).

The in-plane AMR was observed in HfTe₅ thin layers, where the magnetoresistance is defined as $\Delta R_{xx} = \frac{R_{xx}(B) - R_{xx}(0T)}{R_{xx}(0T)}$. Figure 1(c) shows the temperature dependence of AMR. The obvious AMR signals can be found under high magnetic fields even at room temperature. The conventional fitting formula of AMR can be written as^[6]

$$R_{xx} = R_{\perp} - \Delta R \cos^2 \alpha,$$

$$\Delta R = R_{\perp} - R_{\parallel}, \qquad (1)$$

where $R_{\perp}(R_{\parallel})$ represents the longitudinal resistance when the in-plane magnetic field is perpendicular (parallel) to the current. We have fitted the curves at different temperatures according to Eq. (1), as shown in Fig. 1(c) by black lines. One can find that the measured data relatively coincide with the fitted curves at various temperatures. There are two reasons for the slightly deviating parts: (i) The sample placement is not exactly parallel to the magnetic field, and the outof-plane magnetoresistance components are mixed in the data, which can be proved by the analysis of Hall effect R_{xy} . (ii) We have performed symmetry analysis on the data to remove the Hall component by adding the data under positive and negative magnetic field, but this operation cannot cancel the planar Hall resistance, so that the planar Hall resistance is also mixed in the data, making the data deviate from the fitting. The data at 150 K is obviously different from the others and the inversion of anisotropy also occurs near this temperature: $\Delta R = R_{\perp} - R_{\parallel} > 0$ at higher temperatures while $\Delta R = R_{\perp} - R_{\parallel} < 0$ at lower temperatures. We find that this phenomenon is closely related to the temperature-dependent resistance peak $T_{\rm p}$ of $HfTe_5$, as shown in Fig. 1(b), because the peak temperature of 140 K is close to the anisotropy inversion

temperature. In addition, in the temperature range of $\Delta R = R_{\perp} - R_{\parallel} < 0$, the NLMR effect occurs in most of the in-plane directions and reaches the maximum at the perpendicular direction of $\beta = 90^{\circ}$, which is completely different from the NLMR effect induced by chiral anomaly.^[11,12] Figure 1(d) shows the magnetic field dependences of AMR at high temperature (230 K) and at low temperature (20 K). The AMR under different magnetic fields agrees with Eq. (1), and the higher the magnetic field, the larger the AMR.

Furthermore, we also study the variation of the Hall resistance R_{xy} around the inversion temperature. The raw data can be found in Section I of the Supplementary Materials. Hall magnetoresistance shows obvious sinusoidal dependence, which means that the sample is not exactly parallel to the magnetic field. There will be a sinusoidal out-of-plane perpendicular magnetic field component during rotation, thus the Hall signal varies sinusoidally with the angle. Adding the positive and negative fields' data can remove the Hall resistance brought by the out-of-plane magnetic field. However, the base line of Hall magnetoresistance is not at zero because some longitudinal signals are inevitably doped in the data. The whole analysis procedure can be found in Section I of the Supplementary Materials. Figures 1(e) and 1(f) show the Hall magnetoresistance at 50 K and 180 K, respectively. It can be found that there is a significant sinusoidal Hall resistance at low temperature, but not at high temperature. The conventional fitting formula of the planar Hall resistance can be written as

$$R_{xy} = -\Delta R_{\rm H} \sin \alpha \cos \alpha. \tag{2}$$

The black fitting line in Fig. 1(e) shows that the data fits well, indicating that the Hall signal is actually the planar Hall resistance. It is worth mentioning that the parameters in fittings of Eqs. (1) and (2) are different, i.e., $\Delta R_{\rm H} \neq \Delta R$, implying that the planar Hall effect may be induced by multiple reasons.

Next, we study the angle dependence of negative longitudinal magnetoresistance. When the temperature is lower than the inversion temperature, we could observe the in-plane NLMR in HfTe₅ thin layers. Since the signal itself can arise from an extrinsic origin, i.e., current $jetting^{[18]}$ that is a measurement error caused by the non-uniform distribution of current, we first confirm the source of the NLMR using a special electrode structure in sample S7. We prepare a six-electrode pattern and make sure that the electrodes completely vertically go across the sample, ensuring that the measured signals are not affected by the uneven current distribution (see detailed information in Section II of the Supplementary Materials). One can find that the NLMR effect can still be measured in this sample, and both angle and temperature variation trends are completely consistent with those of the ordinary sample, indicating that the NLMR signal is an intrinsic characteristic of the sample itself, rather than the measurement error.



Fig. 2. Out-of-plane magnetic field dependence of NLMR effect of HfTe₅ with $\Delta R = \frac{R(B) - R_0}{R_0}$. (a) The α dependence of magnetoresistance from 2.00° to -2.15° when we fix $\beta = 30^{\circ}$. (b) The data after subtracting the *b* direction component according to different α from (a). (c) The α dependence of magnetoresistance from 4.37° to -3.66° when we fix $\beta = 120^{\circ}$. (d) The data after subtracting the *b* direction component according to different α from (c).

Then, we carefully study the angle and temperature dependences of the NLMR effect. In some samples, the amplitude of NLMR can reach tens of percent, which is much higher than those in some common ferromagnetic materials. Figure 2(a) shows the α dependence of NLMR. The magnetic field is mainly placed in plane with $\beta = 30^{\circ}$, and it slightly deviates from the plane by a small angle α . The precise angle α in the figure comes from the Hall measurements. By comparing the magnetic fields with the same Hall resistance between in-plane and out-of-plane directions, one can determine the component of the magnetic field offset in b direction and calculate the α angle. The detailed calculation method is provided in Section II of the Supplementary Materials. Figure 2(a) shows that as the out-of-plane component increases, the NLMR quickly disappears. A small angular offset can induce a large positive magnetoresistance component that covers the NLMR signal, because of the large positive magnetoresistance of $HfTe_5$ in b direction. Since the deflection angle α is small and the sample is very anisotropic, we can treat the total magnetoresistance as the vector sum of the in-plane and out-of-plane magnetoresistances. By subtracting the bdirection component, one can get the in-plane component shown in Fig. 2(b). All the curves with different deflection angles are coincident, confirming that the NLMR signal is indeed caused by the in-plane magnetic field.

When changing the in-plane magnetic field to $\beta =$

 120° , as shown in Fig. 2(c), we can find some similar characteristics, i.e., the positive magnetoresistance component outside the plane will cover the NLMR signal, and after the component in *b* direction is subtracted, the curves are partly coincident, as shown in Fig. 2(d). The data are only overlapped in the low magnetic field, whereas the NLMR signal is greatly suppressed in high magnetic field due to the SdH oscillations. This indicates that when the Landau levels are formed in the presence of high magnetic field, the NLMR effect becomes obviously suppressed, implying that the existence of NLMR effect may be related to the band structure near the Fermi energy.



Fig. 3. Temperature-dependent negative longitudinal magnetoresistance of HfTe₅. (a) Temperature dependence of negative longitudinal magnetoresistance from 10 K to 180 K, where $\alpha = 0^{\circ}$, $\beta = 75^{\circ}$. The inset gives the zoomin display at high temperatures from 120 K to 180 K, where negative and positive magnetoresistance coexist. (b) The temperature dependence of amplitude of magnetoresistance ΔR under different magnetic fields.

Now we continue to study the effect of temperature on NLMR effect, i.e., temperature dependence of negative longitudinal magnetoresistance, as shown in Fig. 3(a). When the magnetic field is applied in different in-plane directions, it shows the same variation trend, i.e., as the temperature increases, the amplitude of the NLMR gradually decreases. However, it always keeps a good NLMR profile till reaching the inversion temperature. Below the inversion temperature, the NLMR effect will be more obvious in higher magnetic field at lower temperature, as shown in Fig. 3(b). Above the inversion temperature, the negative longitudinal magnetoresistance is gradually replaced by the positive magnetoresistance from low to high field. When the temperature reaches $\sim 180 \,\mathrm{K}$, the NLMR effect completely disappears. In bulk HfTe₅, the angular resolution photoelectron spectroscopy (ARPES) and transport measurements suggest that the Fermi level can shift from the valence band to conduction band as the temperature decreases, and the Fermi level will go through Dirac point at temperature $T_{\rm p}$, so that the correlation between the critical temperature of the NLMR and $T_{\rm p}$ confirms again that the existence of NLMR is closely related to the special band structures. It is noteworthy

that we do not find distinct evidences for the transition of Fermi energy by transform measurements in the thin layers' system. In the range of 2–300 K, the major carriers are always holes (see Section IV of the Supplementary Materials). In following study, if the Fermi energy in the thin-layer system shares the same transition tendency as the bulk, it is expected to be experimentally verified by ARPES measurements.

Summarizing the experimental results, we can find that the inversion of AMR, and the appearance of planar Hall effect and NLMR effects are all related to the resistivity anomaly temperature $T_{\rm p}$. Meanwhile, the formation of Landau levels also strongly suppresses the NLMR effect. Therefore, we attribute the appearance of these phenomena to the topological electronic band structures of HfTe₅. According to some similar experimental phenomena,^[16,17,19,20] we believe that the temperature dependent anisotropy and the NLMR effect could be induced by the hybrid coupling of surface states, in which the carriers are spin-momentum locked. The Dirac-like surface electronic band structure is displayed in Fig. 4(a). When an in-plane magnetic field is applied, a momentum shift occurs in the band structure of the top and bottom surface states. Therefore, the top and bottom surfaces will hold two kinds of carriers with completely opposite spin polarizations. When the sample is thin enough, the hybrid coupling between the top and bottom surface states can generate a net spin polarization S_{net} , which can suppress the spin scattering and induce negative longitudinal magnetoresistance, as shown in Fig. 4(b). Different directions of in-plane magnetic field can change the net spin directions, resulting in AMR. In addition, when the Fermi level lies in the conduction band or valence band, the directions of S_{net} are opposite, corresponding to the inversion of AMR around T_{p} .

Furthermore, we find that the NLMR only exists in the samples with thickness of tens of nanometers, as shown in Table 1. In thick samples, the coupling between surface states becomes weaker, so that the NLMR effect disappears, which can be verified in samples S8, S9 and S10. Although the NLMR effect should be more pronounced in thinner samples, we do not observe it in thinner samples S1 and S2. We also find that the temperature-dependent longitudinal resistance peak $T_{\rm p}$ disappears in these thinner samples (see detailed information in Section V of the Supplementary Materials). Usually, the Fermi energy slightly rises as the temperature decreases. Therefore, the absence of $T_{\rm p}$ indicates that the Fermi energy is deeply located inside the valence band. Consequently, the bulk states are dominant in electrical transport, and covering the negative longitudinal magnetoresistance.



Fig. 4. The schematic diagram of AMR and NLMR effect. (a) The topological surface states of HfTe₅ (001) plane. (b) Shifted band structure of top and bottom surface states under in-plane magnetic field and the generation of net spin polarization $S_{\rm net}$. The red and blue arrows represent the spin polarization direction.

Sample thickness	Abnormal cooling peak $T_{\rm p}$	Negative magnetoresistance $(B \parallel c \text{ axis})$
S1-16 nm	No	No
S2-25nm	No	No
S3-28 nm	Yes	Yes
$S4-35\mathrm{nm}$	Yes	Yes
$S5-38\mathrm{nm}$	Yes	Yes
$S6-51\mathrm{nm}$	Yes	Yes
m S7-60nm	Yes	Yes
S8-88 nm	Yes	No
S9-100 nm	Yes	No
$\mathrm{S10} ext{-}330\mathrm{nm}$	Yes	No

Table 1. Negative longitudinal magnetoresistance in different thickness samples.

In conclusion, we have observed distinctive electrical transport phenomena in non-magnetic $HfTe_5$ thin layers, e.g., in-plane anisotropic magnetoresistance, planar Hall effect and negative longitudinal magnetoresistance effect. In some samples, the amplitude of NLMR can reach tens of percent, which is much higher than those in common ferromagnetic materials, such as iron, cobalt, nickel and their alloys. All these effects are found to be related to the resistivity anomaly $T_{\rm p}$. Especially, the in-plane AMR reverses around temperature $T_{\rm p}$. The corresponding physical origin of these effects can be attributed to the hybrid coupling of surface states that produces a net polarized spin $S_{\rm net}$ in the presence of in-plane magnetic field, thus leading to anisotropic spin scattering and reducing the resistance.

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