Supplementary Materials: Temperature dependent in-plane anisotropic magnetoresistance in $HfTe_5$ thin layers

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Section I: Data processing of planar Hall effect

Due to slightly unparallel between the sample and the magnetic field , a small amount of out-of-plane magnetic field component that changes sinusoidally with the angle β is doped into the data, which in turn produces a sinusoidally varying Hall resistance, shown as the red and blue lines in Fig. S1(a). By combining the Hall data under the same intensity of positive and negative magnetic fields, we can remove the out-of-plane Hall component, thereby obtaining a pure planar Hall effect resistance, shown as the green line in the Fig. S1(a). An obvious planar Hall effect with a period of π appears, but this symmetry analysis inevitably imports a small amount of longitudinal resistance, which shifts the baseline from 0. According to the plane Hall effect formula:

$$R_{xy} = -\Delta R_H \times \sin\alpha \times \cos\alpha \quad (1)$$

we find that the fitting is relatively coincident with the green line, indicating that the data showed in green line describe the planar Hall effect. Figure S1(b) shows the raw data of the Hall effect measured under different in-plane magnetic field. Through the same processing method, we get Fig. 1(e) in the text.

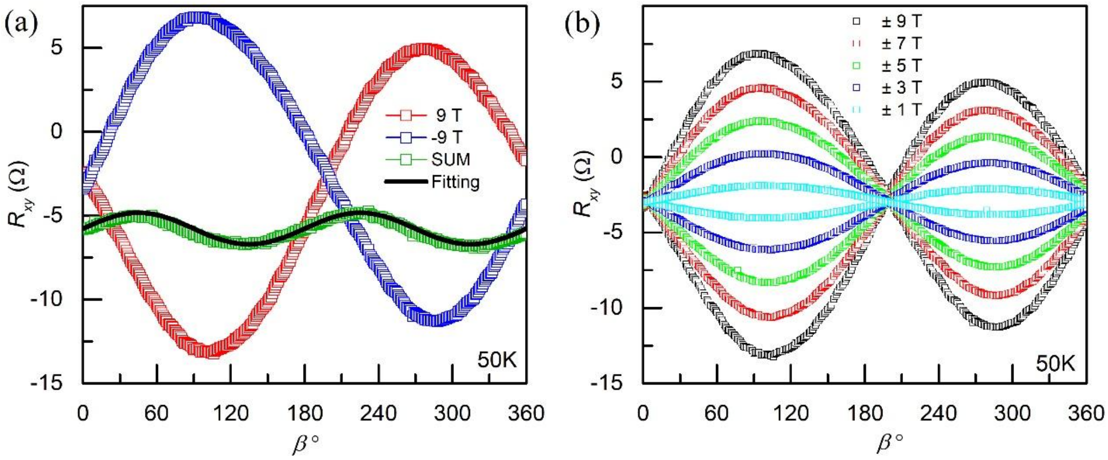


FIG. S1. In-plane angle-dependent Hall signal and planar Hall effect. (a) The planar Hall effect resistance (green line) obtained by symmetrization. The blue and red line are the raw data at 9 T and -9 T, and the black line is the fitting according to Equation (1). (b) The raw data of Hall signal under different in-plane magnetic fields.

Section II: Eliminating the extrinsic origin of negative longitudinal magnetoresistance

In order to confirm whether the negative longitudinal magnetoresistance is intrinsic or extrinsic (current jetting), we apply specific electrodes pattern onto the sample S7, as shown in Fig. S2(a), where all the electrodes completely vertically go across the sample, assuring the same potential. So that the measured signal will not be affected by uneven current distribution in this sample. And the same angle and temperature dependence of negative longitudinal magnetoresistance are also observed in sample S7, as shown in Fig. S2(b) and S2(c).

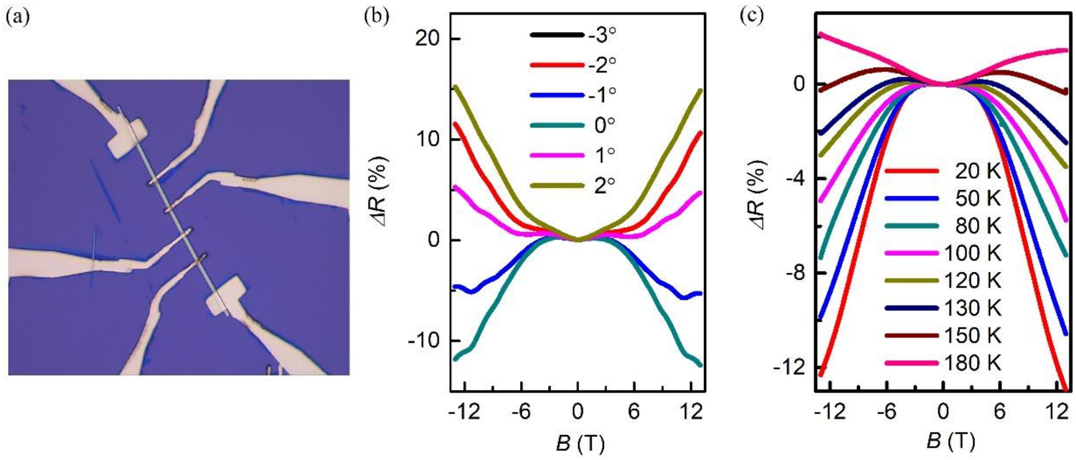
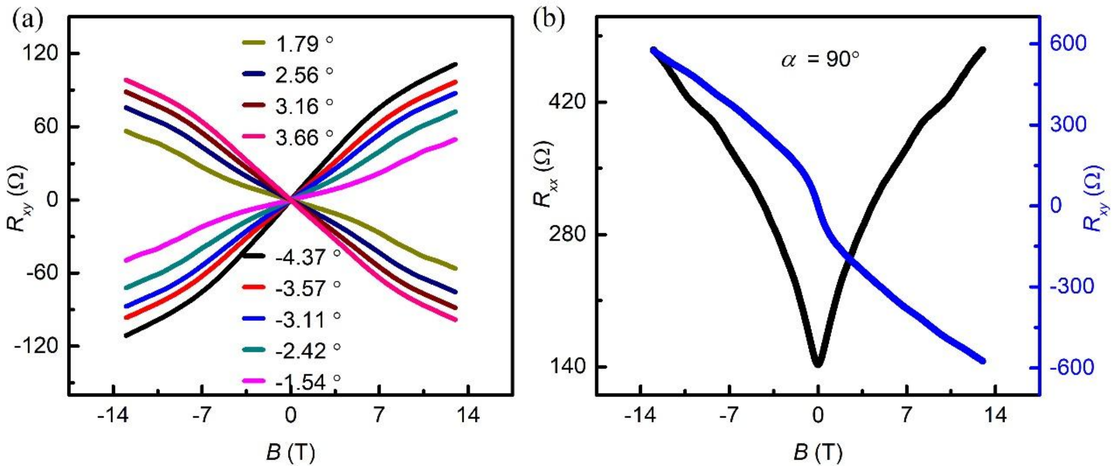


FIG. S2. Negative longitudinal magnetoresistance in the sample with homogeneous current distribution. (a) The microscope pattern of the sample S7. (b) Out-of plane angle-dependent negative longitudinal magnetoresistance where the magnetic fields mostly lie in *ac* plane with $\beta = 30°$, and slightly deviates with $\alpha = -3°, -2°, -1°, 0°, 1°, 2°$. (c) Temperature-dependent negative longitudinal magnetoresistance from 20 K to 180 K where the magnetic field is fixed in $\alpha = 0°, \beta = 30°$.

Section III: Detecting deflection angle α by Hall resistance

We calculate the detailed angle α in Fig. 2 by comparing the Hall resistance between out-of-plane perpendicular Hall resistance and in-plane Hall resistance. In Figure S3, the slope is contrary for the angle α with different sign. As the red line with $\alpha = 3.66°$, when in-plane magnetic field reaches 13 T, the Hall resistance is about 93 Ω which is all from the deviated (out-of-plane) component of magnetic field. We find that in Fig. S3(b), when magnetic field is out-of-plane perpendicular to the current, the same 93 Ω Hall resistance realizes at 0.83 T which equals to the deviated component mentioned when the magnetic field is in plane. So that we can infer the

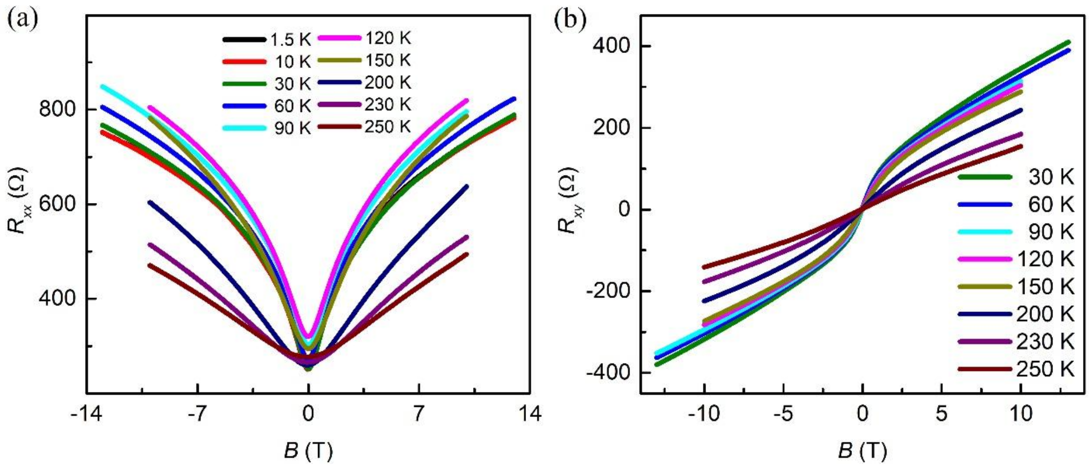


precise α from the trigonometric function $\alpha = \arcsin\left(\frac{0.83\ \mathrm{T}}{13\ \mathrm{T}}\right) = 3.66°$. The other precise angles in Fig.S3(a) are deduced by the same method.

FIG. S3. Comparison of Hall resistance between in-plane and out-of-plane magnetic field. (a) In-plane Hall measurement where the Hall resistance is from the deviated (out-of-plane) component of the magnetic field. The legends are deflection angles α. (b) Out-of-plane longitudinal and Hall measurements.

Section IV: Temperature dependence of longitudinal and Hall resistance

In thin-layers samples around dozens of nanometers, the Hall slope is always positive rather than inversing around temperature T_p, as shown in Fig. S4(b). We notice that the Hall resistance changes from linear to nonlinear when decreasing the temperature, which means that there might be more than one kind of carriers participating the transport process. This could be caused by the shift of Fermi level during decreasing the temperature. But we still can not conclude that the Fermi level goes through Dirac point at T_p by transport measurements. Whether the process occurs in thin-layers samples still needs further exploration and research, such as



ARPES.

FIG. S4. Temperature-dependent longitudinal and Hall measurements. (a) Longitudinal magnetoresistance measurements under different temperature. (b) Hall measurements under different temperature. It shows that the slope is always positive under different temperature which is different from the bulk samples that the slope is inversed at T_p.

Section V: Electrical transport measurement of samples with different thicknesses

In both thicker and thinner samples, we have not observed the in-plane NLMR, as shown in Fig. S5 (b). The resistivity anomaly peak T_p of $HfTe_5$ indicates that the Fermi surface goes through the Dirac point. By comparing the temperature dependent resistance of samples with different thicknesses, we find that as the thickness decreases, T_p gradually disappears, indicating that the Fermi energy may be deeply located inside the valence band. So that the system mainly exhibits the transport properties of bulk states at this time, and the NLMR effect cannot be observed.

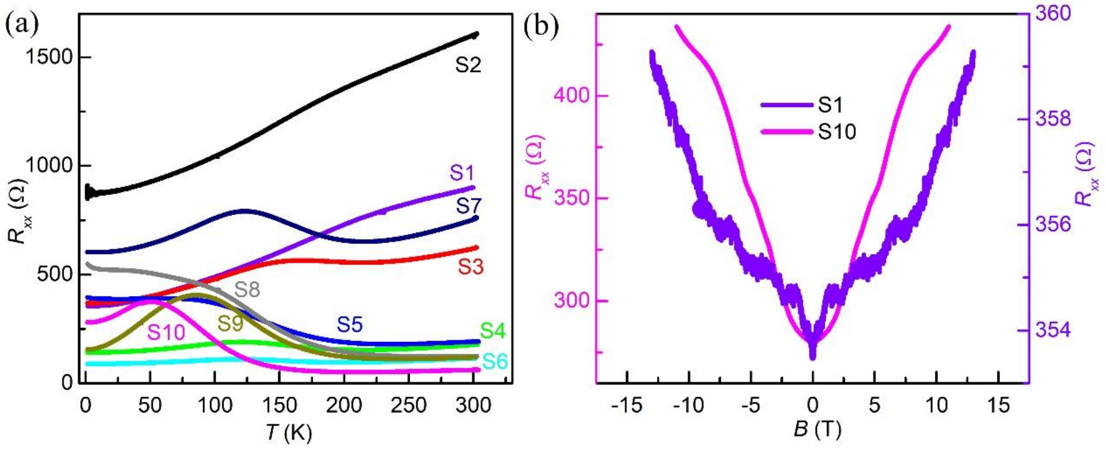


FIG. S5. Temperature and magnetic field dependence of longitudinal resistance for samples with different thickness. (a) Temperature dependence of longitudinal resistance for sample S1 to sample S10 in table I. (b) In-plane perpendicular magnetic field dependence of longitudinal resistance for sample S1 and sample S10. The NLMR effect disappears in both samples.