

Fermi surface and nested magnetic breakdown in WTe₂

Supplemental Material

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ASSIGNMENT OF ORBITS

Figure S1 shows a Fast Fourier Transform (FFT) from the data of Fig. 1(c) in the main text in the range [5-13.3]T. The main peaks below 200 T correspond to extremal areas of individual pockets in the Fermi surface of WTe₂. For the assignment of the individual electron and hole pockets α , β , γ and δ , we follow the labelling of Zhu *et al.* [1]. Their carrier type, hole-like for α and δ , electron-like for β and γ , has been identified from the density functional theory (DFT) calculations.

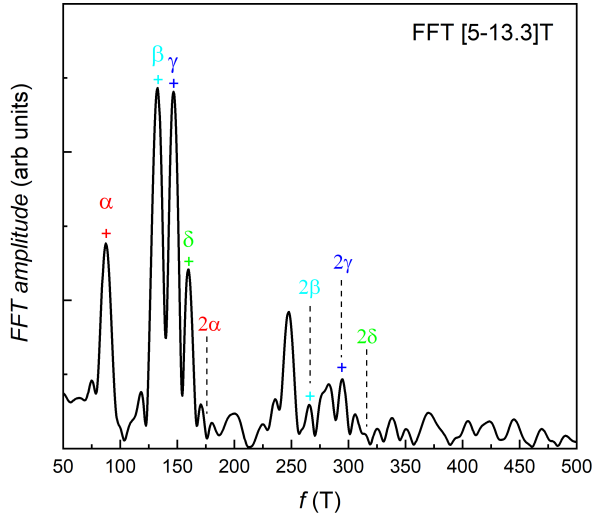


FIG. S1: FFT spectrum in the field range [5-13.3]T at 0.34 K for $B \parallel c$.

In this field range, all individual pockets for α , β , γ and δ are well resolved. In addition, we attribute some peaks in the FFT spectrum to their first harmonics 2β and 2γ .

DENSITY FUNCTIONAL THEORY CALCULATIONS

DFT electronic structure calculations were performed using a full-potential linearized augmented plane-wave plus local orbital method as implemented in the Wien2K package [2]. The experimental structure was used [3] along with the PBE-GGA exchange correlation potential [4]. The spin-orbit interaction was included on both W and Te using a second variational method [2]. The calculation was converged with 10^4 k -points in the full Brillouin zone. To calculate accurately the quantum oscillation (QO) frequencies a dense k -mesh was used with k -points spacing of 2×10^{-3} , 4×10^{-3} , 8×10^{-3} in reciprocal lattice units (rlu) along the a^* , b^* and c^* directions respectively. The difference in k -point spacing (in rlu) results in an approximately uniform mesh in ($\text{\AA}^{-1}$) units because of the different lattice constants in the a, b and c directions. For $B \parallel c$ a single slice at $z = 0$ was used, whereas the full 3-D structure was needed to calculate the angle dependence. To determine the extremal areas the Fermi-surface was sliced at 100 positions along the $k \parallel H$ direction. Calculated Fermi areas A_k were converted into QO frequencies using the Onsager relation $f = (\hbar/2\pi e)A_k$ and masses calculated from $m_b = (\hbar^2/2\pi)dA_k/dE$.

The calculation with spin-orbit interaction included but without an orbital potential ($U = 0$) gives hole and electrons bands, similar to Fig. 4 in the main text, but with the symmetric pockets of the larger hole band joining together across Γ . This joining is caused by a very flat band, close to Γ (see Fig. S2), and a very small shift (7 meV) of the band energy causes the Fermi surface to revert to the 2 sets of 2 nested pockets shown in Fig. 4. These 4 pockets have 4 extremal frequencies which are reasonably close to the experimental areas particularly when viewed as a fraction of the total Brillouin zone area which is equivalent to a QO frequency of 19 kT. However, with this $U = 0$ calculation, the masses of the hole pockets are ~ 2 times larger than experiment, which re-

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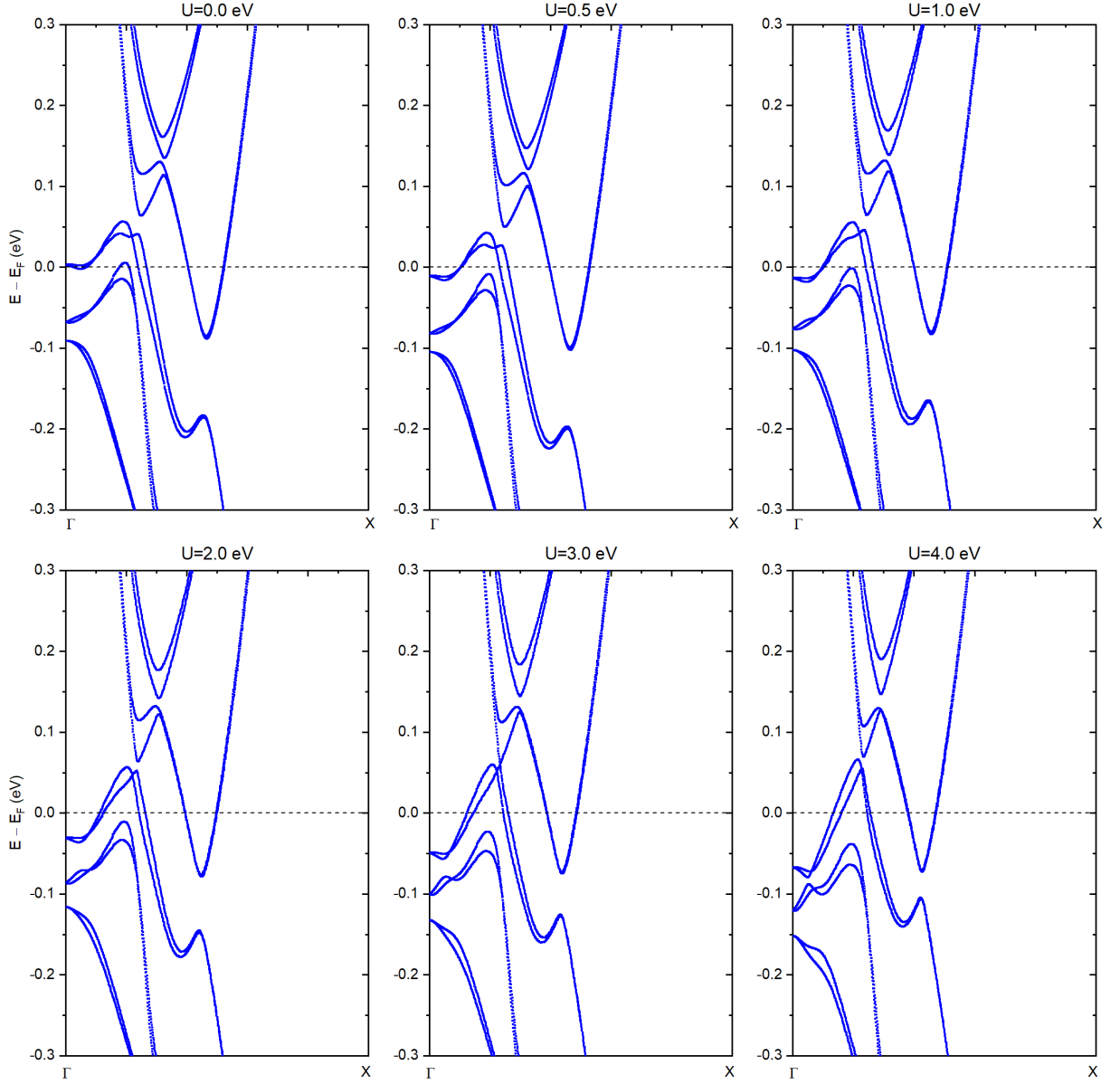


FIG. S2: Band-structure, energy-momentum along the $\Gamma - X$ direction for several values of U .

sults from the influence of the aforementioned flat band. Often the measured mass $m^* > m_b$ because of many-body effects not taken into account within DFT (e.g., the electron-phonon interaction), but the opposite $m^* < m_b$ rather indicates a deficiency in the DFT calculation at the one-electron level. To improve agreement with experiment, we therefore performed calculations with $U > 0$. The effect of U is to depress the energies of the portion of the bands near Γ but leaving the rest of the bands close to E_F relatively unchanged (see Fig. S2). For U of a few eV the hole band becomes much more symmetric (v_F becomes much more isotropic) and the portion of low v_F which causes the high value of m is removed. Similar conclusion were made by Di-Sante *et al.* [5] who

TABLE S1: Calculated quantum oscillation parameters for $U = 5$ eV, compared to experimental values. A shift of 8 meV for the hole bands, changes the frequencies(masses) to 72 T($0.34 m_e$) and 178 T($0.51 m_e$) for bands 1 and 2 respectively.

band	Type	f (Exp)	m^* (Exp)	f (DFT)	m_b (DFT)
1	h	88 T	0.47 ± 0.06	98 T	0.40
2	h	158 T	0.46 ± 0.04	216 T	0.58
3	e	148 T	0.40 ± 0.03	145 T	0.37
4	e	133 T	0.40 ± 0.02	128 T	0.34

also found LDA+ U with $U \sim 2$ eV provided the best agreement with their ARPES measurements on WTe_2 .

The changes in the frequencies and masses as a func-

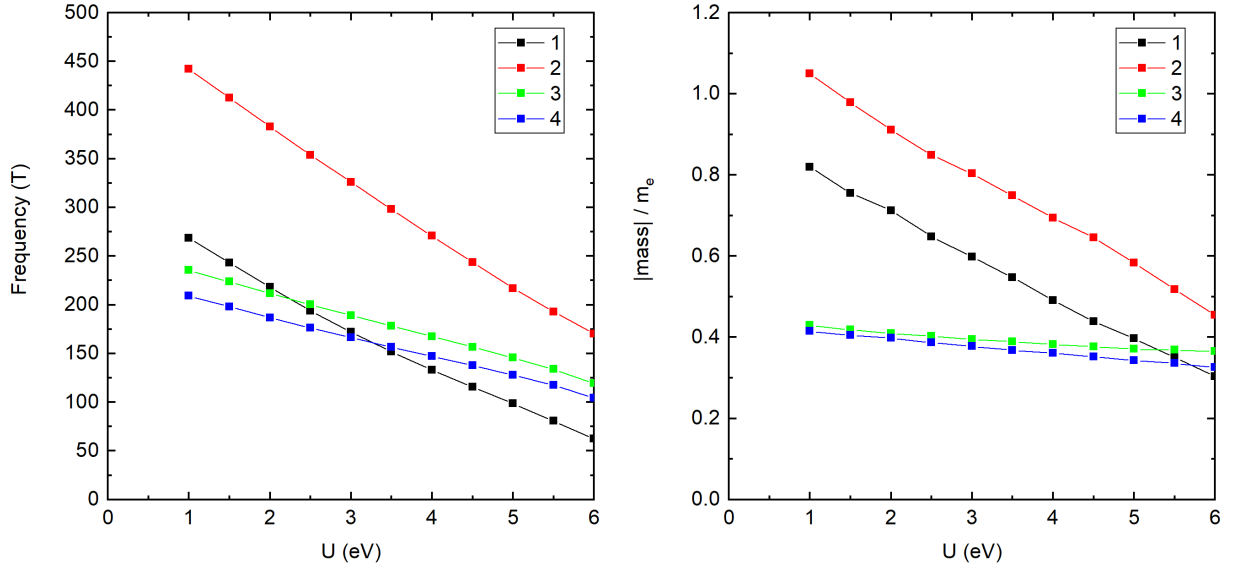


FIG. S3: Calculated frequencies and masses for the 4 pockets as a function of U . Bands 1 and 2 are the hole bands, and 3 and 4 the electron bands.

tion of U are shown in Fig. S3. In the main text, we use a value of $U = 3$ eV to maintain approximate agreement with the ARPES study [5] which in contrast with our experiment, also probes the band-structure away from the Fermi level but has much less accuracy in terms of Fermi pocket size and masses and is potentially surface sensitive. This value of U still results in frequencies and masses which are not in perfect agreement with experiment. In order to provide the best match we make small energy shifts of the bands. For the hole bands $\Delta E = -23$ meV and for the electron bands $\Delta E = +10$ meV. These shifts are in opposite directions, making both pockets smaller in size and approximately maintaining charge neutrality (note we cannot rule out a very small doping in our samples). For the electron bands this procedure results in DFT frequencies which are within a few Tesla of the experimental ones with almost exactly the same difference between the pockets. For the holes, the difference in area between the two orbits is larger than experiment. However, we have chosen not to shift the two pockets by different energies so as to preserve the size of the Dresselhaus splitting which is important for the magnetic breakdown gaps. Instead we have chosen the energy shift so that the average frequency is the same as the average experimental value.

Our solution for tuning the DFT is not unique, a similar solution could be found for a larger value of U . For example, with $U = 5$ eV the calculated parameters are shown in Table S1. The electron-band frequencies are now in almost perfect agreement with experiment without any shift in energy, although the masses are slightly too low. The hole-bands however, still require a small shift to bring the average frequencies in line with experi-

ment, but this is much smaller (8 meV) than when compared to the $U = 3$ eV result. Again the splitting of the hole bands is higher than in experiment. Although our solution is not perfect, it is very close to experiment and shows clearly the nature of the deficiency of the $U = 0$ result. It is of course possible that better agreement could be found using more sophisticated methods to handle the Coulomb interaction, but this is beyond the scope of the present work.

For $U = 3$ eV, the resultant QO frequencies calculated for field rotations from $H \parallel (001)$ to either (010) or (100) are shown in Fig. S4. It can be seen that there are marked differences in the evolution of the frequencies for the two rotation directions, which reflects the shape of the Fermi surface. The electron Fermi surfaces can roughly be approximated by an ellipsoid with axes ratio $(k_{a^*} : k_{b^*} : k_{c^*}) = (0.22 : 0.44 : 1)$ (see Fig. S5). For an ellipsoid the area (and hence frequency) for rotation from $H \parallel (001)$ to $H \parallel (010)$ varies as

$$A_k^{c^* \rightarrow b^*} = \frac{\pi k_{a^*} k_{c^*}}{\sqrt{\sin^2 \theta + \left(\frac{k_{c^*}}{k_{b^*}} \cos \theta\right)^2}}$$

whereas for the rotation $H \parallel (001)$ to $H \parallel (100)$ it is

$$A_k^{c^* \rightarrow a^*} = \frac{\pi k_{b^*} k_{c^*}}{\sqrt{\sin^2 \theta + \left(\frac{k_{c^*}}{k_{a^*}} \cos \theta\right)^2}}.$$

With $\theta = 0$, $A_k = \pi k_{a^*} k_{b^*}$, whereas for $\theta = 90^\circ$, $A_k^{c^* \rightarrow b^*} = \pi k_{a^*} k_{c^*}$ and $A_k^{c^* \rightarrow a^*} = \pi k_{b^*} k_{c^*}$. For $k_{c^*} \gg (k_{a^*}, k_{b^*})$ then $A_k \sim 1/\cos \theta$, as for a pure two-dimensional cylindrical Fermi surface. As $k_{b^*} \simeq 2k_{a^*}$ it can therefore be understood why the increase with θ is

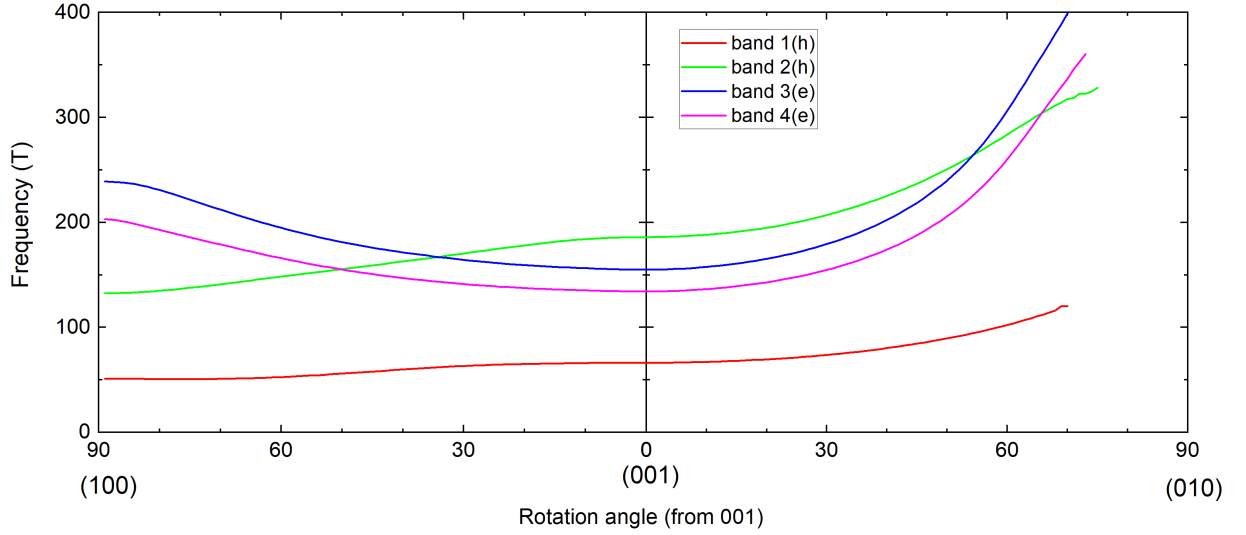


FIG. S4: Calculated quantum oscillation frequencies as a function of field angle for $U = 3$ eV. Bands 1 and 2 are the hole bands, and 3 and 4 the electron bands.

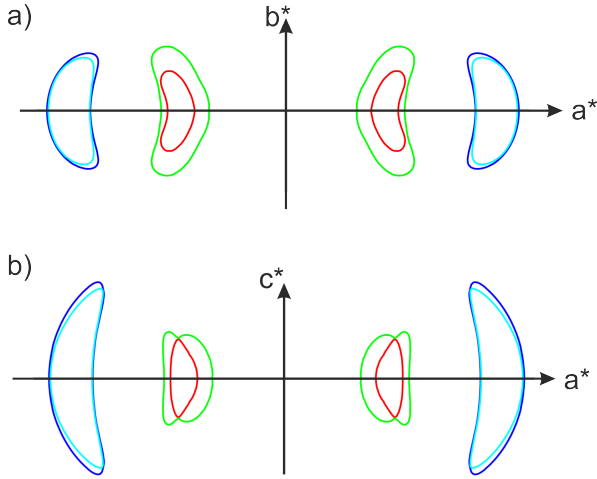


FIG. S5: 2D Fermi contours (a) in the $\Gamma - X - Y$ plane, and (b) in the $\Gamma - X - Z$ plane, for $U = 3$ eV with energy shifts: hole bands $\Delta E = -23$ meV and electron bands $\Delta E = +10$ meV

larger for the rotation towards (010) compared to (100). Furthermore, as $k_{c^*} \simeq 2k_{a^*}$ it can be understood why for the rotation towards (010) $F \sim 1/\cos \theta$ for $\theta \lesssim 40^\circ$ (see Fig. 2).

MASS DETERMINATION

As described in the main text, the mass m^* was determined by fitting R_T to the amplitude of the FFT of dR/dB , between $B_{\min} = 5$ T and $B_{\max} = 28.7$ T. B in the formula for R_T is set to the average inverse field in this interval $1/\langle B^{-1} \rangle$. In principle, determining m^* in this way can lead to an underestimate of the mass because if

there is significant variation of the QO signal amplitude over the FFT field window then the effective B in the R_T expression will differ from $1/\langle B^{-1} \rangle$. If the damping with field is due solely to R_T the error is minimal, but if there is significant impurity scattering or other (non-exponential) terms then the fitted m^* can be less than the true value. If the field variation of the signal is known, then this effect can be checked by doing the same analysis on simulated data with a known input mass. Unfortunately, in the present case the frequencies are too closely spaced to do this for all the orbits. However, the α orbit is sufficiently separated from the rest to perform two different checks. The first is simply to vary $B_{\min}$ as we have done in Fig. 3(c) of the main text. The minimal change of m^* as $B_{\min}$ increases up to the limit where the β FFT peak starts to merge with the α peak, indicates there is no significant underestimate.

The second check is to use a Fourier filter to isolate the α oscillations from the rest of the signal and then perform an inverse FFT transformation on this filtered data. To do this we used a smooth band pass filter with $F_{\min} = 75$ T and $F_{\max} = 105$ T with 5 T rise width to isolate the α signal. The filtered data is shown in Fig. S6(a) for the temperatures indicated. A temperature dependent amplitude can be obtained by fitting the data between 15.5 T and 21.1 T and this is shown in Fig. S6(b) along with a fit to R_T . The fit yields a value of $m^* = 0.44(2)m_e$ which is the same, within error, as the value of $m^* = 0.47(6)m_e$ obtained from the FFT analysis. As the field range for the Fourier filter analysis is much smaller than the FFT analysis we can be confident that m^* has been accurately determined. We note that we expect the Dingle damping for the other hole δ orbit to be similar to α and that for the electron orbits (β and γ) to be less, so we can also be confident that these

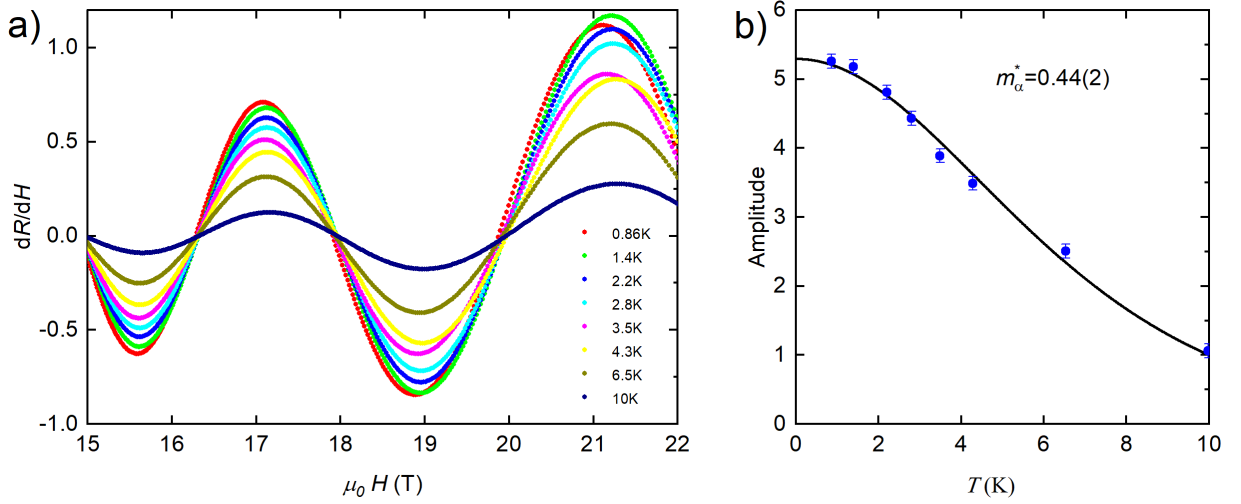


FIG. S6: Mass determination for the α orbit using Fourier filters. (a) shows the Fourier filtered data and (b) the temperature dependent amplitude obtained by fitting the data between 15.5 T and 21.1 T along with a fit to R_T yielding a value of $m^* = 0.44(2)m_e$.

masses have been accurately determined too.

MAGNETIC BREAKDOWN AND DINGLE FIELD

Magnetic breakdown (MB) in WTe_2 occurs between Russian doll nested electron- or hole-pockets which is schematically illustrated in Fig. S7. Different paths and directions, indicated by Arabic numbers 1-7 and arrows are sketched that correspond to hole-hole ($\alpha+\delta$) and electron-electron ($\beta+\gamma$) tunneling. The point or breakdown gap k_g at which MB occurs, is marked with a coloured box. While electron-electron tunneling occurs at a single point with $k_y = 0$ (2,5 in the sketch), hole-hole tunneling may take place at two different spots with finite k_y (2*,5* in the sketch).

The breakdown field, $B_0 = (\hbar/e)(k_g/2)^2$, has been extracted from DFT calculations and found to be $B_0 = 0.2$ T for electrons and 0.5 T for holes, respectively. From the experiment, we observe MB to occur for much higher magnetic fields ($B \geq 8$ T). In order to estimate the Dingle fields and mean-free-paths on the various orbits we have used a filter in Fourier space to isolate the frequency components. We first remove a background (non-oscillatory resistance) from the raw data and compute the fractional change in oscillatory resistance. $(R - R_{bg})/R_{bg}$ (R_{bg} was estimated by fitting a 4th order polynomial to the $R(B)$ data). The Fourier filtered data was then fitted to the standard LK expression $B^{0.5}R_T \cdot \exp(-B_D/B)$, with R_T calculated from the m^* values given in the table in the main paper. The fit is shown in Fig. S8 and the extracted parameter are summarized in Table S2. The results show that the appearance of the peaks in the FFT corre-

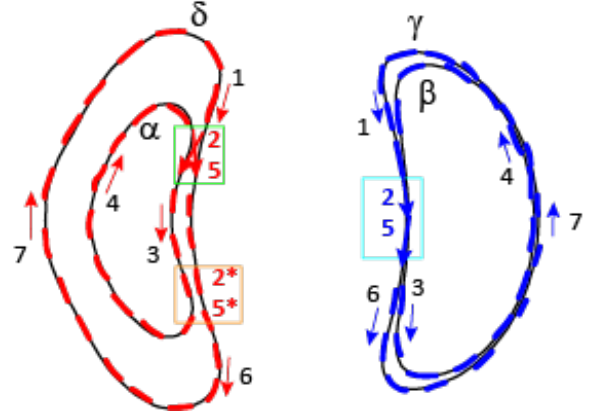


FIG. S7: Magnetic breakdown in WTe_2 between Russian doll nested electron- or hole-pockets. Different paths and directions of motion are indicated.

sponding to MB between Russian doll nested electron- or hole-pockets is determined by impurity damping (Dingle term) since B_D is larger than B_0 . Furthermore the long mean-free-paths for all the orbits show the high quality of our sample.

TABLE S2: Scattering parameter for our sample derived from fit in Fig. S8

band	Type	f (Exp)	m^* (Exp)	B_D (T)	ℓ (Å)
1	h	88 T	0.47 ± 0.06	11 ± 1 T	970
2	h	158 T	0.46 ± 0.04	11 ± 1 T	1200
3	e	148 T	0.40 ± 0.03	11 ± 1 T	1260
4	e	133 T	0.40 ± 0.02	18 ± 2 T	800

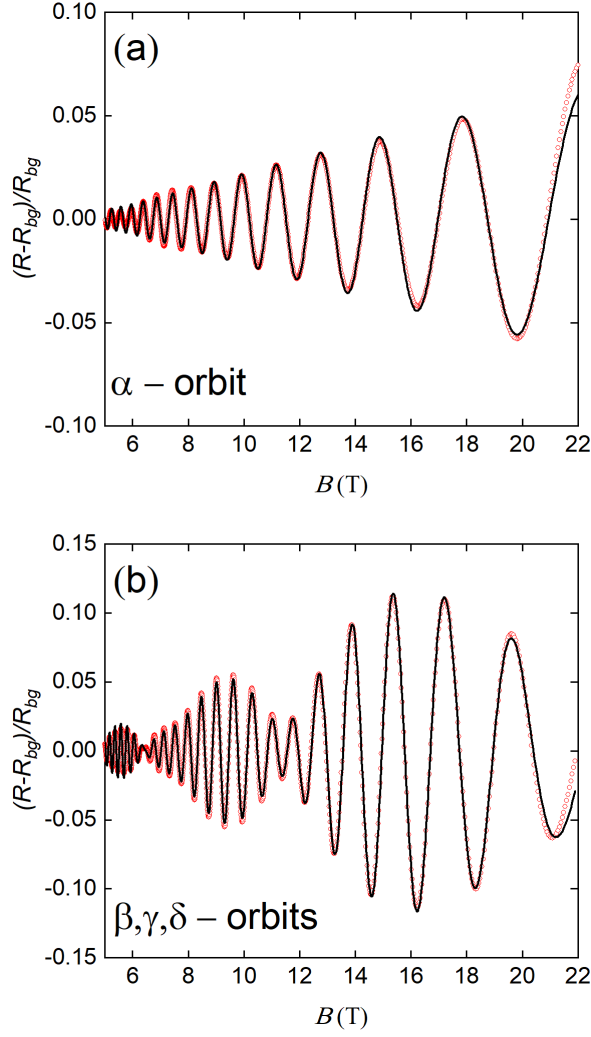


FIG. S8: LK fit to the oscillatory fractional resistance. (a) shows the filtered data (symbols) and fit (solid line) for the α orbit and (b) for β, γ and δ . The data have been filtered to isolate the component by applying a filter in Fourier space. The fit is restricted to the middle of the measured field range to avoid end effect from the Fourier filter. The temperature is $T = 0.84$ K and the masses used for R_T are as given in the main paper.

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