#### Supplementary information for "High-field immiscibility of electrons belonging to adjacent twinned bismuth crystals"

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#### SUPPLEMENTARY NOTES

# Supplementary Note 1. Magneto-optical measurement on another sample

We measured additional magneto-optical results on another bismuth sample with same configuration as descried in the main text in the Supplementary Figure 1. The results are similar to those of the sample shown in the main text.

# Supplementary Note 2. The experimental and theoretical Landau spectrum in linear Y-scale

The Supplementary Figure 2 shows the experimental and theoretical Landau spectrum is in linear Y-scale for magnetic field. The data are same as the Fig.2 of the main text. Four twinned domains are possible. However, our data can be fit by assuming the presence of a single minority domain in addition to the main crystal. It is possible that there are additional minority domains, but their population is below our detection level. It is also possible that during crystal growth pressure inhomogeneity favors the emergence of a minority domain along a Sample #2 Polarizer/Analyzer  $\sim$ 75 deg Magnetic field  $\rightarrow$  normal to the plane



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Supplementary Figure 1. No detectable change of twins under pulsed magnetic field in another sample named #2 with magneto-optical imaging:

specific orientation instead of three.

#### Supplementary Note 3. The experimental and theoretical Landau spectrum including low-field electron spectrum

The Supplementary Figure 3 shows the experimental and theoretical Landau spectrum including low-field elec-



Supplementary Figure 2. The experimental and theoretical Landau spectrum: (a) The illustration of the twinned structure in bismuth. The two crystals tilt from each other by  $108^{\circ}$  and share one common binary. (b) The Landau spectrum of the main and twinned samples from holes according to theory (solid lines) and experiment (symbols) for binary-trigonal, (c) bisectrix-trigonal, and (d) binary-bisectrix planes. The data are the same as the main text, but in linear scale for Y-axis. The red and blue symbols are for the peaks from the holes of the main and twinned crystals from the Fig. 3 of the main text, respectively. The red and blue lines represent the calculated Landau levels from holes of the main and twinned domains, respectively. The magenta dashed lines are for calculated  $0_{e^-}$  electron Landau level which corresponds to the large drop in magnetoresistance as the field is along trigonal. The other electron peaks below 28 T are omitted for clarity, see the ref [1].

tron spectrum. The complex of the Landau spectrum is further complicated by the electron spectrum. In high fields, the electron spectrum is hard to be distinguished from others. And a misalignment could partially explain the discrepancy seen in panels b and c. We also note that experiment cannot distinguish the origin (electron or hole, main or minority crystal) of an observed anomaly. Finally, since these anomalies are peaks in second derivative of magnetoresistance, some of them may be noise rather than signal. The agreement at high field is not as good as low field, which may be related to the higher sensitivity of the Landau spectrum to fine tuning at high field. From the comparison of theory and experiment for common and independent chemical potentials in the Fig.6 in the main text, it is safe to conclude that theory gives a qualitatively better account of the data in

one case compared to the other.

### Supplementary Note 4. Different $V'_3$ for theoretical Landau spectrum

The Supplementary Figure 4 shows the Landau spectrum for main crystal in bis.-tri. plane with different  $V'_3$ . Setting  $V'_3$  to -0.0625, we could achieve a better fit to the experimental values around 40 Tesla. This parameter affects the lowest Landau level only and has no impact on other levels. Consequently, as shown in the figure, the spectra at low magnetic fields remain unchanged from the original parameters, and only a small part of spectra at the quantum limit is modified. Since the value of the g-factor is estimated from spectra below 30 Tesla, there is no change in the g-factor value with the adjustments made in this study.

[1] Z. Zhu *et al.*, Landau spectrum and twin boundaries of bismuth in the extreme quantum limit, Proc. Natl. Acad.

Sci. 109, 14813 (2012).



Supplementary Figure 3. The experimental and theoretical Landau spectrum: (a) The illustration of the twinned structure in bismuth. The two crystals tilt from each other by  $108^{\circ}$  and share one common binary. (b) The Landau spectrum of the main and twinned samples from holes according to theory (solid lines) and experiment (symbols) for binary-trigonal, (c) bisectrix-trigonal, and (d) binary-bisectrix planes. The data are the same as the main text, but in linear scale for Y-axis. The red and blue symbols are for the peaks from the holes of the main and twinned crystals from the Fig. 3 of the main text, respectively. The red and blue lines represent the calculated Landau levels from holes of the main and twinned domains, respectively. The magenta dashed lines are for calculated  $0_{e^-}$  electron Landau level which corresponds to the large drop in magnetoresistance as the field is along trigonal.



Supplementary Figure 4. Landau spectrum for main crystal in bis-tri plane with different  $V_3$ '. Red and black points show the spectrum with  $V_3$ '= -0.0625 and -0.0025 respectively