Extremely large magnetoresistance in a high-quality WTe$_2$ grown by flux method

K. Tsumura$^1$, R. Yano$^1$, H. Kashiwaya$^1$, M. Koyanagi$^1$, S. Masubuchi$^2$, T. Machida$^2$, H. Namiki$^{3,4}$, T. Sasagawa$^3$, and S. Kashiwaya$^1$

$^1$National Institute of Advanced Industrial Science and Technology, 1-1-1 Umezono, Tsukuba, Ibaraki 305-8568, Japan
$^2$Institute of Industrial Science, The University of Tokyo, 4-6-1 Komaba, Meguro-ku, Tokyo 153-8505, Japan
$^3$Laboratory for Materials and Structures, Tokyo Institute of Technology, 4259 Nagatsuta, Midori-ku, Yokohama 226-8503, Japan

E-mail: kohei.tsumura@aist.go.jp

Abstract. We have grown single crystals of WTe$_2$ by a self-flux method and evaluated the quality of the crystals. A Hall bar-type device was fabricated from an as-exfoliated film on a Si substrate and longitudinal resistance $R_{xx}$ was measured. $R_{xx}$ increased with an applied perpendicular magnetic field without saturation and an extremely large magnetoresistance as high as 376,059 % was observed at 8.27 T and 1.7 K.

1. Introduction

Tungsten ditelluride WTe$_2$ is a layered transition metal dichalcogenide and has been the center of attention from various aspects. WTe$_2$ consists of tungsten chains placed between two tellurium sheets. Each layer is combined together by a weak van der Waals interaction between Te-Te, and neighboring layers are rotated by 180$^\circ$ due to the buckling of layers originating from a strong intermetallic bonding [1]. Monolayer is expected to be a quantum spin Hall insulator and much experimental effort has been made to reveal the topological nature [2, 3]. Bulk is considered as a type-II Weyl semimetal where the Lorentz invariance of Weyl fermion is broken [4]. One of the properties of Weyl semimetal, the chiral anomaly, has been observed as a negative magnetoresistance (MR) in a thin film [5]. On the other hand, an extremely large magnetoresistance (XMR) without saturation has been reported for a thick WTe$_2$ film [6]. A perfect electron-hole compensation is thought to be responsible for the XMR [7], however, there is a controversial argument which suggests alternative explanations [8].

Here we report on the magnetoresistance MR and the quality of WTe$_2$ grown by a flux method. We have measured a longitudinal resistance $R_{xx}$ as functions of temperature $T$ and a magnetic field $B$. Residual resistivity ratio ($RRR$) was 704 and the XMR up to 376,059 % was observed at $B = 8.27$ T and $T = 1.7$ K. Moreover, quantum oscillations on the $R_{xx}$ became visible from the magnetic field as small as 2.5 T. These results confirm the high quality of

$^4$ Present address: Tokyo Metropolitan Industrial Technology Research Institute, 2-4-10 Aomi, Koto-ku, Tokyo 135-0064, Japan
2. Experimental

Single crystals were grown by a self-flux method reported by several groups [8, 9, 10]. The mixture of polycrystalline WTe$_2$ (0.1 g) and Te (5 g) were loaded onto a silica tube and heated up to 835 °C. Then, the tube was cooled down to 535 °C at the rate of 1 °C/h. Excess Te was removed by centrifuging and the following annealing. As a result, sizable platelet crystals with a typical size of 1 × 3 mm$^2$ shown in Fig. 1(a) was obtained.

WTe$_2$ film was prepared by mechanical exfoliation on a Si substrate covered with a thermally oxidized SiO$_2$ layer. Au wires were directly connected to one of the exfoliated WTe$_2$ flakes using silver paste immediately after the exfoliation. Figure 1(b) shows the optical microscope image of the fabricated Hall bar-type device. The thickness of the flake is estimated to be around 100 nm judging from the brightness and the color of the flake though it has yet to be confirmed.

Transport measurements were carried out in a variable-temperature cryostat using the wirings shown in Fig. 1(b). $R_{xx} = R_{d4}$ was measured by a standard lock-in technique as functions of temperature and applied magnetic fields. The magnetic field was applied both in-plane and perpendicular to the $ab$-plane up to $B = 8.27$ T.

3. Results and Discussion

Figure 2 shows the temperature dependence of $R_{xx}$ in a perpendicular magnetic field at $B = 0$ and 8.27 T. $R_{xx}$ decreases with temperature drop at $B = 0$ T. $RRR$ define as $R_{300K}/R_{2K}$ is 704. $RRR$ is the indicator of crystal quality, such as the amount of impurities and crystallographic defects. The obtained $RRR$ is not the best but fairly large compared with previous reports, which assures the high-quality of our WTe$_2$ grown by the flux method [6, 9, 11, 12]. When the field at $B = 8.27$ T is applied, the $R_{xx}$ deviates from the zero-field curve and starts to increase at $T^* = 50.5$ K, which is called the turn-on temperature [6]. We will mention this behavior later.

MR versus $B$ under an in-plane and a perpendicular magnetic fields at $T = 1.7$ K are shown in Fig. 3. Here, the MR is defined by the following equation; $MR = |R_{xx}(B) - R_{xx}(0 \, T)|/R_{xx}(0 \, T)$. The MR in the in-plane field is no more than 6,600 %, on the other hand, the MR increases without saturation in the perpendicular field and the XMR of 376,059 % is observed at $B = 8.27$ T. It has been shown that WTe$_2$ is a semimetal, so that both the conduction band and the
Figure 2. Temperature dependence of $R_{xx}$ in a perpendicular magnetic field at $B = 0$ and 8.27 T. The temperature $T^*$ where $R_{xx}$ becomes minimum is indicated by arrow.

Figure 3. Magnetoresistance MR versus applied magnetic field $B$ at $T = 1.7$ K.

Valence band cross the Fermi energy at the same time [6]. Moreover, it has been also identified that the electron density $n_e$ is perfectly compensated with the hole density $n_h$ in WTe$_2$. In the carrier compensated system, the longitudinal resistivity $\rho_{xx}$ can be described by the two-carrier model [13]:

$$\rho_{xx} = \frac{1}{e} \frac{(n_h \mu_h + n_e \mu_e) + (n_e \mu_e \mu_h^2 + n_h \mu_h \mu_e^2)B^2}{(n_h \mu_h + n_e \mu_e)^2 + \mu_h \mu_e B^2(n_h - n_e)^2}.$$

where $e$ is the elementary charge, and $\mu_e$ ($\mu_h$) is the mobility of electrons (holes). As observed in our experiment, the MR increases without saturation in the perfect $n$-$p$ compensated system.
because $B^2$ term in the denominator of eq. (1) vanishes. When the temperature is increased, the balance between $n_e$ and $n_h$ is lost. As a result, the XMR effect is turned off and it is observed as the turn-on behavior of $R_{xx}$ [7]. However it has been also pointed out that the non-saturating XMR cannot be explained solely by the perfect carrier compensation because the compensation is no longer maintained in high magnetic fields [8, 14]. This issue has not been settled yet. Note that although the thickness of the sample was not uniform as seen in Fig. 1(b), the observed XMR and $RRR$ values are consistent with a previous study, where the XMR is proportional to the square of the $RRR$. [10]. This implies that the effect of inhomogeneous thickness is not significant in our experiment.

Finally, we examine quantum oscillations on $R_{xx}$ in the perpendicular field. Figure 4(a) shows $\Delta R_{xx}$ as a function of $B^{-1}$. Here, the $\Delta R_{xx}$ is extracted by subtracting a smoothly increasing component obtained by fit to eq. (1) from $R_{xx}$. The oscillations become visible from $B \sim 2.5$ T, which also implies the high mobility in our crystal. We have performed fast Fourier transformation (FFT) analysis on $\Delta R_{xx}$ and obtained an FFT spectrum shown in Fig. 4(b). Four pronounced peaks at $F_{\alpha 1} = 84$ T, $F_{\beta 1} = 120$ T, $F_{\beta 2} = 145$ T, and $F_{\alpha 2} = 160$ T are observed. On the Fermi surface of the semimetallic WTe$_2$, there are two electron and two hole pockets due to spin-orbit coupling, which has been revealed by band calculation [4]. $F_{\alpha}$ and $F_{\beta}$ in the spectrum are attributed to those hole and electron pockets, respectively. This result is consistent with previous reports [9, 14].

4. Conclusions
We have grown single crystals of WTe$_2$ by the self-flux method and fabricated a hall bar-type sample. The XMR as high as 376,059 % was observed at $B = 8.27$ T and the $RRR$ was 704. These values are fairy large, which guarantees the quality of our crystals. Such high-quality enabled the manifestation of quantum oscillations from low magnetic fields and revealed four Fermi pockets corresponding to the spin-orbit split electron and hole pockets.

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