Large magnetothermopower and Fermi surface reconstruction in Sb$_2$Te$_2$Se

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We report the magnetoresistance, magnetothermopower and quantum oscillation study of Sb$_2$Te$_2$Se single crystal. The in-plane transverse magnetoresistance exhibits a crossover at a critical field $B^*$ from semiclassical weak-field $B^2$ dependence to the high-field unsaturated linear magnetoresistance which persists up to the room temperature. The low-temperature Seebeck coefficient is negative in zero field contrary to the positive Hall effect. The magnetic field induced the sign reversion of the Seebeck coefficient between 2 K and 150 K. The quantum oscillation of crystals reveals the quasi-two-dimensional (quasi-2D) Fermi surface. These effects are possibly attributed to the large Fermi surface which touches Brillouin zone boundary to becomes quasi-2D and the variation in the chemical potential induced by the magnetic field.

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I. INTRODUCTION

The VA-VIA compounds (such as Bi$_2$Te$_3$, Sb$_2$Te$_3$) are well known and have been extensively studied long time ago for their excellent thermoelectric properties.$^{12}$ Recently a new property of these materials, topological insulator (TI), became one of the mostly focused subject of the condensed matter physics and material science.$^{3,4}$ In the three dimensional (3D) TIs, the existence of the nontrivial topological invariance dictates that the excitation gap must vanish at the boundaries, thereby inducing the robust metallic surface states in contrast to the full insulating gap in the bulk. The spin-momentum-locked gapless surface states consisting of spin helical Dirac fermions with a quantum Berry phase could result in a variety of interesting quantum phenomena, such as the quantum anomalous Hall effect and topological superconductor, because the back-scattering and localizaton is suppressed.$^{22}$ A number of materials (including HgTe quantum well, Bi$_{1-x}$Sb$_x$, Bi$_2$Se$_3$, Bi$_2$Te$_3$, and Sb$_2$Te$_3$) have been identified to be 3D TIs, both through theoretical calculations and angular resolved photoemission spectroscopy (ARPES).$^{8,12}$ Among them, the tetradymite-like compounds such as Bi$_2$Te$_2$Se, Bi$_2$Te$_2$S, Bi$_2$Se$_2$S and Sb$_2$Te$_2$Se were predicted to host an isolated Dirac cone on their naturally cleaved surface and attracted intensive attention.$^{12-16}$

Besides the intensive study of the surface state in TIs by ARPES, scanning tunneling microscopy (STM) and theoretical calculation, the magnetotransport behavior of bulk TIs is also important since it is directly related to the practical application. The gapless surface Dirac states will induce some quantum transport behavior in TIs that results in very large linear magnetoresistance (MR)$^{6,7,17}$ and quantum oscillation with Landau sublevels.$^{18}$ In addition, in the highly doped TIs which have bulk carriers and exhibit metallic behavior, the competition between the contribution of the bulk carriers and the surface Dirac carriers could induce some interesting transport properties, such as the quantum Hall effect and layered transport of bulk carriers in doped Bi$_2$Te$_3$, and the field-induced polarized transport of valleys in p-type Sb$_2$Te$_3$. Especially in Bi$_2$Se$_3$ crystals with bulk carriers, thermolectric/thermamagnetic studies reveal the large Zeeman splitting of the three-dimensional bulk band and the variation of the chemical potential above the quantum limit. These demonstrate the complexity of the surface/bulk states and the rich magnetotransport in TIs with different bulk carrier densities. Besides that, although the thermoelectric properties of VA-VIA compounds have been intensively studied, there are few reports about the magnetothermpower behavior. The magnetic field influence on the thermal transport in ordinary metals is usually very small. Initially the large magnetothermopower effect was observed in doped InSb which was attributed to the effects of the sample geometry on the minority carriers.$^{23}$ In a system with large magnetoresistant effect the magnetic field has significant influence on the properties of carriers and large magnetothermopower effect could be expected. The giant magnetothermopower effect was achieved in the giant magnetoresistant multilayer/granular systems and the colossal magnetoresistant manganites, which could be of interest for magnetic field sensors or magnetic controllable thermolectric devices.$^{23,24}$

In this paper, we report the magnetoresistance, magnetothermopower and quantum oscillation study of Sb$_2$Te$_2$Se single crystal. The in-plane transverse magnetoresistance exhibits a crossover at a critical field $B^*$ from semiclassical weak-field $B^2$ dependence to the high-field unsaturated linear magnetoresistance which persists up to the room temperature. The low-temperature Seebeck coefficient is negative in zero field contrary to the positive Hall resistivity, indicating the multiband effect. The magnetic field induced the sign reversion of the Seebeck coefficient between 2 K and 150 K. The quantum oscillation of crystals reveals the quasi-2D Fermi surface. These effects are possibly attributed to the large Fermi
measurements, the current path was in the four-wire method. In the in-plane resistivity and Hall measurements, the current path was in the bc-plane, whereas magnetic field was parallel to the a-axis except in the angular dependent MR measurement. Seebeck coefficient was measured using steady state method and one-heater-two-thermometer setup with silver paint contact directly on the sample surface. The heat and electrical current were transported within the bc-plane of the crystal, with magnetic field along the a-axis and perpendicular to the heat/electrical current. The relative error in our measurement for both κ and S was below 5% based on Ni standard measured under identical conditions. The de Haas-van Alphen (dHvA) oscillation experiments were performed at National High Magnetic Field Laboratory, Tallahassee. The crystals were mounted onto miniatur Seiko piezoresistive cantilevers which were installed on a rotating platform. The field direction can be changed continuously between parallel (θ = 0°) and perpendicular (θ = 90°) to the c-axis of the crystal.

II. EXPERIMENTAL

Single crystals of Sb₂Te₂Se were grown using a high-temperature modified Bridgman method. X-ray diffraction (XRD) data were taken with Cu Kα (λ = 0.15418 nm) radiation of Rigaku Miniflex powder diffractometer. Electrical transport measurements up to 9 T were conducted in Quantum Design PPMS-9 with conventional four-wire method. In the in-plane resistivity and Hall measurements, the current path was in the bc-plane, whereas magnetic field was parallel to the a-axis except in the angular dependent MR measurement. Seebeck coefficient was measured using steady state method and one-heater-two-thermometer setup with silver paint contact directly on the sample surface. The heat and electrical current were transported within the bc-plane of the crystal, with magnetic field along the a-axis and perpendicular to the heat/electrical current. The relative error in our measurement for both κ and S was below 5% based on Ni standard measured under identical conditions. The de Haas-van Alphen (dHvA) oscillation experiments were performed at National High Magnetic Field Laboratory, Tallahassee. The crystals were mounted onto miniatur Seiko piezoresistive cantilevers which were installed on a rotating platform. The field direction can be changed continuously between parallel (θ = 0°) and perpendicular (θ = 90°) to the c-axis of the crystal.

III. RESULTS AND DISCUSSIONS

Sb₂Te₂Se has tetradymite structure consisting of three quintuple layers and can be represented as -Te-Sb-Se-Sb-Te-Se-Sb-Se-Sb-Te- (as shown in the inset of Fig. 1(b)). Powder XRD pattern of our crystals which were fitted by RIETICA software in Fig. 1(a) can be indexed in the R3m space group. The crystals are plate-like and the base plane is the ab-plane (Fig. 1(b)). SEM elementary analysis revealed that the composition of our crystals is Sb₂.02(7)Te₁.95(3)Se₁.05(7). The in-plane resistivity of the crystal is metallic above ∼ 20 K (Fig. 2(a)) and then increases with decreasing temperature below 20 K (as shown in the inset of Fig. 2(a)). The external magnetic field enhances the resistivity in the whole temperature range significantly, while it has barely any influence on the thermal conductivity (Fig. 2(b)). The thermopower behavior of Sb₂Te₂Se is interesting (Fig. 2(c)). The Seebeck coefficient is positive above ∼ 100 K. But it decreases with decreasing temperature and becomes negative below 100 K. The magnetic field induces the decrease of the sign reversal temperature of Seebeck coefficient.

Below we will discuss the magnetic field effects on the resistivity and Seebeck coefficient. Fig. 3(a) shows the magnetic field dependence of the MR at several temperature. At 2 K, the MR reaches ∼ 30% in 9 T field (Fig. 3(a)). In Fig. 3(b), the field derivative of MR, dMR/dB, initially decreases with increase in field indicating B₁/² dependence of MR, and then linearly increases with field in the low field region which indicates a B² dependent MR by linear fitting (lines in the low field region). But above a characteristic field B°, dMR/dB saturates to a very much reduced slope This indicates that in the high fields the MR is dominated by a linear field dependence plus a very small quadratic term (MR = A₁B + O(B²)) as shown by lines in the high-field region, which extends to a very low crossover fields B° where the MR naturally reduces to a weak-field semiclassical quadratic response. The high field linear MR persists even at 300 K. Similar room temperature linear MR was observed in Bi₂Te₃ nanosheet.

The linear magnetic field response was also found in the Seebeck coefficient (Fig. 4(a) and (b)). As shown in Fig. 2(c) and Fig. 4(a), the magnetic field enhances the Seebeck coefficient above 150 K but the Seebeck coefficient is positive in whole field range. Below ∼ 120 K, the Seebeck coefficient in zero field is negative in low fields.
where the absolute value of $S$ decreases linearly with increase in magnetic field. Above specific crossover field there is a sign change of $S$ from negative to positive where the absolute value of $S$ increases with increase in field. At 120 K, the crossover field is around 3 T. With decreasing temperature, the crossover field initially increases to about 8 T at 40 K and then decreases to around 2 T at 10 K.

Fig. 4(c) shows the Hall resistivity and the apparent carrier density as function of temperature and field. The Hall resistivity exhibits linear field dependence, the signature of single-band behavior. But contrary to the sign change of Seebeck coefficient with changing temperature and field, the Hall resistivity for $\text{Sb}_2\text{Te}_2\text{Se}$ is always linear and positive in whole temperature and field range (Fig. 4(c)). The apparent carrier density $n_{\text{app}} = B/\rho_{xy}$ increases with decreasing temperature which is consistent with the metallic behavior (the inset in Fig. 4(c)).

The tetradymite-like compounds such as $\text{Bi}_2\text{Te}_2\text{Se}$, $\text{Bi}_2\text{Te}_2\text{S}$, $\text{Bi}_2\text{Se}_2\text{S}$ and $\text{Sb}_2\text{Te}_2\text{Se}$ were predicted to host an isolated Dirac cone on their naturally cleaved surface and attracted intensive attention. In order to clarify the possible topological surface state and the electronic structure, we performed the quantum oscillation measurements up to 35 T field on $\text{Sb}_2\text{Te}_2\text{Se}$ crystals. Both the magnetoresistance and the magnetic torque shows quantum oscillation. The temperature-dependent and angular ($\theta$) dependent cantilever or magnetoresistance signals are shown in Fig. 5 and Fig. 6, respectively. The Fourier transform spectrum of the oscillation at 0.4 K reveals a periodic behavior in $1/B$ with a frequency $F \sim 225$ T when field is perpendicular to the $ab$-plane ($\theta=0$), as shown in Fig. 6(b). The temperature dependence of the oscillation amplitude can be used to determine cyclotron effective mass through the Lifshitz-Kosevitch formula. Using the highest oscillation peak, the fitting gives a $m \approx 0.13(5)m_e$ where $m_e$
is the bare electron mass (the inset of Fig. 5(a)). Fig. 6(a) shows the magnetic field direction dependence of the typical dHvA oscillations of Sb$_2$Te$_2$Se single crystal, and the detailed angular dependence of the oscillation frequency are shown by squares in Fig. 6(b). For a surface state or two-dimensional Fermi surface, the cross section has $S_F(\theta) = S_0 |\cos(\theta)|$ angular dependence and the oscillation frequencies should be inversely proportional to $|\cos(\theta)|$. In Fig. 6(b), the angular dependence of the oscillation frequency can be fitted very well by $1/|\cos(\theta)|$ (the solid line). This indicates that the observed states in the quantum oscillation is quasi-2D Fermi surface.

However, the low-temperature resistivity of our material is $\sim 5$ m$\Omega$ cm and the density of carrier from the Hall resistivity is about $2 \times 10^{19}$ cm$^{-3}$, and both values are close to that observed in doped TIs such as Bi$_2$Se$_3$ and Bi$_2$Te$_3$. This means the bulk states should dominate the transport behavior in this material and the quasi-two-dimensional Fermi surface observed in quantum oscillation should not come from the Dirac surface states. In highly doped Bi$_2$Se$_3$ with a carrier density $\sim 4.7 \times 10^{19}$ cm$^{-3}$, it was reported that the Fermi surface is large enough to touch the Brillouin zone boundary. So the quasi-2D magnetotransport features in this system arise from the bulk of the sample acting as many parallel 2D electron systems to give a multilayered quantum Hall effect, instead of the Surface state. Our material has similar carrier density and then the quasi-2D magnetotransport behavior observed in quantum oscillation (Fig. 6) should have similar origin. Besides that, the band structure calculation reveals that the bulk state in Se-doped Sb$_2$Te$_2$ most likely is massive Dirac state, which possibly induce the linear magnetoresistance in high field as observed in other materials with Dirac states.

In a nonmagnetic metal, the diffusion of carriers and the phonon drag effect will contribute to the Seebeck coefficient. The diffusion mechanism of electron and holes will determine the sign of the Seebeck coefficient. The phonon drag mechanism often gives a peak structure at temperature $T \sim \Theta_D/5$ where $\Theta_D$ is the Debye
The Seebeck coefficient is given by the Mott relationship, with diffusion mechanism and electron-type carriers, Seebeck coefficient on field $B$ appears only in the conductivity matrix element $\sigma_{ij}(B)$ ($\sigma = \sigma_{xx}$). The electron contribution to Seebeck coefficient $S_e$ is usually negative while the hole contribution $S_h$ is always positive.\textsuperscript{40,41} For a two-band metal comprising electron and hole bands, $S$ is expressed as

$$S = \frac{\sigma_h |S_h| - \sigma_e |S_e|}{\sigma_h + \sigma_e},$$

where $\sigma_{e(h)}$ and $S_{e(h)}$ are the contributions of electrons (holes) to the electric conductivity and Seebeck coefficient, respectively.\textsuperscript{40} So the different sign of Seebeck coefficient and Hall resistivity implies the multiband effect in Sb$_2$Te$_2$Se, while the linear positive Hall resistivity implies that the single hole band dominates the Hall transport.

Similar sign change in the Seebeck coefficient by decreasing temperature was also observed in Se-doped Bi$_2$Se$_3$ and one possible reason for this and the different sign between the Hall and Seebeck coefficient is the negative phonon drag peak superposed by the positive diffusive thermoelectric response.\textsuperscript{40,41} This could explain the positive-negative transition of Seebeck coefficient by decreasing the temperature in our material. However, it could not induce the sign change by increasing magnetic field since the magnetic field should have no influence on the phonon drag, and this means the phenomenon should come from the carriers. Similar phenomena where Hall resistivity and Seebeck coefficient has different sign were also observed in some noble metals such as Cu and Ag and was attributed to the neck structure of the Fermi surface.\textsuperscript{44} The Fermi surface in Ag and Cu is far from spherical, but Fermi surface just touches the Brillouin boundary and has a set of "necks" at the Brillouin boundary.\textsuperscript{45} Our material has high carrier density and the quasi-2D magnetotransport behavior in quantum oscillation suggests that the Fermi surface (hole) in doped Sb$_2$Te$_2$Se is large enough to touch the Brillouin zone boundary and should give similar neck structure as Cu/Ag. Usually these necks are almost perpendicular to the Brillouin boundary and the curvatures near the necks are oppositely to the residual Fermi surface, as shown in Cu/Ag case. The neck structure gives a heavy electron-like (negative) contribution to the Seebeck coefficient. The density of this electron-like states associated with the necks, is extremely large due to the rapidly varying surface with energy. But its contribution to the Hall coefficient is very small because it has the very large effective mass and the residual Fermi surface is massive Dirac state with very large mobility. Consequently the different sign between the Hall and Seebeck coefficient appears.

The sign reversion of Seebeck coefficient in magnetic field should be related to the different response of two kind of contribution from different parts of the Fermi surface. Similar effects were also reported in several other materials such as high-$T_c$ cuprates, and was considered as a signal of the Fermi surface reconstruction.\textsuperscript{46,47}
For example, the high-field $R_H$ and $S/T$ are found to drop with decreasing temperature and become negative in HgBa$_2$CuO$_{4+\delta}$, which is attributed to the charge-order wave and related Fermi surface reconstruction.\textsuperscript{22} In similar materials Bi$_2$Se$_3$ with high bulk carrier density, a strong variation of the chemical potential and Fermi energy above the quantum limit is observed and is believed to induce the observed increase in the Nernst response.\textsuperscript{9} The change of the chemical potential could also happen in Sb$_2$Te$_2$Se and it could suppress the contribution of the states at the Necks of the Fermi surface to the Seebeck coefficient by suppressing the number of that states. Besides that, with increasing magnetic field, the Dirac holes will be very easy to form Landau splitting and then occupy the zeroth LLs (achieve quantum limit) gradually because of the high mobility and small effective mass, while the moderate field hardly has any influence on the states at the necks due to the very large mass. So under magnetic field the Dirac holes will dominate the thermal transport behavior in the quantum limit since the Fermi level locates between the zeroth and first LLs and the DOS at the Fermi level is suppressed. The positive Hall resistivity and Seebeck coefficient confirm the dominant hole-like carriers up to some crossover field above which there is a sign change in $S(B)$ due to enhanced contribution of bulk hole-like carriers.

**IV. CONCLUSION**

In summary, we report the magnetoresistance, magneto-thermopower and quantum oscillation study of Sb$_2$Te$_2$Se single crystal. The in-plane transverse magnetoresistance exhibits a crossover at a critical field $B^*$ from semiclassical weak-field $B^2$ dependence to the high-field unsaturated linear magnetoresistance which persists up to the room temperature. The low-temperature Seebeck coefficient is negative in zero field contrary to the positive Hall resistivity, indicating the multiband effect. The magnetic field induced the sign reversion of the Seebeck coefficient between 2 K and 150 K. The quantum oscillation of crystals reveals the quasi-2D Fermi surface. These effects are possibly attributed to the large Fermi surface which touches Brillouin zone boundary to become quasi-2D and the variation in the chemical potential induced by the magnetic field.

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