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Giant and anisotropic magnetoresistances in p-type Bi-doped Sb 2Te 3 bulk single crystals

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Antimony telluride (Sb$_2$Te$_3$) compounds have rhombohedral structures, space group $R_{3m}$ ($D_{3d}^3$) and are well-known as narrow gap layered semiconductors [1]. In the past few years, Sb$_2$Te$_3$ crystals have been intensively studied because of their excellent thermoelectric performances in the vicinity of room temperatures [2, 3]. Doping effects on thermoelectric properties have been studied to maximize the thermoelectric figure of merit, $ZT$, which is of critical importance for any potential commercial usage of thermoelectric energy conversion. Sb$_2$Te$_3$ based semiconductors have been widely applied in thermoelectric energy converters, refrigerators, and thermostats operating near room temperatures. V. A. Kulbachinskii and H. Y. Lv, et. al. have investigated the electrical properties and band structure parameters of Se and Bi doped Sb$_2$Te$_3$ from the Hall effects, Shubnikov-de Haas effects (SdH), and thermoelectric effects [4, 5, 6, 7]. Many-valley model was applied to estimate several parameters including the shape and orientation of the energy ellipsoids as well as the density of carriers and the relaxation times. The anisotropy of the relaxation times and effective masses of electrons in Fermi surface was also determined by the comparison of the observed galvanomagnetic coefficients with those predicted from De Haas–van Alphen effects (dHvA) experiments.

Recently, Sb$_2$Te$_3$ compounds were confirmed as three dimensional topological insulators with robust surface states comprising a single Dirac cone by angle resolved photoemission spectroscopy (ARPES) measurements and band structure calculations [8, 9]. Topological insulators are quantum materials with an insulating bulk state and a topologically protected metallic surface state with spin and momentum helical locking and a Dirac-like band structure [10, 11]. Unique and fascinating electronic properties, such as the quantum spin Hall effects, magnetoelectric effects, magnetic monopoles, and elusive Majorana states, are expected from topological insulators [12, 13]. Topological insulators have great potential applications in spintronics and quantum information processing, as well as magnetoelectric devices with higher energy efficiency [14, 15]. Up to now, the surface states in topological insulators have been mainly investigated by angle-resolved photoemission spectroscopy (ARPES), scanning tunneling microscopy (STM) and theoretical calculations [16]. To understand the thermoelectric and topological insulator properties and to investigate the possibility of device applications, bulk magnetotransport measurements are indispensable. Shubnikov-de Haas and Aharonov-Bohm oscillations have been observed in Bi$_2$Se$_3$ and Bi$_x$Sb$_{1-x}$ topological insulators [17, 18]. And low temperature linear MR (defined as $\text{MR}=R_0-R/2R_0)*100$) was observed in Bi$_2$Se$_3$ crystal flakes and Bi$_2$Se$_3$ nanoribbons [19, 20]. However, to the best of our knowledge, the magnetotransport study of Sb$_2$Te$_3$ is very limited. Here, we report the angular dependent MR in Bi doped Sb$_2$Te$_3$ bulk single crystals over a broad range of temperatures and magnetic fields. Giant MR of up to 230% was observed in Bi doped Sb$_2$Te$_3$ bulk. The observed MR exhibits quadratic field dependences in low fields and becomes linear at high fields. The giant MR represents anisotropic MR in Bi doped Sb$_2$Te$_3$ bulk single crystals could find applications in anisotropic magnetoelectronic devices.

The Sb$_2$Te$_3$ single crystals with 9 % Bi doping used for this study were cleaved from bulk crystals with 99.99% purity. Four-probe transport measurements were performed on a rectangular sample with dimensions of $4 \times 3 \times 0.16 \text{ mm}^3$ between 2.5 and 200 K using a Quantum Design 14 T Physical Properties Measurement System.
Figure 3(a) shows polar plots of the angular dependence of the MR measured in several magnetic fields at 2.5 K. The magnetic fields, ranging from 4 to 13 T, were kept constant during each rotation. The observed angular dependence of the MR displays strong anisotropy and a twofold symmetry. Wide peaks appear around the perpendicular field configuration (about $\theta = 15^\circ$, $195^\circ$) and dips around the parallel field configuration (about $\theta = 105^\circ$, $285^\circ$). This suggests that the MR is most strongly pronounced in out-of-plane high fields. In contrast, the MR anisotropy ratio (defined as $(MR_{\theta=15^\circ}-MR_{\theta=105^\circ})/MR_{\theta=15^\circ} \times 100\%$) increases with magnetic fields decreasing and reaches a maximum of 210\% in 4 T at 2.5 K, as presented in Fig. 3(b). This suggests that the MR anisotropy ratio is most strongly pronounced in low fields at a fixed temperature. The common feature is that the peaks and dips of the plots are present at the same $\theta$ values. Figure 4(a) displays polar plots of the angular dependence of the MR measured at 13 T for various temperatures. The temperatures, ranging from 200 to 2.5 K, were kept constant during each rotation. The observed angular dependence of the MR displays anisotropic features and a twofold symmetry, which is similar to that shown in Fig. 3. Furthermore, both the MR and the MR anisotropy ratio are strongly pronounced at low temperatures.

The well established Kohler’s rule suggests that the MR of a semiconductor is a universal function of B as a result of the Lorentz force deflection of carriers. At high field, most materials show saturating MR. Therefore, such a giant anisotropic non-saturating MR in our Bi doped Sb$_2$Te$_3$ bulk crystals is unusual. Linear quantum-MR theory was originally developed by Abrikosov based on gapless semiconductors and at the extreme quantum limit to explain the observed giant linear MR in doped silver chalcogenides [22]. However, it is difficult to reach the extreme quantum limit in our Bi doped Sb$_2$Te$_3$ because of high hole density. Metals that contain Fermi surfaces with open orbitals in some crystallographic directions, including Cu, Ag, Au, Mg, Zn, Sn, Pb and Pt, will also exhibit large no-saturated MR for fields applied in those directions [23]. These MR are positive and linear in high magnetic fields. And, they present obvious anisotropy because of anisotropy of Fermi Surface, which are similar what we observed in p-type topological insulator Sb$_2$Te$_3$ [24]. In addition, no-saturated MR can occur in semiconductors as a consequence of strong electrical disorder, which is related to the carrier mobility but independent of carrier density. They are also positive and have transitions from a quadratic field dependence at low fields to a linear dependence at higher fields. And the high-field MR was found to be linear at all temperatures ranges [25].

The giant and high-field linear MR with twofold symmetry of anisotropy in our samples is likely to be related to the configurations of the valence bands. The valence bands of undoped Sb$_2$Te$_3$ are multi-valley bands and consist of upper and lower...
valence bands. Sb$_2$Te$_3$ has a non-spherical Fermi surface consisting of six ellipsoids tilted at an angle to the basal plane, where the two valence bands are responsible for conduction [26]. Both valence bands are filled by holes with different mobilities, effective masses, and concentrations in different valleys. The non-saturating linear MR suggests that holes have open orbits along the Fermi surface, because the MR should saturate at high B if the orbits are closed in high fields [23]. Doping not only increases the density of holes and decreases the mobility of holes, but also increases the additional impurity scattering. The main mechanisms of scattering in Bi doped Sb$_2$Te$_3$ are acoustic phonon scattering, impurity scattering, and intravalley and intervalley scattering. The giant MR might come from intravalley and intervalley scattering in the upper valence band and impurity scattering from doping induced impurity concentrations in different valleys. The non-saturating linear MR observed, especially in high magnetic fields and low temperatures. Giant MR and strong anisotropy of MR were observed, especially in high magnetic fields and low temperatures. The giant MR might result from anisotropic carrier mobility, relaxation time and effective mass in Fermi surface. Based on the observed giant, high field linear MR and strong anisotropy, the Bi doped Sb$_2$Te$_3$ single crystals might have potential applications in anisotropic magneto-electronic devices, such as anisotropic magnetic field sensors, operated at low temperatures and high fields.

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REFERENCES

FIGURE CAPTIONS

FIG. 1 Resistivity of $p$-type Sb$_2$Te$_3$ as a function of temperatures in 0 T and 13 T. The insets show the measurement schematic diagram (left) and Hall resistance as a function of field at 100 K (right).

FIG. 2 MR as a function of magnetic fields for several temperatures from 2.5 K to 200 K. The inset shows a comparison of MR under out-of-plane and in-plane fields of 13 T at 2.5 K.

FIG. 3 Polar plots of (a) angular dependences of MR and (b) anisotropy ratios of MR at 2.5 K in different fields.

FIG. 4 Polar plots of (a) angular dependences of MR and (b) anisotropy ratios of MR in 13 T at different temperatures.