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Heavy doping effects on $PbBi_2Te_4$ topological insulator

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Abstract

Heavy doping effects on $PbBi_2Te_4$ topological insulator (TI) were experimentally investigated, where Sb and Se were chosen as dopants. By Sb doping, we successfully obtained bulk insulating samples for the first time in this system by fine-tuning the composition. As for Se doping, structure analyses by X-ray diffraction indicated that Se inner ordering configuration was realized, which is one of the three possible Se configurations in the $PbBi_2(Te,Se)_4$ crystal structure. A previous theoretical work indicated that this Se configuration should result in the formation of the largest bandgap of all the TIs identified so far.

1. Introduction

Topological insulators (TIs) are known to exhibit characteristic 2D surface states similar to graphene and other Dirac materials. TIs' surface states have a peculiar spin property compared to graphene, and therefore, have been attracting much attention from condensed matter physics and many other fields. However, the detection and practical use of the surface states are generally difficult. The reason is that the bulk resistivity of TIs is considerably low in most of the TIs identified so far, and that the characteristic surface conduction is masked by bulk conduction. Therefore, many efforts have been made to find new TI materials exhibiting bulk insulation or to improve those already identified.

In the present study, we chose $PbBi_2Te_4$ as a target material, and attempted to increase bulk resistivity by heavy doping of other elements such as Sb and Se. A series of $Pb(Bi,Sb)_2(Te,Se)_4$ TIs have three advantages: (1) the highest surface carrier density, (2) the largest energy gap and (3) an unusual 1D dislocation conduction.

First, the surface carrier density is the highest among TIs identified so far[1], and therefore, it is expected that the utilization of surface states is easier than other TI materials. Second, it has been reported that a large band gap over 350meV can be achieved by Se doping[2]. Third, this material can yield unusual conduction along 1D crystalline defects, i.e., dislocations[3]. While conventional TIs such as $(Bi,Sb)_2(Te,Se,S)_3$ have the set of topological numbers of $(\nu_0;\nu_1\nu_2\nu_3)$=(1;000), this material has an unusual set of $(\nu_0;\nu_1\nu_2\nu_3)$=(1;111). A previous theoretical study has shown that dislocations in TIs with this set of topological numbers can form 1D gapless states, which are topologically protected against disorder scattering [3]. Recently, Hamasaki et al. showed excess conductivity attributable to dislocation conduction in a Bi-Sb TI[4].

Existence of topological surface states in a $PbBi_2Te_4$ TI was first verified independently by Souma et al.[1] and Kuroda et al.[5] using ARPES technique. Souma et al. also reported that by controlling Sb doping ratio, Fermi level can be tuned. However, bulk insulating samples have never been obtained in this material.

Recently, heavily Se doped $PbBi_2Te_4$, i.e., $PbBi_2(Te,Se)_4$ has gathered attentions. As described before, there are three possible configurations of Se-site distribution in the $PbBi_2Te_4$ crystal structure: random distribution, outer Se ordering, and inner Se ordering. Shvets et al.[2] calculated band structure for each configuration, and showed that maximum bandgap is achieved when inner Se ordering occurs. In addition, the gap size is expected to be about 350meV, which is the largest of all the TIs identified so far.

In view of these trends, we examined heavy doping effects on $PbBi_2Te_4$ TI. As for Sb doping, we fabricated a series of $Pb(Bi_{1-x}Sb_x)_2Te_4$ samples and systematically studied the transport properties in order to enhance bulk resistivity. As for Se doping, the configuration of Se ordering was determined.

2. Experimental Procedures

Bulk crystals were grown by vertical Bridgman method for the two series of alloys, $Pb(Bi_{1-x}Sb_x)_2Te_4$ ($0.6<x<0.9$) and $PbBi_2(Te_{1-y}Se_y)_4$ ($y=0.4$). First, constituent elements were sealed in a quartz ample under a vacuum of 10^{-3} Torr. Then, Bridgman method was applied with the ample translation speed of 2mm/h.

The solidified rod was studied for phase identification. Crystal structure and composition were determined using powder X-ray diffraction (p-XRD) (RINT2500V, Rigaku) and Electron Probe Micro Analyzer (EPMA, JXA8800).

The crystals were easily cleaved on (111) plane, and single crystal flakes of grown TI phase were taken out, and used for resistivity measurements. Ohmic contacts were fabricated on the plane using room-temperature cured silver paste. Resistivity measurements were conducted using Physical Property Measurement System (PPMS, Quantum Design inc.) in the temperature range of 2-300 K, and in the magnetic field range from -9 to +9 T.

3. Results and Discussion

Sb-doped samples

Fig. 1 shows the temperature dependences of resistivity measured for the samples of $Pb(Bi_{1-x}Sb_x)_2Te_4$ ($0.6<x<0.9$).

The samples with compositions around x=0.79 show insulating behavior while others are metallic. Insulating samples show conventional thermal activation behavior in the temperature range 120-200 K: Arrhenius plots of these data showed the excitation energies of $\Delta \approx 32$meV. This value is almost the same as those reported for $(Bi,Sb)_2(Te,Se)_3$ TIs [6].

In the low temperature region (2K<T<120 K), thermal activation behavior has vanished, and the resistivity appears to be saturated. The magnetoresistance of this region also showed an unusual behavior. The low-temperature data were found to agree well with a 3D weak-antilocalization (WAL) and weak-localization (WL) model combined with electron-electron interaction[7]. This model is originally based on electron scatterings by random potential. Previous studies have shown that $PbBi_2Te_4$ TI has a lot of antisite defects. In addition, our $Pb(Bi,Sb)_2Te_4$ materials should have randomly distributed Sb. These antisite defects and doped Sb atoms may create impurity bands within the gap. The behavior in the low temperature range in our samples can be interpreted as the conduction of carriers subjected to disorder scatterings in the impurity band.

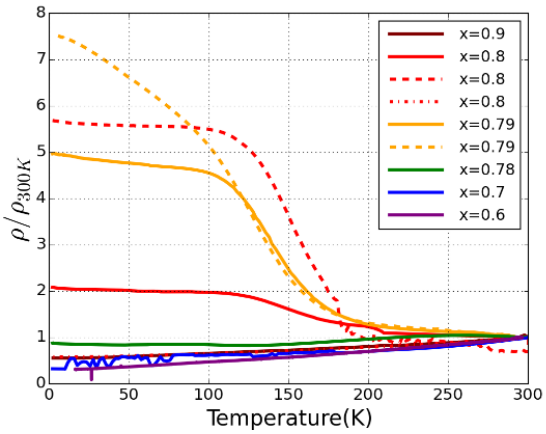

Fig. 1 Temperature dependences of resistivity measured for the samples of $Pb(Bi_{1-x}Sb_x)_2Te_4$ (0.6<x<0.9). The data are normalized to the room temperature value.

Se-doped samples

There are three possible configurations of Se distribution in $PbBi_2(Te,Se)_4$ crystal, as described in introduction. We have simulated the p-XRD patterns for the three configurations using RIETAN-FP (Fig.2 (a)-(c)). Overall features are similar for the three patterns, except for the intensity ratios among the three low-angle peaks indexed as 003, 006, and 009. This is due to the fact that the stacking order of layer structure in the c-direction substantially differs for the three configurations.

Fig. 2 (d) presents the p-XRD pattern measured for $PbBi_2(Te_{1-y}Se_y)_4$ (y=0.4). First, we notice other minor peaks compared with the simulated patterns, indicating the existence of contaminant phase(s). With regard to the intensity ratios among the low-angle peaks, they agree well with those in the pattern of Fig. 2 (c), corresponding to the inner Se ordering. This configuration has theoretically been predicted to yield the largest bandgap (≈350meV) of all the TIs identified so far.

To study the Se ordering in our samples in more detail, Rietveld analyses of p-XRD patterns and direct observations of Se configuration by scanning transmission electron microscopy are in progress. The expected large bandgap for the inner Se ordering should lead to the fabrication of highly insulating TI samples. To realize it, it is necessary to obtain the samples free from contaminant phases.

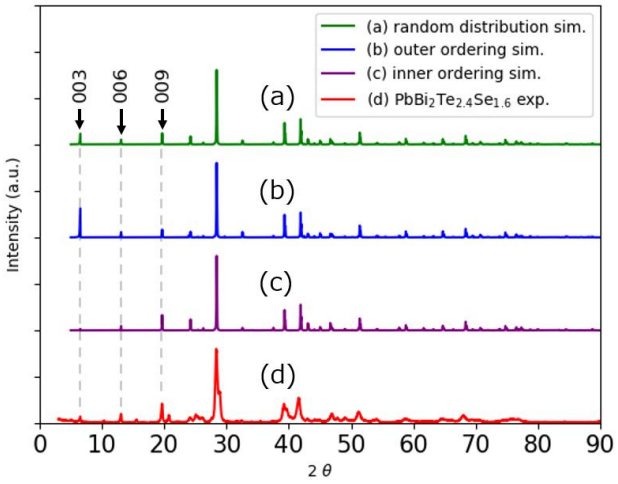

Fig. 2 Simulated p-XRD patterns for three possible Se configurations ((a)-(c)), and the pattern measured for $PbBi_2(Te_{1-y}Se_y)_4$ (y=0.4) (d).

4. Conclusions

Effects of Sb or Se doping on $PbBi_2Te_4$ TI were experimentally investigated. With regard to the Sb doping, we successfully obtained bulk insulating samples in $Pb(Bi_{1-x}Sb_x)_2Te_4$ ($x \approx 0.79$). Temperature dependence of resistivity and magnetoresistance in a low temperature region agreed well with a 3D WAL-WL model. As for Se doping, structure analyses by X-ray diffraction indicated that Se inner ordering configuration was realized, which has previously been indicated to yield the largest bandgap of all the TIs identified so far.

Acknowledgements

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