

Magneto-optical evidence of the topological phase transition in (111)-Pb_{1-x}Sn_xTe

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Abstract. Recently, the study of topological crystalline insulators has become of great interest in condensed matter physics. In this work, we present infrared magneto-optical investigation of such novel quantum states in narrow gap rocksalt IV-VI semiconductor Pb_{1-x}Sn_xTe which exhibits a topological phase transition at a critical value $x_c \approx 0.40$. High-mobility Bi-doped trivial and non-trivial (111)-Pb_{1-x}Sn_xTe ($0 \leq x \leq 0.56$) films grown on BaF₂ substrates by MBE were examined at 4.5K and magnetic fields $B=0-15$ T. Massive and massless Dirac fermion models are used to analyze transmission spectra. We are able to determine the band parameters of the bulk and the topological surface states of such material.

1. Introduction

The search for topological states of matter beyond the existing Z_2 topological insulators (TIs) protected by time-reversal symmetry has recently become one of the major topics in physics of semiconductors. A novel topological state called topological crystalline insulator (TCI) protected by mirror symmetry of the crystal has been discovered in narrow gap rocksalt IV-VI semiconductors such as Pb_{1-x}Sn_xTe and Pb_{1-x}Sn_xSe alloys [1]. Such materials were shown to exhibit semiconducting bulk states accompanied by gapless Dirac surface states. In Pb_{1-x}Sn_xTe a band inversion occurs at four equivalent L-points in the three-dimensional Brillouin zone (3DBZ) when the Sn content is above the critical value $x_c \approx 0.40$, leading to a trivial to non-trivial topological phase transition for $x \geq x_c$ [2]. Since the L-points are mirror symmetric with respect to six equivalent {110} diagonal crystallographic planes, this band inversion results in an even number of surface Dirac cones at points of (001), (110) or (111) surfaces of the crystal where the L-points are projected [3].

In this work, (111)-oriented Pb_{1-x}Sn_xTe films were studied. Pb_{1-x}Sn_xTe crystallizes in a face-centered-cubic structure as NaCl. The small direct band gap of the compound is located at the L-points of the 3DBZ shown in figure 1a. (111)-Pb_{1-x}Sn_xTe possesses four ellipsoidal bulk carrier pockets at the L-points: one bulk longitudinal valley (black) of which the major axis is parallel to the [111] direction and three bulk oblique valleys (red) of which the major axes are tilted by $\pm 70.5^\circ$ with respect to the [111] direction. A band inversion occurs at four equivalent L-points when going from PbTe to SnTe as



illustrated in figure 1b. In the topological trivial phase, the lowest conduction and the highest valence bands are labelled respectively by L_6^- and L_6^+ . The energy gap ($E_g > 0$) initially decreases with increasing Sn content, closes at $x = x_c$, and then re-opens when $x > x_c$ ($E_g < 0$). In the topological non-trivial phase (inverted regime), L_6^+ and L_6^- states form respectively the conduction and valence bands. This band inversion results in the emergence of the topological surface states (TSS) at four different points on the corresponding two-dimensional Brillouin zone (2DBZ). As can be seen in figure 1a, the projections of the L-points are located at the $\bar{\Gamma}$ and $\bar{M}$ points of the (111) 2DBZ. The bulk longitudinal valley results in the $\bar{\Gamma}$ -point Dirac cone (blue), while the three oblique valleys give rise to the $\bar{M}$ -point Dirac cones (green).

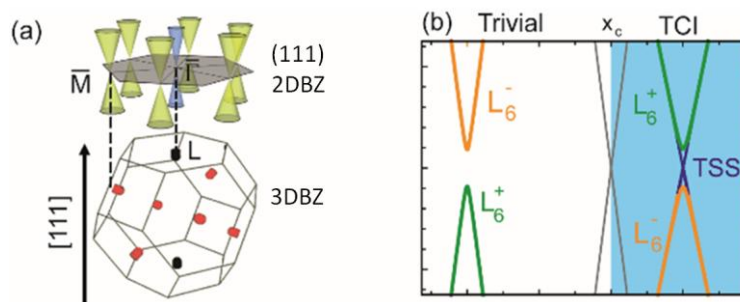


Figure 1. (a) The first 3D Brillouin zone of $Pb_{1-x}Sn_xTe$ accompanied by a corresponding (111) 2D Brillouin zone. (b) Schematic illustration of topological phase transition in $Pb_{1-x}Sn_xTe$ system.

2. Methods

Bi-doped (111)- $Pb_{1-x}Sn_xTe$ ($0 \leq x \leq 0.56$) films were grown by molecular beam epitaxy (MBE) on BaF_2 substrates. PbTe and SnTe effusion cells were employed to control the layer composition [4]. Light Bi-doping is necessary for high Sn content $Pb_{1-x}Sn_xTe$ films for compensating the intrinsic p-type carrier concentration ($p > 10^{19} \text{ cm}^{-3}$) originating from native (Pb,Sn) vacancies [5]. Bulk carrier densities $\sim 1 \times 10^{18} \text{ cm}^{-3}$ and high mobilities $\sim 10000 \text{ cm}^2/\text{Vs}$ are obtained from transport measurements at 77K. Far- and mid-infrared (FIR and MIR) magneto-optical absorption measurements in the Faraday geometry were performed on $Pb_{1-x}Sn_xTe$ samples at 4.5K and magnetic fields $B = 0-15\text{T}$. The spectral range is 4-450 meV. The sample pieces were placed in a superconducting coil. B is applied parallel to the [111] direction. Transmission spectra were acquired and analyzed by a Bruker Fourier transform infrared spectrometer. The relative transmission at a fixed B is defined as the normalization of the sample transmission $T(B)$ by the zero-field transmission $T(0)$. Further information about the sample growth, characterization and magneto-optical absorption experiment is given in [6].

3. Experimental results and analyses

3.1. Bulk states

Figure 2(a,b) show typical IR transmission spectra obtained for $x = 0.11$ and $x = 0.56$ at 4.5K. Strong transmission minima dispersing as a function of B can be clearly seen. Two series of transitions are associated with different bulk valleys, known to have a massive Dirac dispersion. The strongest series denoted by black arrows is attributed to the bulk longitudinal valley. Other transitions marked by red arrows result from the bulk oblique valleys.

The transmission minima are plotted as a function of B in figure 2(c,d) in order to construct transition fan diagrams. Full black circles and empty red circles denote respectively the transitions in the longitudinal and oblique valleys. The green rectangle represents the BaF_2 substrate reststrahlen band (22-55 meV) where no absorption can be observed. A massive Dirac model is used to fit the data for both types of bulk valleys. As the joint density of states is optimal for $k_z = 0$ (B and $z \parallel [111]$), the Landau levels (LLs) at $k_z = 0$ are sufficient to describe the observed magneto-optical interband transitions. The interband transition $(N^v - (N - 1)^c)$ energies, for p-type materials, in the massive Dirac model are given by [6] and [7]:

$$E_{N-1}^c - E_N^v = \sqrt{\Delta^2 + 2v_F^2 \hbar e B (N-1)} + \sqrt{\Delta^2 + 2v_F^2 \hbar e B N} \quad (1)$$

The conduction and valence energies of a LL index N are denoted by E_N^c and E_N^v , respectively. Δ is defined to be half of the energy gap E_g , $\Delta = E_g/2$. v_F is the Fermi velocity. The band edge mass of Dirac fermions is given by $m^* = |\Delta|/v_F^2$. e and $\hbar$ have their usual meaning.

The ground cyclotron resonance (CR) can be written as:

$$E_0^v - E_1^v = -|\Delta| + \sqrt{\Delta^2 + 2v_F^2 \hbar e B} \quad (2)$$

The black and red solid lines in figure 2(c,d) are the calculated transition energies. The CR-L and CR-O refer to the CR originating from the bulk longitudinal and oblique valleys, respectively. An excellent agreement with the massive Dirac fermion model is obtained in all investigated samples.

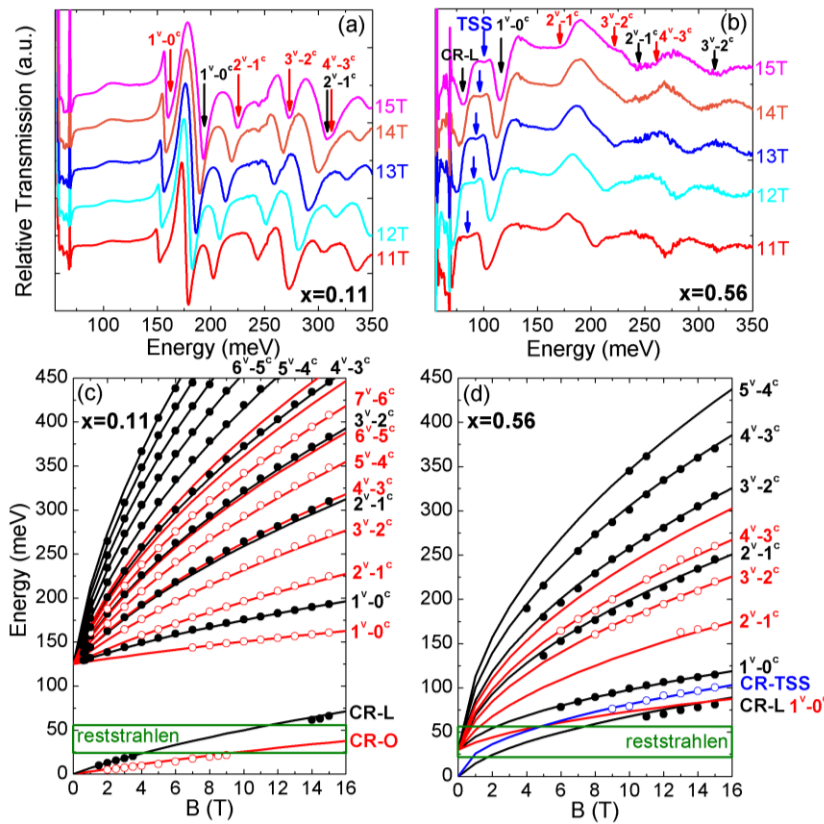


Figure 2. Magneto-optical transmission spectra of (111)- $\text{Pb}_{1-x}\text{Sn}_x\text{Te}$ measured at 4.5K and $B=11\text{--}15\text{T}$ for $x=0.11$ (a) and $x=0.56$ (b). The black and red arrows mark transmission minima resulting from the longitudinal and oblique valleys, respectively. All spectra are shifted vertically for clarity. Transition fan charts are constructed for $x=0.11$ (c) and $x=0.56$ (d). Black and red circles denote respectively the transitions in the longitudinal and oblique valleys. Black and red fits are calculated from a massive Dirac model. The transmission minima shown by blue arrows in (b) and blue circles in (d) are interpreted as the CR-TSS using a massless Dirac model. No absorption can be observed in the range 22-55 meV due to the BaF_2 reststrahlen band.

The massive Dirac model allows us to precisely determine band parameters such as E_g , v_F and m^* . Their values for the two samples $x=0.11$ and $x=0.56$ are shown in table 1.

Table 1. Magneto-optical band parameters obtained from the massive Dirac model analysis in $\text{Pb}_{1-x}\text{Sn}_x\text{Te}$ for $x=0.11$ and $x=0.56$ measured at 4.5K. The letters l and o denote respectively longitudinal and oblique valleys. m_0 is the electron rest mass.

Composition	E_g (meV)	$v_F^l (\times 10^5 \text{ m/s})$	$v_F^o (\times 10^5 \text{ m/s})$	m_l^*	m_o^*
$\text{Pb}_{0.89}\text{Sn}_{0.11}\text{Te}$	125 ± 5	8.15 ± 0.10	5.4 ± 0.1	$(0.017 \pm 0.001)m_0$	$(0.038 \pm 0.003)m_0$
$\text{Pb}_{0.44}\text{Sn}_{0.56}\text{Te}$	-30 ± 10	7.1 ± 0.1	4.9 ± 0.1	$(0.005 \pm 0.002)m_0$	$(0.011 \pm 0.004)m_0$

Moreover, one can estimate the band edge Fermi energy E_F by calculating the Landau level energy at a fixed B where the transmission disappears. In $\text{Pb}_{0.89}\text{Sn}_{0.11}\text{Te}$ the black transition $1^v - 0^c$ is measured

down to 0.6T, thus $E_F \sim 7.5$ meV below the valence band edge of the bulk longitudinal valley. In $\text{Pb}_{0.44}\text{Sn}_{0.56}\text{Te}$ the bulk longitudinal valley transition $1^v - 0^c$ vanishes at about 4-5T, one can thus get $E_F \sim 40$ meV.

3.2. Topological surface states

Besides the CR-L and the first interband transition $1^v - 0^c$ associated with the bulk longitudinal valley, additional weaker transmission minima indicated by blue arrows in figure 2b are observed in $\text{Pb}_{0.44}\text{Sn}_{0.56}\text{Te}$. Such minima can be interpreted as the CR of the TSS (CR-TSS) and are seen to unambiguously satisfy a massless Dirac model. The surface LL transition energies can be simply obtained by replacing $\Delta=0$ in equation 2. The energy of the CR-TSS is then given by:

$$E_{\text{CR-TSS}} = \sqrt{2v_F^2 \hbar e B} \quad (3)$$

The blue solid line in figure 2d is the calculated CR-TSS transition using the same Fermi velocity as the bulk longitudinal valley of $\text{Pb}_{0.44}\text{Sn}_{0.56}\text{Te}$, $v_F = (7.1 \pm 0.1) \times 10^5$ m/s. Since the Fermi velocities of the longitudinal bulk states and the TSS are found to be equal, this indicates that the observed TSS occur at the $\bar{\Gamma}$ -point of the (111) 2DBZ. These observations are in good agreement with recent theoretical calculations [3]. Moreover, since no CR-TSS transition below 8T can be observed, one can deduce a Fermi level in the $\bar{\Gamma}$ -Dirac cone $E_F \sim 75$ meV measured from the Dirac point. Such a CR-TSS is systematically measured in the non-trivial samples and is not observed in the trivial regime. Note that the TSS interband transitions are nearly located at the energy of the strong interband bulk transitions. This can be easily seen by considering the square root terms in equation 1. For $N \geq 1$ and $B > 1\text{T}$, $(2v_F^2 \hbar e B N)^{1/2} \approx (\Delta^2 + 2v_F^2 \hbar e B N)^{1/2}$, so that the TSS interband transitions cannot be experimentally resolved.

4. Conclusion

FIR and MIR magneto-optical absorption spectroscopy allows us to map out the LL spectrum in high-mobility Bi-doped (111)-oriented $\text{Pb}_{1-x}\text{Sn}_x\text{Te}$ ($0 \leq x \leq 0.56$) films grown on BaF_2 substrates using MBE. The bulk band parameters (E_g , v_F , m^*) of all samples can be accurately determined using the massive Dirac model. Topologically non-trivial samples ($x > 0.40$) are experimentally demonstrated to exhibit massless Dirac TSS occurring at the $\bar{\Gamma}$ -point. This is one of the first observations of TSS-related features by magneto-optics. In topologically trivial samples ($x < 0.40$), only bulk band transitions can be observed. This study paves the way for further work on the optical properties of topological Dirac fermions that are predicted to hold great promise for quantum electronic devices and data storage applications.

References

- [1] Ando Y and Fu L 2015 *Annu. Rev. Condens. Matter Phys.* **6** 361-81
- [2] Bauer G 1980 Magneto-optical Properties of IV-VI Compounds *Narrow Gap Semiconductors Physics and Applications* vol 133, ed Zawadzki W (Berlin Heidelberg: Springer) pp 427-46
- [3] Safaei S, Kacman P and Buczko R 2013 *Phys. Rev. B* **88** 045305
- [4] Springholz G and Bauer G 2013 Landolt-Börnstein – Group III Condensed Matter *Growth and Structuring* vol 34A, ed Klingshirn C (Verlag Berlin Heidelberg: Springer) pp 415-561
- [5] Ueta A Y, Springholz G, Schinagl F, Marschner G and Bauer G 1997 *Thin Solid Films* **306** 320
- [6] Assaf B A, Phuphachong T, Volobuev V V, Inhofer A, Bauer G, Springholz G, de Vaultier L A and Guldner Y 2016 *Sci. Rep.* **6** 20323
- [7] Mitchell D L and Wallis R F 1966 *Phy. Rev.* **151** 581

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