

# Space charge limited current conduction in $\text{Bi}_2\text{Te}_3$ thin films

R. Sathyamoorthy<sup>a,\*</sup>, J. Dheepa<sup>a</sup>, S. Velumani<sup>b</sup>

<sup>a</sup> PG and Research Department of Physics, Kongunadu Arts and Science College, Coimbatore-029, Tamil Nadu, India

<sup>b</sup> Department of Physics, ITESM-Campus, Monterrey, N.L., Mexico

Received 30 June 2006; received in revised form 5 October 2006; accepted 16 November 2006

## Abstract

$\text{Bi}_2\text{Te}_3$  is known for its large thermoelectric coefficients and is widely used as a material for Peltier devices.  $\text{Bi}_2\text{Te}_3$  thin films with thicknesses in the range 125–300 Å have been prepared by Flash Evaporation at a pressure of  $10^{-5}$  m bar on clean glass substrates at room temperature. An Al– $\text{Bi}_2\text{Te}_3$ –Al sandwich structure has been used for electrical conduction properties in the temperature range 303 to 483 K.  $I$ – $V$  characteristics showed Ohmic conduction in the low voltage region. In the higher voltage region, a Space Charge Limited Conduction (SCLC) takes place due to the presence of the trapping level. The transition voltage ( $V_t$ ), between the Ohmic and the SCLC condition was proportional to the square of thickness. Further evidence for this conduction process was provided by the linear dependence of  $V_t$  on  $t^2$  and  $\log J$  on  $\log t$ . The hole concentration in the films were found to be  $n_0 = 1.65 * 10^{10} \text{ m}^{-3}$ . The carrier mobility increases with increasing temperature whereas the density of trapped charges decreases with increasing temperature. The barrier height decreases with an increase in temperature. The increase in the trapping concentration  $V_t$  is correlated with ascending the degree of preferred orientation of the highest atomic density plane. The activation energy was estimated and the values found to decrease with increasing applied voltage. The zero field value of the activation energy is found to be 0.4 eV.

© 2006 Elsevier Inc. All rights reserved.

**Keywords:**  $\text{Bi}_2\text{Te}_3$  thin films; DC conduction; SCLC; Activation energy; Carrier mobility; Barrier height

## 1. Introduction

The materials most commonly used for thermo-electric conversion are semi-metals and narrow gap semiconductors [1,2]. Recently there has been increased interest in finding additional materials for use in cleaner, more efficient cooling systems and hence Bi–Te thin films have attracted considerable attention because of these potential applications in the micro-fabrication of integrated thermo-

electric devices [3,4].  $\text{Bi}_2\text{Te}_3$  have been extensively investigated in bulk crystalline form because of its excellent thermo-electric properties [5], whereas in thin film form so far relatively very little attention has been paid. However, the thin films have the potential to lead key technology in the low power generation [6]. From the measurements of the conductivity, the thermo-electric power and the magnetic susceptibility, Honda and Sone [7] and Endo [8] have found that an intermetallic compound  $\text{Bi}_2\text{Te}_3$  exists and Haken [9] has reported that the compound  $\text{Bi}_2\text{Te}_3$  is of p-type from the measurement of the Hall effect at room temperature.

Considerable interest has been shown in conduction studies on various semiconducting materials, however until recently there are virtually no reports available on the

\* Corresponding author. Department of Physics, Kongunadu Arts and Science College, Coimbatore-641 029, Tamil Nadu, India. Tel.: +91 422 642095; fax: +91 422 644452.

E-mail addresses: rsathya\_59@yahoo.com, rsathya59@yahoo.co.in (R. Sathyamoorthy).

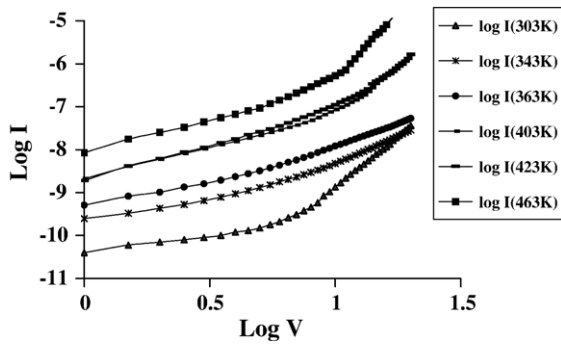


Fig. 1.  $V$ – $I$  characteristics for an Al/Bi<sub>2</sub>Te<sub>3</sub>/Al device at various temperature.

conduction studies on Bi<sub>2</sub>Te<sub>3</sub> thin films. The present article deals with a systematic study of the electrical conduction behavior of a Bi<sub>2</sub>Te<sub>3</sub> thin film sandwiched between aluminium electrodes. Current voltage characteristics under a dc field have been studied at different temperatures (303–483 K) under a rotary vacuum. The influence of the temperature on the electrical parameters is discussed in detail.

## 2. Experimental

Using a Hind Hi Vacuum Unit, aluminium of 5N purity was evaporated from a tungsten filament onto well-cleaned glass substrates through suitable masks to form the base electrodes. High purity Bi<sub>2</sub>Te<sub>3</sub> (99.999% of Aldrich Chem. Comp) was evaporated by a flash evaporation method to form the dielectric layer. The thickness was determined by a quartz crystal thickness monitor and was verified by a multiple beam interferometric technique (MBI). An aluminium counter electrode was evaporated onto the dielectric through suitable masks to complete the Al/Bi<sub>2</sub>Te<sub>3</sub>/Al structure. During the electrical measurements, samples were placed in a metal canister, which was

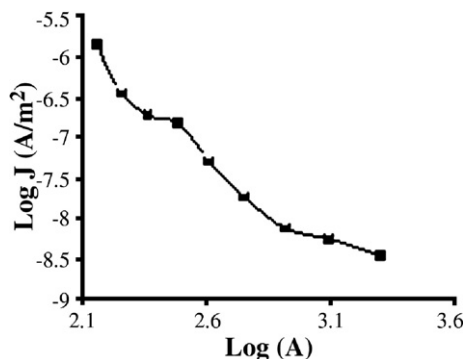


Fig. 2. Dependence of current density ( $J$ ) on thickness ( $d$ ) in the square law region.

Table 1

Free carrier mobility for Bi<sub>2</sub>Te<sub>3</sub> thin film ( $d=120$  Å) at various temperatures

$T$ (K)	Free carrier mobility ( $\mu_0$ ) at 2 V ( $10^{-10}$ m <sup>2</sup> V <sup>-1</sup> s <sup>-1</sup> )
303	1.2
343	1.8
383	4.2
423	7.9
463	21.5
483	28.6

evacuated by a rotary pump. A stabilized dc power supply was used in conjunction with a digital micro-voltmeter and a digital pico-ammeter (DPM 111, Scientific equipment, Roorkee) for voltage and current measurements at different temperatures (303–483 K). A copper constant thermocouple was used to measure the temperature.

## 3. Results and discussion

The variation of current as a function of voltage at different temperatures on a Bi<sub>2</sub>Te<sub>3</sub> thin film of thickness 120 Å is shown in Fig. 1. It is observed that the curve for each temperature exhibits three regions namely AB, BC and CD. In the region AB ( $<2$  V) the conduction is Ohmic ( $I \propto V$ ), which leads us to understand that thermally-generated carriers control the current. At applied voltage  $<2$  V, the slopes of the  $\log I$  vs  $\log V$  plots are approximately equal to unity, while at higher voltages ( $>2$  V), above a well-defined transition voltage, the slopes are approximately equal to two or more. In the region BC, corresponding to approximately 9 to 13 V, a trap square law region ( $I \propto V^2$ ) is obtained followed by the region CD where  $I \propto V^n$  ( $n > 5$ ). In the region BC, it is observed that the current increases, with an increase of the temperature for the same applied voltage. The reduction of the square law region caused by an increase of the temperature may be due to the predominance of injected free carriers over the trapped carriers at higher temperatures [10]. Fig. 2 represents the variation of current density ( $J$ ) with

Table 2

Electrical parameters derived from  $J$ – $V$  plot

Parameters	Al/Bi <sub>2</sub> Te <sub>3</sub> /Al
$P_0$ (m <sup>-3</sup> )	$1.1 \times 10^{16}$
$P_1$ (m <sup>-3</sup> )	$6.21 \times 10^{17}$
$J_s$ (A/m <sup>2</sup> )	$2.66 \times 10^{-6}$
$N_a$ (m <sup>-3</sup> )	$4.21 \times 10^{17}$
$\Delta E$ (eV)	0.411
$\Phi b$ (eV)	0.267
$E_f$ (eV)	0.110

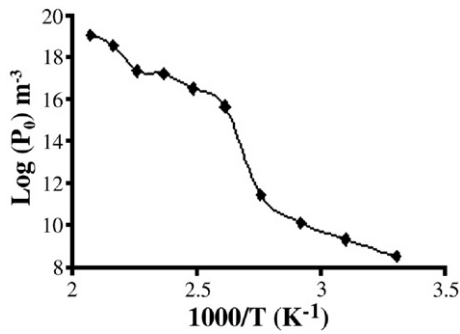


Fig. 3. Effect of temperature on free hole density ( $P_0$ ).

thickness ( $d$ ) at a temperature 303 K in the square law region (12 V) and from the plot, the slope of the straight line is found to be 2.97. Thus it can be concluded that the electrical conduction in  $\text{Bi}_2\text{Te}_3$  thin films is space charge limited due to the presence of traps in  $\text{Bi}_2\text{Te}_3$  thin films [11]. Since holes are the dominant carriers in  $\text{Bi}_2\text{Te}_3$  thin films [12], the equation describing the  $J$ – $V$  characteristics in the Ohmic region is given by:

$$J = P_0 q \mu_0 \frac{V}{d} \tag{1}$$

where  $\mu_0$  is the mobility of holes and  $P_0$  is the concentration of free carrier in the valence band given by:

$$P_0 = N_v \exp\left(\frac{-E_f}{kT}\right) \tag{2}$$

where  $E_f$  is the position of the Fermi level above the valence band edge and  $N_v$  is the density of states in the

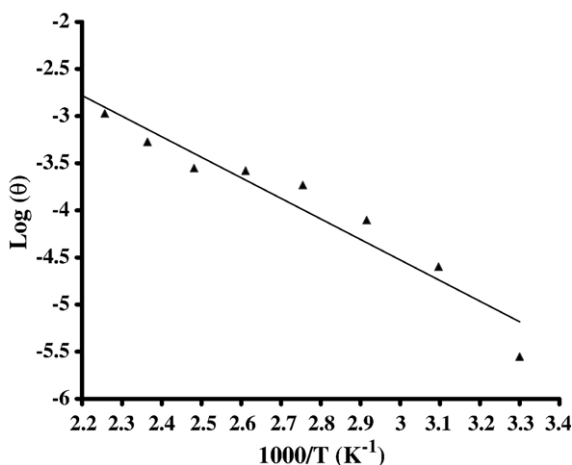


Fig. 4. Variation of  $\theta$  with inverse temperature.

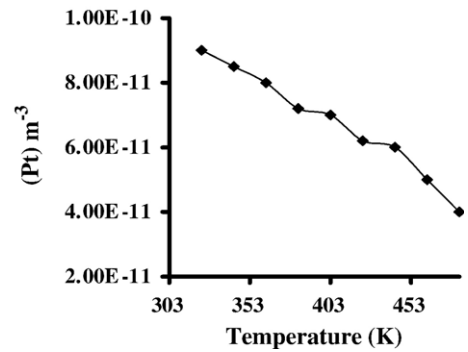


Fig. 5. Variation of trapped hole density ( $P_t$ ) with temperature.

valence band. Using Eqs. (1) and (2) the current density can be evaluated using the relation:

$$J = N_v q \mu_0 \left(\frac{V}{d}\right) \exp\left(\frac{-E_f}{kT}\right) \tag{3}$$

The position of the Fermi level above the valence band edge and mobility of holes can be calculated from the  $\log I$  against  $1000/T$  plot. At higher voltages ( $>2$  V) the slopes of the  $\log J$ – $\log V$  characteristics, are about 2 and more, clarifying that the forward-biased current is Space Charge Limited (SCL) controlled by a trap levels. The relation for the current density [13] is given by:

$$J = \left(\frac{9}{8}\right) \epsilon \epsilon_0 \mu_0 \theta \left(\frac{V^2}{d^3}\right) \tag{4}$$

where  $J$  is the current density,  $V$  the applied voltage,  $\mu_0$  the mobility,  $\epsilon$  the dielectric constant of the material

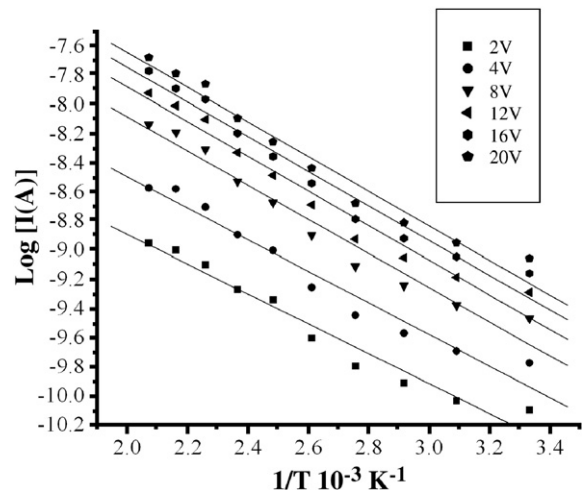


Fig. 6. Variation of  $\log I$  with  $1/T$  in SCLC region for  $\text{Al}/\text{Bi}_2\text{Te}_3/\text{Al}$  device.

(0.053),  $d$  the distance between the electrodes and  $\theta$  is the ratio of free to trapped charge densities, with  $N_t(s)$  representing the total trap density at the energy level  $E_t$ .

The total trap concentration at the valence band energy is given by the expression:

$$P_t = P_0 kT \tag{5}$$

where  $P_0$  and  $P_t$  are the free and trapped hole densities respectively.  $N_v$  is the effective density of states for the valence band and  $E_f$  the trap level measured from the top of the valence band. Experimentally  $\theta$  is proportional to the ratio between the current densities at the beginning ( $I_1/A$ ) and at the end of the rise ( $I_2/A$ ) in the square law region [14] where  $A$  is the area of the capacitor ( $3.49 \times 10^{-6} \text{ m}^2$ ). Thus:

$$\frac{I_1}{I_2} = \theta = \left( \frac{P_0}{(P_0 + P_t)} \right) \tag{6}$$

The growth of the current faster than  $V^2$  is attributed to the effect of traps after the square law region. Using the experimental value of  $\theta$  and  $\epsilon$  the free carrier mobility  $\mu_0$  was calculated at 2 V and the values are given in Table 1.

The equilibrium concentration of charge carriers in the valence band is given by [11]:

$$P_0 = \left[ \frac{\epsilon \epsilon_0 \theta}{qd^2} \right] V_{tr} \tag{7}$$

where  $V_{tr}$  is the voltage at which transition from Ohmic to the square law region takes place,  $q$  is the electronic charge and  $d$  is the thickness of the film. Using Eq. (7)  $P_0$  and  $P_t$  are calculated and the values are given in Table 2. Fig. 3 represents the dependence of  $P_0$  with the inverse absolute temperature and this plot yields a straight line. From the slope and intercept on the current axis, the value of  $E_f$  is determined, as 0.1 eV and  $N_a$  is found to be  $4.21 \times 10^{17} \text{ m}^{-3}$ . From Fig. 4 the zero field activation energy has been estimated by additional plotting of the linear portion and it is found to be dependent on temperature. The decrease in  $P_t$  shown in Fig. 5 may be

Table 3  
Activation energy as a function of applied voltage

Voltage (V)	DC activation energy (eV)	Zero field energy (eV)
2	0.32	
4	0.28	
8	0.26	0.45
12	0.23	
16	0.15	

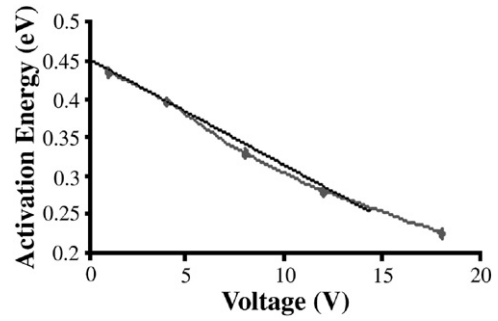


Fig. 7. Variation of activation energy with inverse applied voltage.

due to the fact that a rise in temperature lowers the trapping probability in semiconductors. The increase in  $\mu_0$  with temperature may be due to the semiconducting nature of  $\text{Bi}_2\text{Te}_3$  thin films.

#### 4. Activation energy

The temperature dependence of the current at various applied voltages were recorded and analysed. The activation energy has been determined using the relations [11]:

$$I \propto \exp\left(\frac{-\Delta E}{kT}\right) \tag{8}$$

$$I = I_0 \exp\left(\frac{-E}{kT}\right) \tag{9}$$

From the slope of the plot Fig. 6 of logarithmic current against  $1000/T$  and by using Eq. (8), the activation energies have been estimated and are given in Table 3. It is clear from the table that the activation energy decreases with increasing applied voltage. The intercept with the origin gives the zero field value of the activation energy. From Fig. 7, it is clear that the fit is giving a zero field value of the trap energy as 0.445 eV.

#### 5. Conclusions

Current–voltage measurements showed Ohmic conduction at lower voltages and SCL conduction at higher voltages. The transition voltage,  $V_t$ , between the Ohmic and the SCL conduction was proportional to  $t^2$ . Various electrical parameters such as saturation current density, barrier height, density of states and activation energies in the valence band were determined and it was found to be decrease with an increase in applied voltage. The barrier height decreases with an increase in temperature.

## Acknowledgments

The authors acknowledge the Secretary and the Management of Kongunadu Arts and Science College, Coimbatore for their encouragement and support to carry out this work.

## References

- [1] Van Her Waarden AW, Sarro PM. Thermal sensors based on the seebeck effect. *Sens Actuators* 1986;10:321–46.
- [2] Lovett DR. *Semimetals and narrow band semiconductors*. London: Pion; 1977.
- [3] Shafi C, Brett MJ. Optimization of  $\text{Bi}_2\text{Te}_3$  thin films for microintegrated Peltier heat pump. *J Vac Sci Technol A Vac Surf Films* 1997;15:2798–801.
- [4] Min Gao, Rowe DM. Cooling performance of integrated thermoelectric microcooler. *Solid State Electron* 1999;43:923–9.
- [5] Goldsmid HJ. *Electronic refrigeration*. Lindon: Pion; 1986.
- [6] Boyer A, Cisse E. Properties of thin film thermoelectric materials: applications to sensor using the Seebeck effect. *Mater Sci Eng B Solid-State Mater Adv Technol* 1992;13:103–11.
- [7] Honda K, Sone T. Science reports of Tohoku Imperial University, First series, vol. 2; 1913. p. 1.
- [8] Shing YH, Chang Y, Mirshafii A, Hayashi L, Roberts SS, Josefowicz JY, et al. Sputtered  $\text{Bi}_2\text{Te}_3$  and  $\text{PbTe}$  thin films. *J Vac Sci Technol A Vac Surf Films* 1983;1:503–6.
- [9] Haken W. Growth of p and n type bismuth telluride thin films by co-evaporation. *Ann Phys* 1910;32:291.
- [10] Mangalaraj D, Radhakrishnan M, Balasubramanian C, Kasilingam AR. Current–voltage characteristics of aluminium nitride films formed by RF glow discharge. *J Phys D* 1980;13:L101–5.
- [11] Goswami A. *Thin film fundamentals*. India: New Age International (P) Limited; 1996.
- [12] Helin Zou, Rowe DM, Min Gao. Growth of p- and n-type bismuth telluride thin films by co-evaporation. *J Cryst Growth* 2001;222:82–7.
- [13] Gertstenberg D, Maissel LI, Glang R. *Handbook of Thin Film Technology*, Eds. McGraw-Hill Co., New York, (1970).
- [14] Subbarayan A, Balasubramanian C, Narayandass Sa K. Structural, aging, Annealing and Electrical properties of Perylene thin film. *Indian J Pure Appl Phys* 1988;26:410.