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Characterization of Bismuth Telluride thin films — Flash evaporation method

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Abstract

Semiconducting Bi_2Te_3 thin films were grown by a flash evaporation method. The optical characteristics of Bi_2Te_3 samples have been analyzed using a spectrophotometer in the wavelength range of 400–800 nm. The transmittance falls steeply with decreasing wavelength. It reveals that Bi_2Te_3 films have considerable absorption throughout the wavelength region. The effect of thickness on the fundamental optical parameters like refractive index and extinction coefficient are studied. The optical band gap energy decreases typically from 1.4 to 1.0 eV with increasing thickness, which may be due to the increased grain size of the films at higher thicknesses. Bi_2Te_3 thin films with a thickness of 120–300 Å exhibit semiconducting behavior with an exponential decrease in resistivity with increasing temperature ranging from 303 to 483 K. To examine the temperature degeneracy of the resistivity in more detail, log ρ Vs $1/T$ graphs are drawn and the activation energy calculated from these graphs decreases with increasing thickness in the range 0.16–0.06 eV. This decrease in activation energy with thickness of the film may be due to the change in barrier height. The change in barrier height is due to the change of crystallite size in the films.

Keywords: Bismuth Telluride; Thin films; Optical properties; Resistivity; Activation energy

1. Introduction

Thermoelectric devices can be used in a wide temperature range as solid-state coolers, as power generators and as sensors or detectors [1,2]. Specific cooling applications include shipboard air conditioning, IR detection systems, cryogenic electronics, etc. However, their use has been limited by the relatively low energy conversion efficiency. Bismuth Telluride (Bi_2Te_3) and its alloys are most important semiconductor thermoelectric materials used in fabrication of devices for the 200–400 K temperature range. The figure of merit of the best Bi_2Te_3 based alloys is about one [3]. Many studies have been made in recent years to improve the thermoelectric properties of Bi_2Te_3 [4–7]. It is a common practice to fabricate Bi_2Te_3-based thermoelectric materials by single crystal growth [8,9], sintering [10], mechanical alloying [11–13], hot pressing [14], hot extrusion [15], electrochemical atomic layer epitaxy [16,17] etc. However, not much work has been reported on flash evaporated Bi_2Te_3 thin films. Hence this paper reports the optical and electrical properties of Flash Evaporated Bi_2Te_3 thin films.

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2. Experimental

Bismuth Telluride thin films were deposited onto ultrasonically cleaned glass substrates at room temperature. High purity material (99.999% Aldrich Chem. USA) was used for the evaporation. The vaporising source in the flash evaporation technique is a thin molybdenum boat heated by the Joule effect. Its temperature can be controlled by means of chromel–alumel thermocouple. The powder distributor consisted of an ultrasonic storage chamber ending in a slightly inclined V-shaped chute. The Bi_2Te_3 vapor flows continuously along the chute at a rate controlled by the vibrational frequency of the ultrasonic storage chamber. The thickness of the film was measured by a Multiple Beam interferometer technique by forming Fizeaue [18] fringes. The samples were analysed by an X-ray diffractometer (Rigaku, Japan) operated at 30 kV with filtered CuKα radiation of wavelength 1.5405 Å. Optical investigations in the visible range were performed using a spectrometer (Jasco Corp, V-570). Electrical resistivity measurements in the range 303–453 K as a function of temperature were performed on Bi_2Te_3 films of different thickness using a Four Probe method.

3. Results and discussion

X-ray diffraction studies were made on Bi_2Te_3 thin films deposited on glass substrates to determine their structural parameters. Fig. 1 shows the X-ray diffraction pattern of Bi_2Te_3 thin films of thicknesses 125, 200 and 280 Å and they have been found to be amorphous in nature. The compositional analysis of the films was carried out by EDAX and the elemental percentages were found to be 52.48% and 47.52% for Bi and Te respectively.

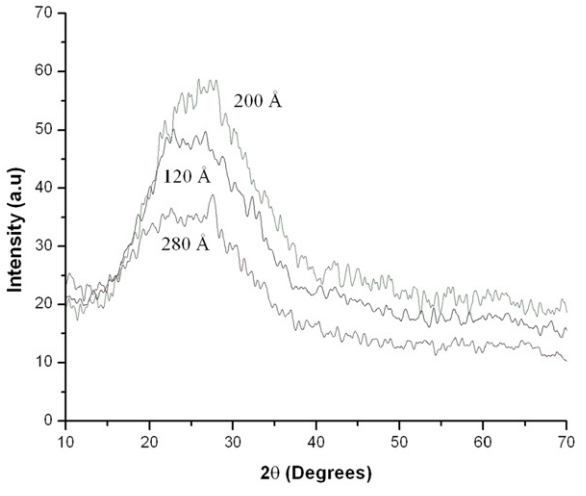

Fig. 1. XRD spectrum of Bi_2Te_3 thin films of different thickness.

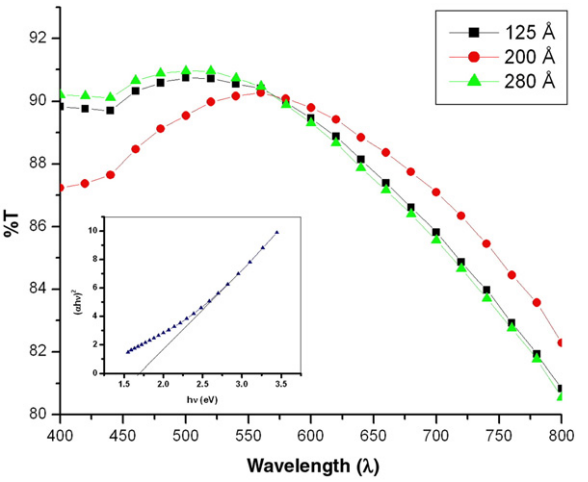

Fig. 2. Transmission spectra of Bi_2Te_3 films of various thickness. Inset Fig. $(\alpha h\nu)^2$ Vs $h\nu$ plot for the Bi_2Te_3 film of thickness 125 Å.

Transmission characteristics of Bi_2Te_3 films are given in Fig. 2. The figure clearly indicates that the transmission is found to decrease with increasing wavelength and no specific trend is observed with change in thickness. The transmittance falls steeply with decreasing wavelength. This reveals that the Bi_2Te_3 films are having considerable absorption throughout the visible region. The energy band gap of the films was calculated with the help of transmission spectra, using the Tauc [19] relation:

$$\alpha h\nu = A(h\nu = \mathrm{Eg})^n$$

where

$h\nu$	Photon energy
α	Absorption coefficient
Eg	Band gap
A	Constant
n	½ for direct band gap
n	2 for indirect band gap.

From the inset figure in Fig. 2, $(\alpha h\nu)^2$ Vs $h\nu$ plot, the optical band gap energy has been estimated for different thickness of the films and given in Table 1. The possible transitions in the Bi_2Te_3 thin films are direct and allowed. It

Table 1
Variation of the optical band gap and activation energy with film thickness

Thickness (Å)	Band gap (eV)	Activation energy (eV)
125	1.62	0.16
200	0.96	0.07
280	0.43	0.06

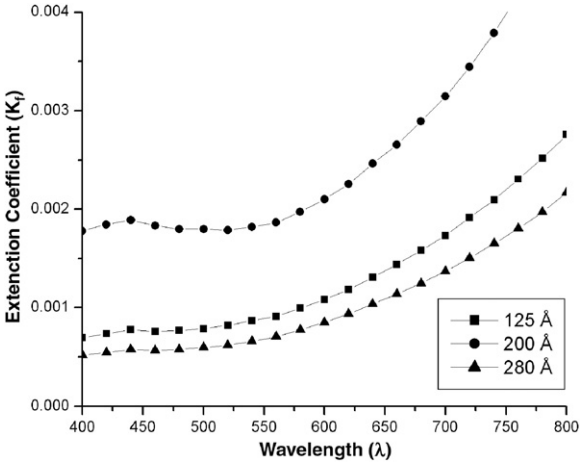

Fig. 3. Variation of extinction coefficient with wavelength.

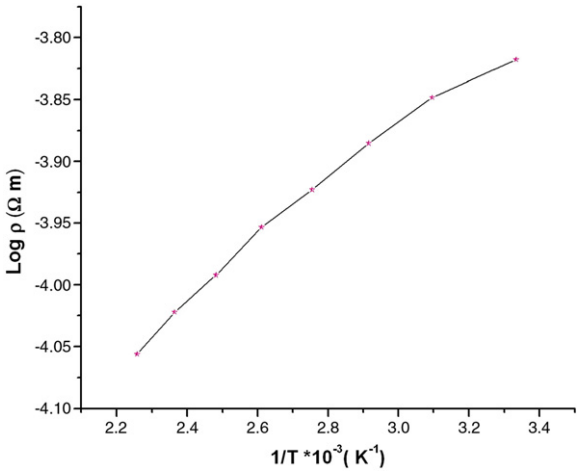

Fig. 5. Variation of log ρ as a function of the $1/T$ of Bi_2Te_3 thin films.

is observed from the table that the optical band gap energy decreases with an increase in film thickness. The decrease in optical band gap energy with increase in film thickness is due to the increase in crystallite size for thicker films. The adatom mobility also increases with thickness, which also results in an increase in the crystalline nature, which in turn decreases the band gap [20–22].

Fig. 3 shows the variation of extinction coefficient with wavelength for various thicknesses from which it is observed that an increase in wavelength increases the extinction coefficients [23–25]. Fig. 4 shows the variation of refractive index with wavelength for different thickness. It clearly shows that the refractive index increase as the thickness increases. There is a slight variation with respect to the wavelength, which may be attributed to the strong effects of surface and volume imperfections at the microscopic scale.

The temperature dependence of the resistivity was examined by electrical resistance measurements in the temperature range 303–453 K for various thicknesses under a constant current source (1 mA). The resistance is found to have a linear dependence on inverse thickness at all temperatures, which may be probably due to the discontinuous structure of the film. It is observed that the carriers were strongly, non-degenerate and hence the resistivity decreases with increase in temperature [26]. Also the Bi_2Te_3 thin films studied in the present study exhibit the normal semiconducting behavior of an exponential decrease of resistivity with temperature.

The activation energies have been evaluated from the graph plotted between ln ρ Vs $1/T$. The graph to calculate the activation energy for the Bi_2Te_3 thin film of a thickness 125 Å is shown in Fig. 5 and the activation energies for other films of various thicknesses were calculated in a similar way and the values are given in Table 1. It is seen that the activation energy decreases as the thickness increases [27] which can be explained on the basis of an island structure of the film formation and thereby, tunneling of charged carriers between islands separated by a few Angstroms in distance.

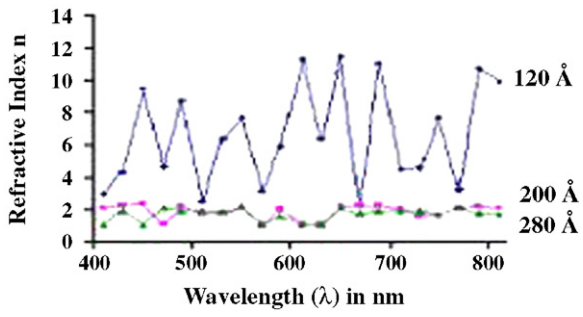

Fig. 4. Spectral response of the refractive index for samples of different thickness.

4. Conclusions

The Bi_2Te_3 thin films grown by flash evaporation have been found to exhibit an amorphous nature. The possible optical transitions in these films are found to be direct and allowed. The optical band gap decreases with an increase in wavelength. The resistivity decreases with an increase of film thickness. The activation energy increases with decreasing film thickness.

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