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Electrical properties of polycrystalline Sb_2Te_3 films

N. G. PATEL, P. G. PATEL
Department of Electronics, Sardar Patel University, Vallabh Vidyanagar 388 120, Gujarat, India

Single-phase polycrystalline stoichiometric films of Sb_2Te_3 with different thicknesses were prepared on glass substrates by a flash evaporation technique at constant substrate temperature of 423 K. The electrical properties of these films, such as resistivity, Hall mobility, carrier concentration and activation energy, were determined for different film thicknesses. The optical absorption of these films was also studied. The implications are discussed.

1. Introduction

The electrical and optical properties of V_2–VI_3 (V = Bi, Sb; VI = S, Se, Te) compounds are interesting because of possible use in electromechanical devices [1], optical recording systems [2], and thermoelectric devices [3–6]. It is known that these compounds have a narrow band gap and possess a layered crystal structure [1, 7]. It appears from a survey of the literature that so far few reports on the electrical properties of thin films of such compounds have been published, particularly Sb_2Te_3 compound. Rajagopalan and Ghosh [8] reported electrical and thermoelectric properties of Sb_2Te_3 films deposited by a direct evaporation method. Patel and Patel [9] reported that Sb_2Te_3 films grown at 423 K by flash evaporation method were found to be single-phase polycrystalline, stoichiometric and to exhibit a minimum electrical resistivity. The present work was undertaken with a view to obtain information on the variation of the electrical and optical properties of Sb_2Te_3 thin films as a function of the thickness, keeping other variables such as substrate temperature, rate of evaporation, pressure in the chambers, nature of the substrate, etc., constant, which may be important for the fabrication of devices.

2. Experimental procedure

The Sb_2Te_3 solid ingot was prepared by fusing high-purity antimony and tellurium (99.999% pure) in stoichiometric proportions in a vacuum-sealed quartz ampoule at a temperature about 1140 K. Sb_2Te_3 films were prepared by flash evaporation from fine grains of Sb_2Te_3 solid ingot on thoroughly cleaned glass substrates in a vacuum of 10^{-4} Pa. The substrate temperature was kept constant at about 423 K. The deposition rate was kept constant at about 1–2 nm s^{-1}, and the thickness of the films was measured using the multiple-beam interference technique. Structural studies of the films were made by transmission electron microscope (EM400 Philips) and the composition of the films was checked by energy dispersive analysis of X-rays (EDAX 9100/60 System). The electrical properties were measured along the plane perpendicular to the c-axis using the Van der Pauw method. Vacuum-deposited silver films with silver-pasted leads provided ohmic contacts. The optical absorption spectra were recorded using a Beckman DK-2A spectrophotometer.

3. Results and discussion

The transmission electron micrographs and the corresponding diffraction patterns of Sb_2Te_3 films of different thicknesses are shown in Fig. 1. It is observed from the electron micrographs that the grain size increases as the thickness of the film increases, which leads to a decrease in the intercrystalline barrier. It is revealed from the electron diffraction patterns that Sb_2Te_3 films have a single-phase polycrystalline structure. The calculated d-values from the electron diffraction patterns are in good agreement with the standard d-values [10]. Thus, significant textural changes are not observed as the film thickness changes.

Fig. 2 shows the variation in the electrical resistivity with increasing film thickness. The resistivity decreases with increasing film thickness and starts to saturate at a thickness of about 300 nm. Generally, for thicknesses up to about 20 nm, the film has an island structure and carrier transfer between the islands occurs by either field-emission or field-assisted thermionic emission. Beyond this thickness the film is essentially continuous, but resistivity changes with thickness because of an appreciable surface scattering effect. Sondheimer's [11] theory qualitatively explains that the scattering of carriers at the surface of the film effectively reduces the mean free path of the carriers, so that the resistivity decreases with increasing thickness. It is inadequate to offer a quantitative explanation, because during the initial stage of growth of films, it contains defects, voids, intercrystalline barriers and impurities which also influence the resistivity. Beyond 300 nm thickness, the resistivity starts to approach that of a bulk value, because the surface scattering

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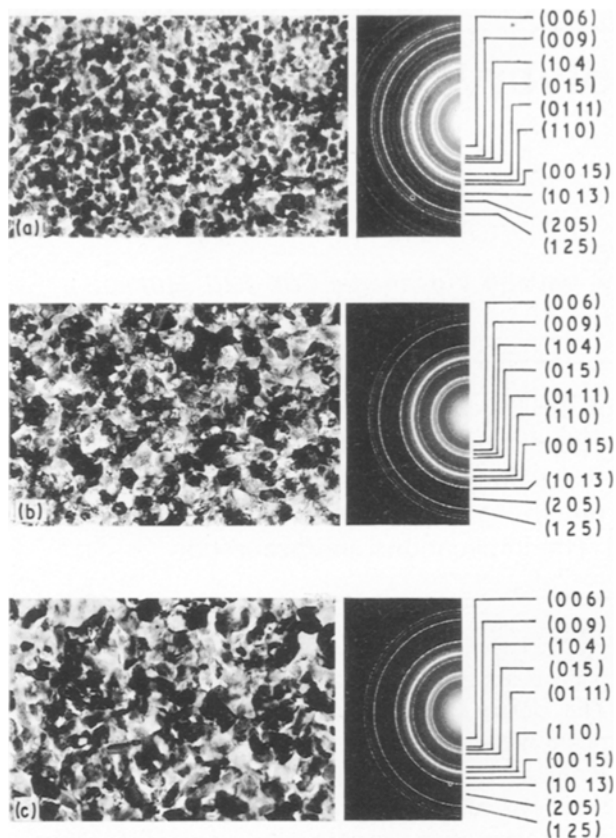

Figure 1 Transmission electron micrographs and the corresponding diffraction patterns of Sb_2Te_3 films with different thicknesses: (a) 100 nm, (b) 200 nm, (c) 300 nm (× 64 000).

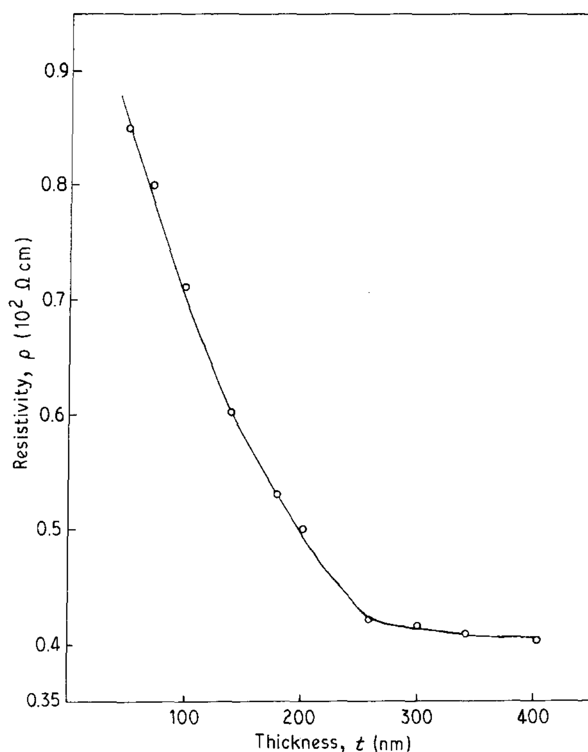

Figure 2 Dependence of the electrical resistivity, ρ, on thickness, *t*, of Sb_2Te_3 film.

effects become negligible and lattice scattering becomes prominent. It has also been observed that the resistivity of very thick film is higher than the reported value of 10^{-2}–10^{-3} Ω cm in bulk Sb_2Te_3 crystal [12]. This may be due to surface unevenness of thin films, as reported by Lucas [13].

Hall mobility (μ_H) and carrier concentration (n) of the Sb_2Te_3 films of different thicknesses have been measured at room temperature and are shown in Fig. 3. During Hall measurements, it was observed that all films are p-type. It was also observed that an increase in carrier concentration with decreasing thickness, ultimately results in a decrease in Hall mobility in the films. This may be explained as being due to the increase in the grain size and imperfection of these films. It should be mentioned that Hall mobility in films drops with increase in carrier concentration more vigorously than in single crystals. This may be due to the presence of a scattering effect in the films, whose contribution increases with decrease in film thickness.

Table I shows the estimated value of the grain size of Sb_2Te_3 films from the electron micrographs and the calculated value of the mean free path for the collision of the carriers (l) using the formula [14]

$$l = \frac{h}{2e}\left(\frac{3n}{\pi}\right)^{1/3} \mu_H \qquad (1)$$

where h is Planck's constant and e is charge of elec-

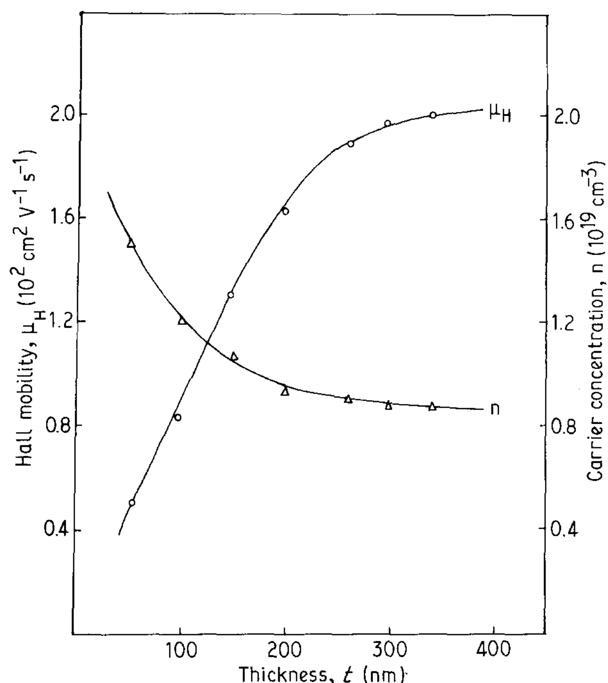

Figure 3 Variation in the Hall mobility, μ_H, and carrier concentration, n, of Sb_2Te_3 films with different thicknesses.

TABLE I Values of the grain size and the mean free path of the carriers at different thicknesses of Sb_2Te_3 films

Thickness, *t* (nm)	Grain size, *D* (nm)	Mean free path of carriers (nm)
50	24.0	2.8
100	46.0	3.7
200	78.0	7.1
250	90.0	7.9
300	93.0	8.1

tron. It is observed from Table I that the increase in the thickness of the films increases the grain size, which leads to increase in the mean free path for the collision of the carriers. It is also observed that the mean free path of the carriers is shorter than the grain size. It is likely that the scattering of carriers in thicker film may be due to imperfection and ionized impurity centres, as reported by Noguchi and Sakata [14]. Rajagopalan and Ghosh [8] have reported variation of the Hall mobility with thickness of Sb_2Te_3 films. However, the observed value of mobility in the present case is about an order of magnitude higher than that reported by them. This may be due to the difference in the growth conditions.

The variation of resistance as a function of temperature is used to determine the activation energy (ΔE). The variation of $\log R$ with inverse temperature for different thicknesses of Sb_2Te_3 films is shown in Fig. 4. It is observed that as the temperature increases the resistance decreases. The straight line nature of these plots indicates the single-phase nature of Sb_2Te_3 polycrystalline films. The activation energy is found to be 0.297, 0.215 and 0.137 eV for thickness of 100, 150 and 200 nm, respectively, by equating the slope to $\Delta E/2K$. It is seen that ΔE increases with decrease in film thickness. This behaviour is usually explained by the island structure theory of Negubauer [15].

The study of the interband absorption spectrum near the threshold of its rapid increase can obviously provide information on the structure of the electron's energy spectrum near the top edge of the valence band and the bottom edge of the conduction band; this information is of essential importance in the determination of the semiconductor's electrical properties. The optical absorption, α, for interband transitions [16] in a semiconductor close to the gap is given by

$$\alpha = (A/h\nu)(h\nu - E_g)^{n/2} \quad (2)$$

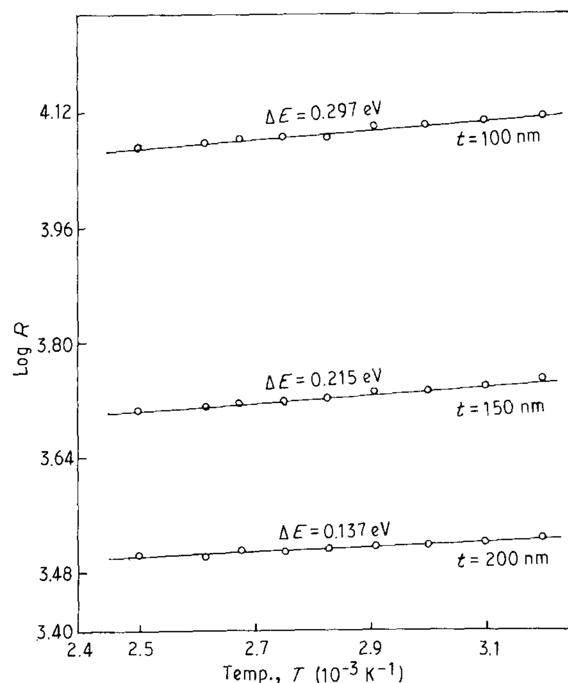

Figure 4 Plot of log of resistance, R, versus inverse of temperature T^{-1}, for different thicknesses of Sb_2Te_3 films.

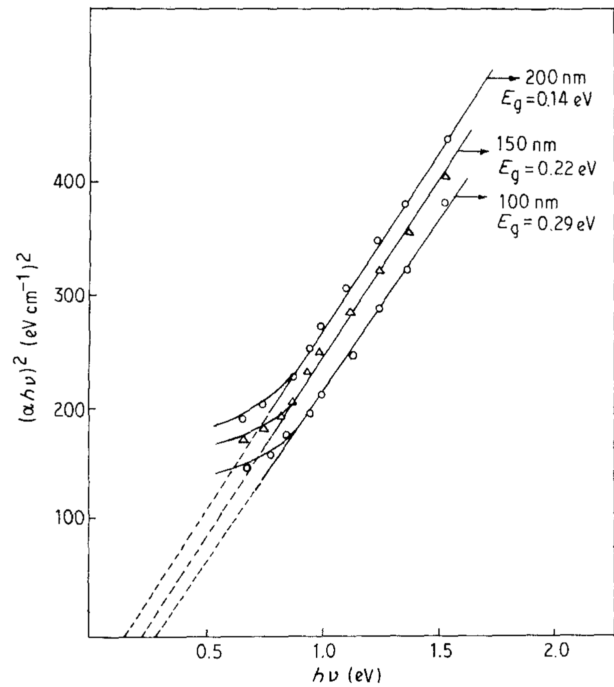

Figure 5 Plot of $(\alpha h\nu)^2$ versus the photon energy ($h\nu$) for different thicknesses of Sb_2Te_3 films.

where A is a constant, $n = 1$ for direct and $n = 4$ for indirect transitions, E_g is the energy gap and $h\nu$ is the incident photon energy. Plots of the average values of $(\alpha h\nu)^2$ versus the photon energy ($h\nu$) of Sb_2Te_3 films of different thicknesses are shown in Fig. 5. The straight line ($n = 1$) which can be seen in the photon energy range 0.8–1.5 eV indicates that Sb_2Te_3 films possess a direct band-gap. When the linear portion of these plots is extrapolated to $\alpha = 0$, the direct energy gaps E_g for thicknesses of 100, 150 and 200 nm are found to be 0.29, 0.22 and 0.14 eV, respectively. Thus, the energy gap decreases as the thickness is increased. This may be due to improvement in grain size and marked variation in the absorption.

Clark [17] has reported a variation of the optical properties of polycrystalline thin films with the grain size. The observed values of optical energy gaps are slightly lower in comparison to the energy gap = 0.3 eV reported by Black *et al.* [18] and Tich'y *et al.* [19] for bulk Sb_2Te_3 crystal.

4. Conclusion

Significant textural changes are not observed with the changes in the film thickness. The electrical resistivity of Sb_2Te_3 decreases with increasing film thickness due to an increase in the grain size and a decrease in the intercrystalline barrier. The dependence of mobility and carrier concentration on the thickness of film may be due to the improvement in the grain size and the mean free path of the carriers. The observed values of the activation energy and the optical energy gap are close to that of reported value. Thus the change in the electrical and optical properties of Sb_2Te_3 films with varying thickness have been attributed mainly to the change in the structural perfection.

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References
1. N. SAKAI, T. KAJIWARA, K. TAKEMURA, S. MINOMURA and Y. FUJI, *Solid State Commun.* **40** (1981) 1045.
2. K. WATANABE, N. SATO and S. MIYAOKA, *J. Appl. Phys.* **54** (1983) 1256.
3. K. YOKOTA and S. KATAYAMA, *Tech. Rep. Kansai Univ. Jpn no.* 16 (1975) 65.
4. H. J. GOLDSMID, J. E. GIUTRONICH and M M. KAILA, *Solar Energy* **24** (1980) 435.
5. B. ROY, B. R. CHAKRABORTY, R. BATTACHARYA and A. K. DATTA, *Solid State Commun.* **25** (1978) 937.
6. K. WOODBRIDGE and M. G. ERTL, *Phys. Status. Solidi (a)* **44** (1977) K 123.
7. H. T. LANGHAMMER, M. STORDEUR, H. SOBOTTA and V. RIEDE, *Phys. Status Solidi (b)* **109** (1982) 673.
8. N. S. RAJAGOPALAN and S. K. GHOSH, *Physica* **29** (1963) 234.
9. T. C. PATEL and P. G. PATEL, *Mater. Lett.* **3** (1984) 46.
10. Powder Diffraction File, Card no. 15–874 (Joint Committee on Powder Diffraction Standards, PA, 1974).
11. E. H. SONDHEIMER, *Adv. Phys.* **1** (1952) 1.
12. B. ROY, B. R. CHAKRABORTY, R. BHATTACHAYA and A. K. DUTTA, *Solid State Commun.* **25** (1978) 617.
13. N. S. P. LUCAS, *J. Appl. Phys.* **36** (1961) 5.
14. S. NOGUCHI and H. SAKATA, *J. Phys. D* **13** (1980) 1129.
15. C. A. NEUGEBAUR, in "Physics of Thin Films", Vol. 2 edited by G. Hass and R. S. Thun (Academic Press, New York, 1969) p. 25.
16. E. J. JOHNSON, *Semiconductor Semimetals* **3** (1967) 153.
17. A. H. CLARK, in "Polycrystalline and Amorphous Thin Films and Devices", edited by L. L. Kazmerski (Academic Press, New York, 1980) p. 135.
18. J. BLACK, E. M. CONWELL, L. SEIGLE and C. W. SPENCER, *J. Phys. Chem. Solids* **2** (1957) 240.
19. L. TICH'Y, J. HORA'K, A. VĂSKO and M. FRUMAR, *Phys. Status Solidi (a)* **20** (1973) 725.

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