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Optical, electrical and thermoelectric power studies of Al–Sb thin film bilayer structure

M SINGH*, J S ARORA, Y K VIJAY and M SUDHARSHAN[†]

Department of Physics, University of Rajasthan, Jaipur 302 004, India

[†]IUC-DAE, Kolkata Centre, Bidhan Nagar, Kolkata 700 091, India

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Abstract. **The III–V semiconductors are of great importance due to their applications in various electro-optic devices. The Al–Sb thin film was deposited on glass substrate by thermal evaporation method at a pressure of 10^{-5} torr. The samples were annealed for 3 h at different constant temperatures in a vacuum chamber at a pressure of 10^{-5} torr. The electrical resistance vs temperature studies show phase transformation from metallic to semiconducting. The observed positive thermoelectric power indicates that Al–Sb thin films are *p*-type in nature. The Rutherford back scattering analysis and optical band gap measurements also indicate that the inter-diffusion concentration varies with temperature.**

Keywords. **Annealing; interdiffusion; semiconductor; bilayer; RBS.**

1. Introduction

The III–V semiconductors are of great importance due to their applications in various electro-optical devices (Yu *et al* 1995). The aluminum antimonide seems to be a promising semiconducting material for high temperature applications specially for transistors and P–N junction diodes, because of large band gap (Zielasek and Archh 1954; Herczog *et al* 1958). The semiconducting compound, aluminum antimonide (Al–Sb), with a energy gap of 1·62 eV, is potentially a high efficiency solar material (Rittner 1954). The Al–Sb has rapidly growing interest in opto-electronics (Lefebvre *et al* 1987; Raisin *et al* 1987). Several workers have studied electrical, thermal, optical and structural properties of stoichiometric Al–Sb in bulk as well as in thin film forms (Patrel and Birander 1983; Matsuo and Soma 1986, 1987; Nikam *et al* 1997; Bedi and Singh 1998). The flash evaporation technique has been employed to grow Al–Sb films (Richards *et al* 1964). The isochronal heat treatment of bilayer results in the inter diffusion and reaction between the elements, accompanied by the consequent nucleation and growth of phases. It was also observed for Al/Ge system with annealing the occurrence of extensive intermixing and asymmetric diffusion across the interface using SIMS technique (Raghavan *et al* 2000). Recently, Singh and Vijay (2004a, b) prepared semiconductor by using the bilayer structure of Zn–Se and In–Sb thin films. From the survey of literature, it can be seen that almost no attempt has been made to study the bilayer diffusion properties of Al–Sb thin films. In the present communication, RBS analysis, optical, electrical and thermoelectric power studies of Al–Sb bilayer structure of thin films prepared by thermal co-evaporation technique is reported.

2. Experimental

Aluminium antimonide films were prepared by thermal evaporation method using a vacuum coating unit at 10^{-5} torr pressure. The high purity aluminium (99·999%, 10 mg) thin foils and pure antimony powder (98·5%, 20 mg) obtained from the BDH Chemicals Ltd., Poole, England, were placed in two different boats in the Hind High Vacuum coating unit at a pressure of 10^{-5} torr. The glass substrate was placed in the substrate holder above the boats carrying materials. Aluminum was first evaporated and later antimony to get bilayer of Al–Sb thin films. The deposited films were annealed at 10^{-5} torr pressure for 3 h to inter-diffuse Al and Sb elements for different constant temperatures. Hitachi spectrophotometer model 330 was used to record transmission spectra of Al–Sb thin films at different temperatures. The electrical resistivity and thermoelectric power measurements were carried out by using the two-probe method.

The RBS analysis was carried out at the Institute of Physics, Bhubaneswar, to confirm compositional variation with temperature in bilayer and inter diffusion of elements, Al and Sb, at different temperatures. The thickness was measured by gravimetric method, $d = M/r*A$ cm, where A is surface area of the hemisphere of radius 10 cm, M the weight of material in boat, and r the density of material. The observed thickness of Al and Sb bilayer was 2356 Å and 1898 Å, respectively.

*Author for correspondence (mangej_singh@yahoo.com)

3. Results and discussion

3.1 *Sheet resistance of Al–Sb thin films*

The Al–Sb bilayer films have been deposited on glass substrate by thermal evaporation technique at a pressure of 10^{-5} torr. The isochronal annealing of bilayer thin film samples is carried out in vacuum chamber at 10^{-5} torr pressure at different temperatures for 3 h to study their inter diffusion process by electrical resistivity method. The measurement of resistance is an effective tool to understand the structural and compositional changes in a layered structure (Raghavan *et al* 2000). The resistance vs temperature variation is shown in figures 1(a) and (b) at temperatures 331 K and 351 K, respectively, which shows the phase transformation due to inter diffusion of bilayer with temperature. In figure 1(a), the sample was isochronal annealed at 331 K and then electrical resistance vs temperature studies were carried out, which shows that the increasing resistance is due to mixing of Al and Sb in each other. This change in resistance indicates possible formation of granularity in the system (Mgbenu 1980). The films initially show positive temperature coefficient of resistance (TCR) suggesting metallic behaviour up to temperature, 320 K and further increasing concentration of Al in Sb due to larger diffusion coefficient of Al in Sb shows negative TCR indicating semiconducting behaviour. The value of positive TCR to negative TCR shows the metallic to semiconducting transition with annealing temperature. The measurements of resistivity of the samples and its variation with temperature have been carried out for Al–Zn–Mg system and negative slope is explained on the basis of mechanism of hopping through the critical states and correlated with phase changes (Joshi *et al* 2002). In figure 1(b), the initial negative TCR shows the semiconducting behaviour up to 320 K and further increasing temperature shows the positive TCR, that sudden increase in resistance at 320 K is justified by our thermopower measurements and also supported increase in grain size and diffusion process with annealing temperature reported in case of Al/Si system by Mgbenu (1980). The 3 h duration of annealing allowed diffusion at such a low temperature, which was reflected by increase in resistance at a particular temperature. The resistance vs temperature graph well agrees with varying compositions of AlSb thin films, which were, metallic to semiconducting (Nikam *et al* 1997).

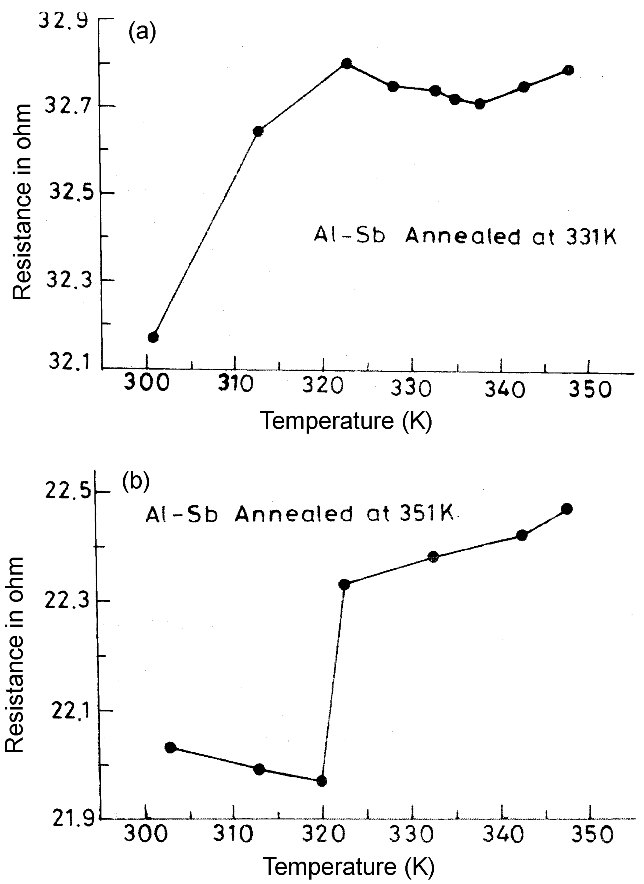

Figure 1. Temperature vs resistance of AlSb thin films annealed at (**a**) 331 K and (**b**) 351 K.

3.2 *Thermoelectric power*

The thermoelectric power measurements were carried out on annealed samples of AlSb, using variable temperature method. In this method, the temperature of one end varied and temperature of another end fixed and induced e.m.f. was observed. The thermoelectric power was calculated using the formula given below

$$S(t) = \Delta V/\Delta T,$$

where ΔT is temperature gradient and ΔV thermo e.m.f. at absolute temperature. The positive sign of TEP suggests that conduction should occur predominantly due to holes, which established the *p*-nature of the AlSb thin films, which agreed with Hall effect measurement observed by Bedi and Singh (1998). According to them, Al–Sb films are *p*-type, which indicates that conduction is primarily due to holes. The temperature dependence of thermoelectric power (S) of the films is shown in figure 2 for annealed samples at two different temperatures. These graphs show that thermoelectric power increases with temperature. The variation of TEP with temperature in our samples may be explained on the basis of defect state model. It may be considered that during TEP measurements the thermal gradients established, changes the density of charged defect state by capturing electrons and holes. The motion of electrons and holes can take place through the process of diffusion. In this case the density of the holes giving TEP is greater than that of electrons and the *p*-type nature established, with increasing temperature the density of holes increased and so TEP also increased.

The hole concentration is (Neugebauer *et al* 1996) given by the relation

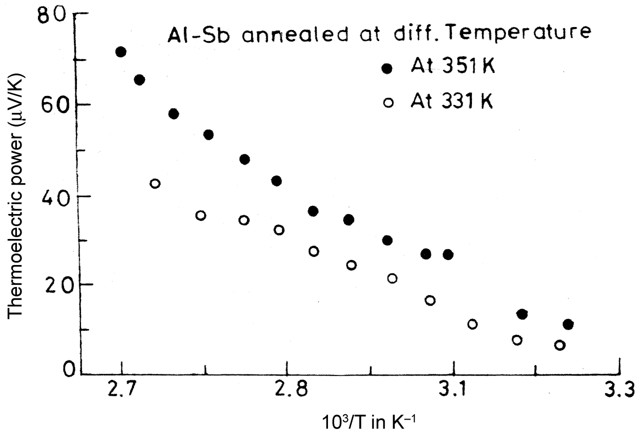

Figure 2. Variation of thermoelectric power vs $10^3/T$ at different temperatures.

$$N_{hole} = N_{acceptors}\exp(-E_a/k_BT),$$

where $N_{acceptors}$ is the number of acceptors, E_a the acceptor ionization energy, k_B the Boltzmann constant and T the device temperature. The hole concentration can, therefore, be increased by (i) increasing the acceptor concentration, (ii) increasing the device temperature and (iii) finding new acceptors with a lower ionization energy.

It was established by SIMS studies that occurrence of extensive intermixing and asymmetric diffusion across the interface and enhanced diffusion of one of the components in Al/Ge bilayer film across the interface can leave behind a large vacancy concentration reported by Raghavan *et al* (2000). In our case this type of vacancy is responsible for increase of positive sign of TEP with temperature. The variation of thermoelectric power at different temperatures may also indicate the phase transformation during the inter diffusion of bilayers. The $10^3/T$ vs temperature plots also indicate that thermoelectric power increases linearly with increasing temperature, which agrees with the behaviour expected of a typical non-degenerate semiconductor observed by Agrawal *et al* (1994).

3.3 *Optical properties*

Transmission spectra of Al–Sb thin film is taken at room temperature with the help of Hitachi spectra photometer model 330. Energy band gap of the films was calculated with the help of transmission spectra, using Tauc (1974) relation

$$ahn = A(hn - E_g)^n,$$

where hn is photon energy, a the absorption coefficient, E_g the band gap, A the constant is 1/2 for direct band gap material and n is 2 for indirect band gap material. Since our material was indirect band gap, the extrapolation of straight line to $(ahn)^{1/2} = 0$ axis gives the value of energy of band gap. Figure 3 shows spectral variation for Al–Sb thin films deposited on the glass substrate extrapolating the lines to $(ahn)^{1/2} = 0$ and it gives the values of optical gap due to indirect transition. In annealed films the optical absorption edge is produced by transitions between two state densities, both of which rapidly change as function of energy and which correspond to the valence and conduction band edge for the ideal stable continuous random network given by Connell and Willian (1972). The band gap observed for these films shows the large variation with annealing temperature. It may be due to the variation in inter diffusion concentration ratio of Al and Sb at interface. The exact concentration of composition requires depth profile studies like SIMS, however, we report in this paper RBS studies to confirm mixing of Al and Sb with annealing temperature. Our optical band gap, 1·45 eV, for isochronal annealed samples for 3 h in vacuum chamber well agreed with the reported band gap which varied from 1·42–1·52 eV (Bedi and Singh 1998) and also for compound, Al–Sb (Rittner 1954) with energy gap, 1·62 eV, for solar cell applications.

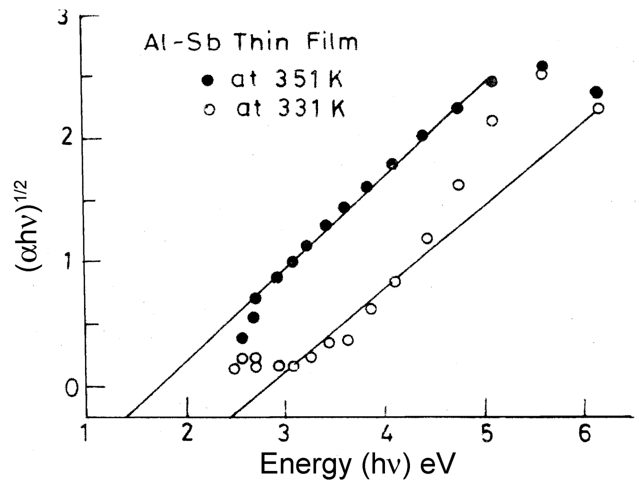

Figure 3. Photon energy (hn) vs $(ahn)^{1/2}$ for indirect band gap material of AlSb thin film at different annealing temperatures.

3.4 *Rutherford back scattering spectrometry*

Rutherford back scattering spectrometry assisted by channeling facility is being successfully employed for quantitative information about stoichiometry and structure of interfaces doped semiconductor as a function of depth. We have observed in figure 4 count vs channel graph of AlSb peaks which shifted with temperature in the downward count direction which shows that the concentration of Al enhanced in Sb or one can say that Al and Sb inter diffuse with increasing temperature. The graph for as deposited films shows separate peaks of aluminium at 230 and antimony at channel number 665, but with increasing temperature aluminium and antimony peaks shifted towards each other in mid region, which confirm the formation of aluminium-antimonide by bilayer structure.

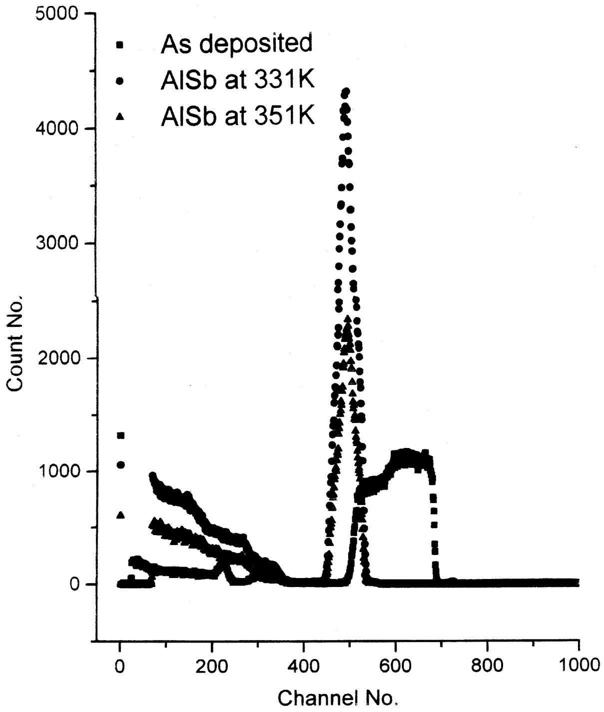

Figure 4. Rutherford back scattering energy of 3 MeV *a* on channel vs count number.

If we compare the graph of as deposited films with different temperature annealed films, then we can find that the peak shifted downwards. It means inter diffusion occurs between the Al/Sb bilayer and the concentration of Al enhanced due to higher diffusion coefficient of aluminium than antimony. Thus we conclude that in AlSb interface is made by inter diffusion of Al and Sb elements to each other. The downwards peak to valley ratio is also observed (Holloway *et al* 1987) in case of Ti–Si layer structure. They had also observed the microstructural changes in multilayered Ti–Si during the thermal annealing by using RBS and XRD techniques. In case of Zn–Se and In–Sb, bilayer structure of thin films also confirm the intermixing using RBS measurements (Singh and Vijay 2004a, b).

4. Conclusions

(I) The resistance variation can be used to understand phase transformation in bilayer structure.
(II) The positive sign of TEP confirms the *p*-type nature of aluminium antimonide.
(III) The band gap measurement confirms the variation of elemental concentration in the bilayer interface with temperature.
(IV) Rutherford back scattering analysis indicates the elemental concentration variation in interface with annealing temperature.

Acknowledgements

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References

Agrawal A, Patel P D and Lakshminarayana D 1994 *J. Cryst. Growth* **142** 344
Bedi R K and Singh T 1998 *Semiconductor materials* (ed.) R K Bedi (Amritsar: Guru Nanak Dev University) p. 189
Connell G A N and Willian P 1992 *J. Non-Cryst. Solids* **8** 215
Herczog A, Haberecht R R and Middleton A E 1958 *J. Electrochem. Soc*. **105** 533
Holloway K and Sinclair R 1987 *J. Appl. Phys*. **4** 61
Joshi S K, Mehta L and Deshpande A P 2002 *Indian J. Pure & Appl. Phys*. **40** 482
Lefebvre P, Gill B, Allegre J, Mathieu H, Chen Y and Raisin C 1987 *Phys. Rev*. **B35** 1230
Matsuo K H and Soma T 1986 *Phys. Status Solidi* **(b)K13** 138
Matsuo K H and Soma T 1987 *Solid State Commun*. **62** 760
Mgbenu F N 1980 *Thin Solid Films* **65** 267
Neugebauer J and Chris van de Walle G 1996 *The physics of semiconductors* (London: World Scientific) **4** p. 2849
Nikam P S, Borse R Y and Pawar R R 1997 *Bull. Mater. Sci*. **20** 1015
Patrel S M and Birander A M 1983 *Indian J. Phys. & Appl. Phys.* **21** 418
Raghavan G, Venugopal Rao G, Amarendra G, Tyagi A K and Viswanathan B 2000 *Appl. Surf. Sci*. **7066** 1
Raisin C, Lassabatere L, Iibert A, Birault B, Abdel G and Voisin P 1987 *Solid State Commun*. **61** 17
Richards J L, Hart P B and Gallone L M 1963 *J. Appl. Phys.* **37** 3418
Richards J L, Hart P B and Muller E K 1964 *Single crystal film* (ed.) M H Francombe (New York: Reinhold Publishing Corp.).
Rittner E S 1954 *Phys. Rev*. **96** 1708
Singh M and Vijay Y K 2004a *Appl. Surf. Sci*. **239** 79
Singh M and Vijay Y K 2004b *Indian J. Pure & Appl. Phys*. **42** 610
Tauc J (ed.) 1974 *Amorphous and liquid semiconductor* (New York: Plenum) p. 159
Yu K M, Moll A J, Chan N, Walukiewicz W and Becla P 1995 *Appl. Phys. Lett*. **66** 2406
Zielasek G and Archh E 1954 *Ubertrag* **8** 529