

Effect of Sn incorporation in the density of defect states in a-Se thin film

N. SHARMA, S. P. SINGH, S. KUMAR*

Department of Physics, Christ Church College, Kanpur-208001, India

The present paper reports the d. c. conductivity measurements at high electric fields in vacuum evaporated amorphous thin films of pure Se and $\text{Se}_{90}\text{Sn}_{10}$ glassy alloys. Current-Voltage (I-V) Characteristics have been measured at various fixed temperatures. In these samples, at low electric fields, ohmic behavior is observed. However, at high electric fields ($E \sim 10^4 \text{V/cm}$), non-ohmic behavior is observed. An analysis of the experimental data confirms the presence of space charge limited conduction (SCLC) in the glassy materials studied in the present case. From the fitting of the data to the theory of SCLC, the density of defect states (DOS) near Fermi level is calculated. The increase in DOS near Fermi level has been found which could be explained in terms of the electronegativity differences between the aforesaid two elements.

(Received April 30, 2007; accepted June 27, 2007)

Keywords: Thin films, Chalcogenide glasses, SCLC, DOS

1. Introduction

Amorphous Selenium has emerged as a promising material because of its potential technological importance. It is widely preferred in the fabrication of electro photographic devices and more recently switching and memory devices [1]. It has been found that Se based alloys are useful due to greater hardness, high photosensitivity, higher crystallization temperature and smaller ageing effect compared to pure a-Se [2]. The transport mechanism of charge carriers in amorphous semiconductors has been the subject of intensive theoretical and experimental investigations for the last few years. These studies have been stimulated by the attractive possibilities of using the structure disorder in amorphous semiconductors for the development of better, cheaper and more reliable solid state devices [3,4].

Due to their low conductivity, amorphous semiconductors are most suitable for high field conduction studies, as the Joule heating is negligibly small in these materials at moderate temperatures. Some such studies have been reported in chalcogenide glassy semiconductors [5-15] and the results have been interpreted in terms of space charge limited conduction or Poole-Frenkel conduction. One of the most direct methods for the determination of the density of the localised states g_0 in the mobility gap involves the measurements of SCLC, which can easily be observed at high fields in chalcogenide materials. Such a technique has already been applied to a-Si:H [16-19]. SCLC technique is not influenced by surface states; unlike field effect experiments where surface states may come into play.

The present paper reports the SCLC measurements in a-Se and a- $\text{Se}_{90}\text{Sn}_{10}$ glassy system. Using the theory of SCLC, for the case of uniform distribution of localised states, the density of localised states near Fermi level is calculated for the present system.

2. Experimental

Glassy alloys of Se and $\text{Se}_{90}\text{Sn}_{10}$ were prepared by quenching technique. High purity (99.999 %) materials were weighed according to their atomic percentages and were sealed in quartz ampoules (length ~ 5 cm and internal dia ~ 8 mm) with a vacuum $\sim 10^{-5}$ Torr. The ampoules containing the materials were heated to 900°C and held at that temperature for 10 - 12 hours. The temperature of the furnace was raised slowly at a rate $\sim 3 - 4^\circ\text{C/min}$. During heating, all the ampoules were constantly rocked, by rotating a ceramic rod to which the ampoules are tucked away in the furnace. This was done to obtain homogenous glassy alloys.

After rocking for about 10 hours, the obtained melts were cooled rapidly by removing the ampoules from the furnace and dropping to ice-cooled water. The quenched samples were taken out by breaking the quartz ampoules. The glassy nature of the materials was checked by XRD technique.

Thin films of these glasses were prepared by vacuum evaporation technique keeping glass substrates at room temperature. Vacuum evaporated indium electrodes at bottom were used for the electrical contact. The thickness of the films was ~ 500 nm. The co-planar structure (length ~ 1.2 cm and electrode separation ~ 0.12 mm) was used for the present measurements. The films were kept in deposition chamber in the dark for 24 hours before mounting them in the sample holder. This was done to allow sufficient annealing at room temperature so that a metastable thermodynamic equilibrium may be attained in the samples as suggested by Abkowitz [20] for chalcogenide glasses. The deposition parameters were kept almost the same for all the samples so that a comparison of results could be made for the various glassy samples. The amorphous nature of thin films was ascertained by X-ray diffraction.

Table 1. Density of localized states (g_0) in a-Se and $Se_{90}Sn_{10}$ glassy alloys.

Samples	Slope of S vs $1000/T$ curve	ϵ_r (120 Hz, 303K)	g_0 (density of localized states in $eV^{-1} cm^{-3}$)
a-Se	6.5×10^{-3}	3.51	4.81×10^{13}
a- $Se_{90}Sn_{10}$	9.7×10^{-4}	4.68	4.29×10^{14}

For the measurements of high field conduction, thin film samples were mounted in a specially designed sample holder. A vacuum of $\sim 10^{-3}$ Torr was maintained throughout the measurements. A d.c. voltage (0 to 300 V) was applied across the sample and the resultant current was measured by a digital Pico-Ammeter. I – V characteristics were measured at various fixed temperatures (285–323 K) in these films. The temperature of the films was controlled by mounting a heater inside the sample holder and measured by a calibrated copper-constantan thermocouple mounted in the vicinity of the films. Before measuring I – V characteristics, thin films were annealed in a vacuum $\sim 10^{-3}$ Torr near glass transition temperature for two hours in the same sample holder that was used for the above measurements.

3. Results and discussion

In the present work, I-V characteristics of thin films of a-Se and $Se_{90}Sn_{10}$ were examined at various temperatures (285 K–323 K). At low fields ($<10^3$ V/cm), an ohmic behaviour is observed in all the samples. However, at higher fields ($\sim 10^4$ V/cm), a superohmic behaviour is observed at all the measuring temperatures.

According to the theory of space charge limited conduction, in the case of a uniform distribution of localized states $g(E) = g_0$, the current (I) at a particular voltage (V) is given by the following relation [21]

$$I = (eA\mu n_0 V/d) \exp(SV) \quad (1)$$

Where d is the electrode spacing, n_0 is the density of the thermally generated charge carriers, μ is the mobility, e is the electronic charge, A is the area of cross section of thin films and S is given by

$$S = 2\epsilon_r \epsilon_0 / e g_0 k T d^2 \quad (2)$$

As evident from Eqn. (1) and (2), in case of space charge limited conduction, the $\ln I/V$ vs V curves should be a straight line and slope (S) of these curves should decrease linearly with the increase of temperature.

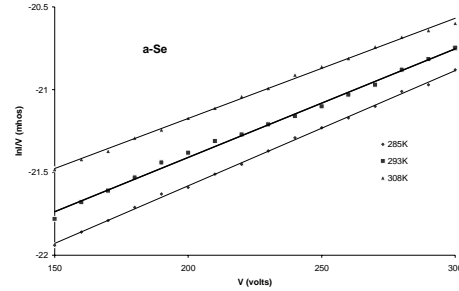


Fig. 1. Plots of $\ln I/V$ vs. V curve for a-Se at different temperatures.

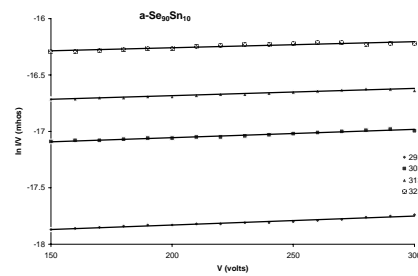


Fig. 2. Plots of $\ln I/V$ vs. V curve for a- $Se_{90}Sn_{10}$ at different temperatures.

In the present case, at higher fields, $\ln(I/V)$ Vs V curves are found to be straight lines with high correlation coefficient at all the measuring temperatures in all the samples. Such curves for amorphous thin films of Se and $Se_{90}Sn_{10}$ are plotted in Figs. 1 and 2. The slope (S) of these curves decreases linearly with temperature for all the samples (see Fig. 3 and 4). These results indicate the presence of space charge limited conduction in all the samples used. Thin films contain a large number of defects due to dangling bonds that give rise to large number of localised defect states. These localised states act as carrier trapping centers and after trapping the injected charge from the respective electrodes, they become charged and thereby expected to build up a space charge. This build up of space charge then play the key role in the determination of SCLC process.

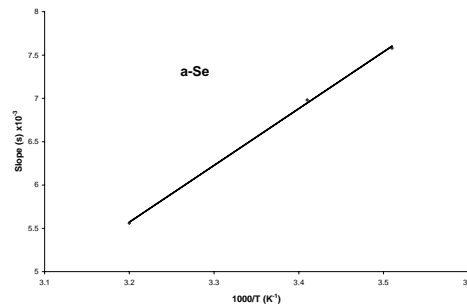


Fig. 3. Plots of S vs. $1000/T$ curve for a-Se glassy system.

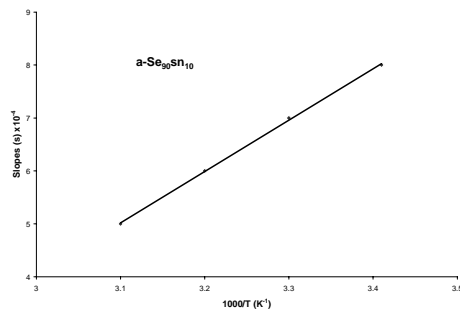


Fig. 4. Plots of S vs. $1000/T$ curve for a - $Se_{90}Sn_{10}$ glassy system.

Using equation 2, we have calculated the density of localised states from the slopes of Figs. 3 and 4. The value of the relative dielectric constant ϵ_r are measured by using capacitance measuring assembly model GR 1620 AP, employing the three terminal technique. The results of these calculations are given in Table 1. It is clear that g_0 increases with the incorporation of Sn in a-Se. The reason for this increase could be understood from the followings:

When isoelectronic atom Te is added to amorphous Selenium, the density of defect states is increased and hence the residual potential increases in xerographic experiment. Onozuka et al. [22] observed that, on introducing Cl to Se-Te system, the residual potential is decreased again. This result was interpreted on the basis of a structural defect model where Te was assumed to form positively charged impurities due to small electronegativity of Te as compared to Se [23], while Cl atoms having higher electro-negativity than Selenium form negatively charged impurities, thereby compensating the effect of Te.

Along the same lines, one can expect that when Sn having lower electro-negativity than Se [23] is introduced, positively charged defects will be created thus increasing the density of defect states in binary Se-Sn system as compared to pure Se.

4. Conclusions

I-V characteristics have been studied in amorphous thin films of Se and $Se_{90}Sn_{10}$. At low fields ($<10^3$ V/cm), an ohmic behaviour is observed. However, at high fields ($\sim 10^4$ V/cm), a super ohmic behaviour is observed

The density of localised states near Fermi level is calculated by fitting the data to the theory of SCLC in case of a uniform distribution of localised states. The incorporation of Sn in a-Se results in an increase in the density of localised states around the Fermi level which could be explained in terms of the electronegativity differences between the aforesaid two elements.

Acknowledgement

Work supported by University Grants Commission, New Delhi.

References

- [1] K. Homma, H. K. Henish, S. R. Ovshinsky, *J. Non-Cryst. Solids* **35-36**, 1105 (1980).
- [2] J. Y. Shim, S.W. Park, H. K. Baik, *Thin solid Films* **292**, 31 (1997).
- [3] S. Kumar, R. Arora, A. Kumar *Physica B* **183**, 172 (1993).
- [4] M. Tabak, S. W. Ing, M. E. Scharfe, *IEEE Trans. Electron Devices* **10**, 91 (1973).
- [5] J.E. Hall *J. Non-Cryst. Solids* **2**, 125 (1970).
- [6] A. Kumar, S. Kumar, R. Arora, *Solid State Commun.* **78**, 65 (1991),
- [7] S. Kumar, R. Arora, A. Kumar, *J. Mater. Sci. Lett.* **10**, 1280 (1991),
- [8] B. T. Kolomiets, E. Lebedev, *Fiz. Tekh. Poluprov. (Russian)* **1**, 815 (1967).
- [9] R. M. Mehra, H. Kumar, S. Koul, P. C. Mathur, *Phys. Stat. Sol. (a)* **83**, 341 (1984).
- [10] D. S. Misra, A. Kumar, S. C. Agrawal, *J. Non-Cryst. Solids* **76**, 215 (1985).
- [11] K. D. McKenzie, P. G. Lecomber, W. E. Spear, *Philos. Mag.* **46**, 377 (1982).
- [12] W. Den Boer, *J. Phys. (Paris)* **42(C4)**, 451 (1981).
- [13] E. Bhattacharya, S. Guha, K.V. Krishna, D.R. Bapat, *J. Appl. Phys.* **53**, 6285 (1982).
- [14] P. S. Nikam, H. S. Aher, *Ind. J. Pure Appl. Phys.* **34**, 393 (1996).
- [15] S. P. Singh, S. Kumar, A. Kumar, *J. Mat. Sci.* **39**, 1 (2004).
- [16] M. Meaudre, R. Meaudre, *Phil. Mag. B* **55**, 417 (1987).
- [17] E. V. Grekov, O. G. Sukhorukov, *Sov. Phys. Semicon.* **22**, 457 (1988).
- [18] I. Soloman, R. Benferhat, H. Tran-Quoc, *Phys. Rev. B* **30**, 3422 (1984).
- [19] S. Okano, M. Suzuki, K. Imura, A. Fukada, J. Hiraki, *J. Non-Cryst. Sol.* **59-60** 969 (1983).
- [20] M. Abkowitz, *Polym. Eng. Sci.* **24**, 1149 (1984).
- [21] M.A. Lampert, M. Park, *Current Injection in Solids*, Acad. Press, New York, (1970).
- [22] A. Onozuka, O. Oda, I. Tsuboya, *Thin Solid films* **149**, 9 (1987).
- [23] L. Pauling, *The Nature of the chemical bond* (Calcutta: Oxford and IBH) **P 93** (1969).

*Corresponding author: dr_santosh_kr@yahoo.com