JOURNAL OF MATERIALS SCIENCE 40 (2005) 481 – 484

Space charge limited conduction in a-$Se_{75}In_{25-x}Pb_x$ thin films*

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The present paper reports the d.c. conductivity measurements at high electric fields in vacuum evaporated thin films of amorphous $Se_{75}In_{25-x}Pb_x$ ($0 < x < 10$). Current-voltage (I-V) Characteristics have been measured at various fixed temperatures. In all the samples, at low electric fields, ohmic behavior is observed. However, at high electric fields ($E \sim 10^4$ V/cm), non-ohmic behavior is observed. In case of sample having 4 at.% of Pb, the experimental data fits well with the theory of space charge limited conduction (SCLC). From the fitting of the data, the density of defect states near Fermi level is calculated. Such type of behavior is not observed at higher concentration of Pb in the present glassy system.

1. Introduction

Because of their potential applications, thin films of chalcogenide glasses have been extensively studied during the past few years. Attempts have been made to produce stable glasses which have good photosensitive properties and can be doped *n* or *p*-type, so that they may be used in various solid state devices [1–3]. It is widely accepted that the addition of a third element to the binary chacogenide glassy alloys produces stability in these glasses. In Ge-Se and Se-In systems, some metallic additives have been found [4–9] to change conduction from *p*-type to *n*-type and hence these binary systems are of great importance.

High field effects are most readily observed in these materials because of their low conductivity, and have been studied by various groups working in this field [10–17]. The result of these workers have been interpreted in terms of heating effect, space charge limited conduction (SCLC) and high field conduction due to the Poole-Frenkel effect. This indicates that the interpretation of the high field data is highly intriguing in these materials and much has to be done in this field. In the present study, we have measured the high field conduction in various glassy alloy of the $Se_{75}In_{25-x}Pb_x$. The results indicate that at low fields, ohmic behavior is observed. However, at higher fields, non-ohmic behavior is observed.

The next section describes the experimental details of the measurements. The results are presented and discussed in the third section. The final section deals with the conclusions drawn from the present work.

2. Experimental

Glassy alloy of $Se_{75}In_{25-x}Pb_x$ ($x = 0, 4, 6, 10$) is prepared by quenching technique. High purity (99.999%) materials are weighed according to their atomic percentages and are sealed in quartz ampoules (length ∼5 cm and internal dia ∼8 mm) with a vacuum $\sim 10^{-5}$ Torr. The ampoules containing the materials are heated to 1000°C and held at that temperature for 10–12 h. The temperature of the furnace is raised slowly at a rate of 3–4°C/min. During heating, all the ampoules are constantly rocked, by rotating a ceramic rod to which the ampoules are tucked away in the furnace. This is done to obtain homogenous glass alloys.

After rocking for about 10 h, the obtained melts are cooled rapidly by removing the ampoules from the furnace and dropping to ice-cooled water. The quenched samples of $Se_{75}In_{25-x}Pb_x$ are taken out by breaking the quartz ampoules. The amorphous nature of the samples was confirmed by the absence of any sharp peak in the X-ray diffraction pattern. Compositional analysis was performed using electron probe micro analysis (EPMA).

Thin films of these glasses are prepared by vacuum evaporation technique keeping glass substrates at room temperature. Vacuum evaporated indium electrodes at bottom are used for the electrical contact. The thickness of the films is ∼500 nm. The co-planar structure (length ∼1.2 cm and electrode separation ∼0.12 mm) are used for the present measurements. A vacuum $\sim 10^{-2}$ Torr is maintained in the entire temperature range (297 K to 327 K).

Before measuring the d.c. conductivity, the films are first annealed at 340 K for one hour in a vacuum $\sim 10^{-2}$ Torr. I-V characteristics are found to be linear

*Work supported by University Grants Commission, New Delhi.

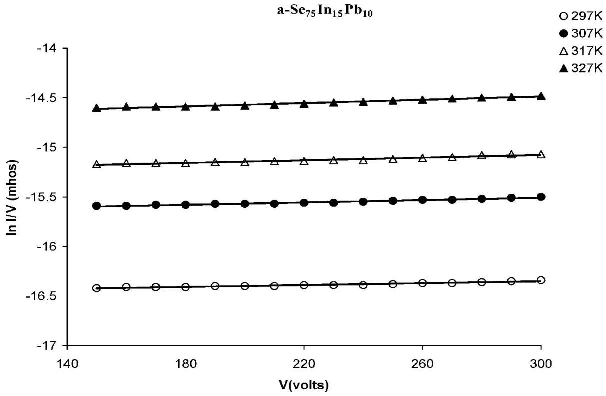

Figure 1 Plots of ln I/V vs. V curves for a-$Se_{75}In_{15}Pb_{10}$ at different temperatures.

and symmetric up to 10 V. The present measurements are, however, made by applying a voltage from 10 to 300 V across the films. The resulting current is measured by a digital Pico-Ammeter. The heating rate is kept quite small (0.5 K/min) for these measurements. Thin films samples are mounted in a specially designed sample holder. A vacuum $\sim 10^{-2}$ Torr is maintained throughout the measurements. The temperature of the films is controlled by mounting a heater inside the sample holder, and measured by a calibrated copper-constantan thermocouple mounted very near to the films.

3. Results and discussion

Results of I-V characteristics at different temperature shows that in all the glassy samples studied here, ohmic behaviour is observed at low voltages, i.e., upto 10 V. However, at higher voltages ($E \sim 10^4$ V/cm), a super ohmic behaviour is observed in all the samples. Here, ln I/V vs. V curves are found to be straight lines in all the samples. Figs 1 and 2 show such curves in case of a-$Se_{75}In_{15}Pb_{10}$ and a-$Se_{75}In_{21}Pb_4$ respectively. Similar curves were found in other glassy samples studied here (results not shown). According to the theory of SCLC, in the case of an uniform distribution of localized states having density g_0, the current (I) at a particular voltage (V) is given by [18]:

$$I = 2eA\mu n_0 V/d[\exp(SV)] \tag{1}$$

Here, e is the electronic charge, A is the cross sectional area of the film, n_o is the density of free charge carriers, d is the electrode spacing and S is given by:

$$S = 2\varepsilon_r \varepsilon_0 / e g_0 k T d^2 \tag{2}$$

Where ε_r is the static value of the dielectric constant, ε_0 is the permittivity of free space and k is Boltzmann's constant. It should be noted that Equation 1 is not an exact solution of SCLC equation, but is a very good approximation of the one carrier space charge limited current under the condition of a uniform distribution of traps.

According to Equation 1, ln I/V vs. V curves should be a straight lines whose slope should decrease with increase in temperature as evident from Equation 2.

In the present case, ln I/V vs. V curves are straight lines in all the materials studied. However, the slopes of these curves decreases with increase in temperature only in case a-$Se_{75}In_{21}Pb_4$. In other samples, slope increases with increase in temperature (see Fig. 1 for example). This indicates that SCLC mechanism is not predominant in these alloys. The reasons for such type of different behavior can not be ascertained from the present measurements alone. There is a scope for doing further work on this problem to get a clear explanation of the observed behavior.

In case of a-$Se_{75}In_{21}Pb_4$, slopes follow the trend as required by Equation 2. This shows that the theory of SCLC is applicable in this case. It may be mentioned here that nearly linear plots of ln I/V vs. V as well as a linear decrease of S with temperature can also be explained in terms of high field conduction due to the Poole—Frenkel effect of screened charged intrinsic defects and field-induced lowering of energy barriers for the charge-carrier hopping within localized states at the band edges. However, in the case of field-dependent conductivity, the plot of ln I/V vs. V must be independent of the electrode spacing 'd'. On the other hand for any SCLC mechanism, the same plot gives different curves for different values of 'd'. We have therefore measured I-V characteristics at room temperature (297 K) for a-$Se_{75}In_{21}Pb_4$ sample having different electrode spacings (Fig. 4). It is clear from this figure that different slopes are obtained at different electrode

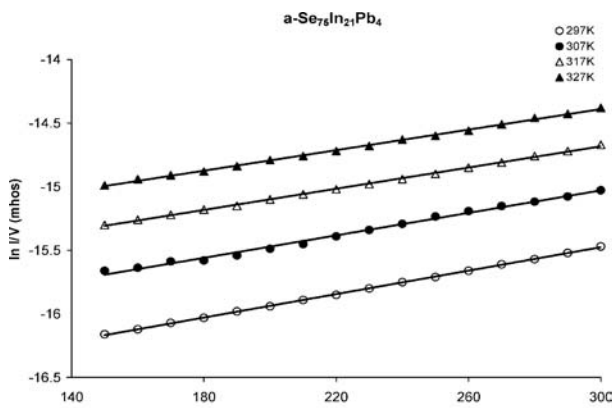

Figure 2 Plots of ln I/V vs. V curves for a-$Se_{75}In_{21}Pb_4$ at different temperatures.

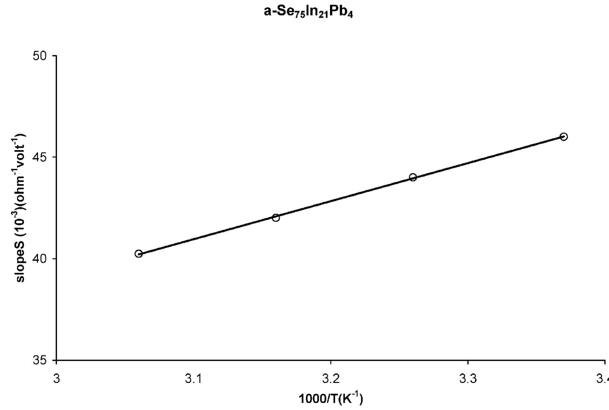

Figure 3 Plots of slope S (of Fig. 2) vs. 1000/T for a-$Se_{75}In_{21}Pb_4$ glassy system.

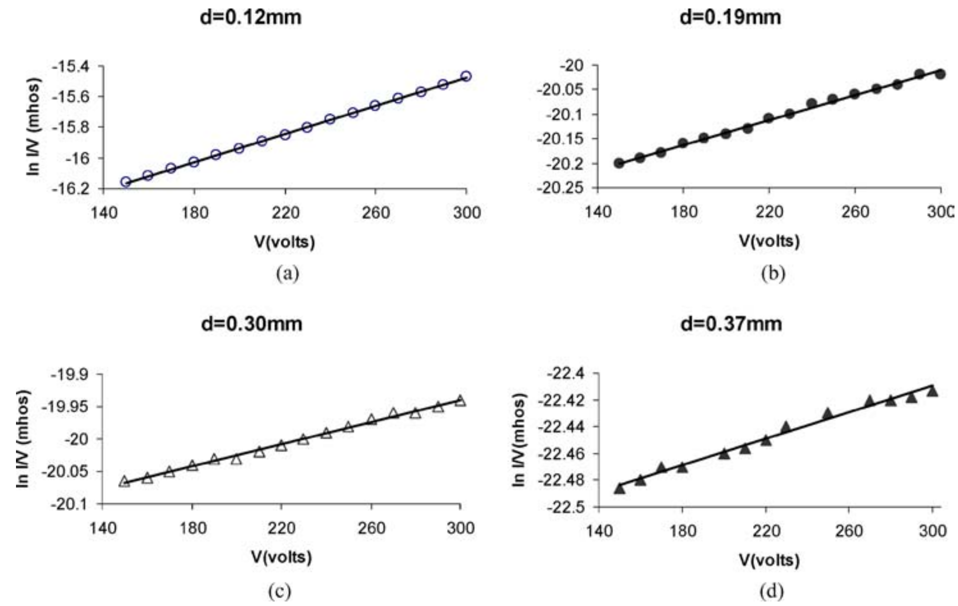

Figure 4 (a)–(d) Plots of ln I/V vs. V curves at room temperatures having different electrode gaps for a-$Se_{75}In_{21}Pb_4$.

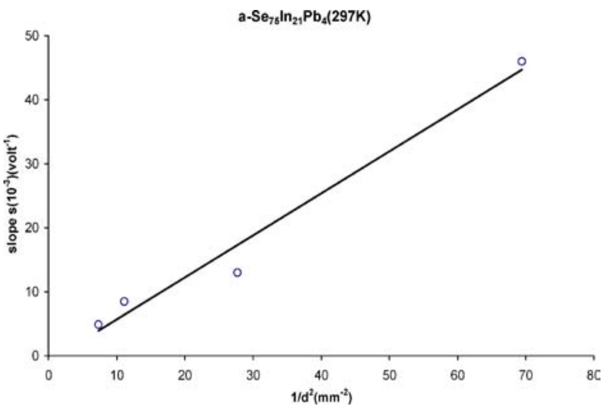

Figure 5 Plots of slope s (of Fig. 4) vs. $1/d^2$ curve for different electrode gaps for a-$Se_{75}In_{21}Pb_4$.

spacing. The values of these slopes are given in Table I and are plotted against $1/d^2$ in Fig. 5. This confirms the validity of Equation 2 in the present case and excludes the possibility of any other high-field conduction processes mentioned above. Hence the present measurements confirm the presence of SCLC in a-$Se_{75}In_{21}Pb_4$ sample.

Using Equation 2, we have calculated the density of localized states from the slope of Fig. 3. The value of the relative dielectric constant ε_r is measured by using capacitance measuring assembly model G R 1620 A P, employing the three terminal technique. The value of density of localized states comes out to be 5×10^{13} eV^{-1} cm^{-3}.

TABLE I Values of slopes of ln I/V vs. V curves for different electrode gaps

Electrode gap (d) (mm)	$1/d^2$ (mm^{-2})	Slopes (s) of ln I/V vs. V at T_{room} (297 K)
0.12	69.44	4.6×10^{-3}
0.19	27.70	1.3×10^{-3}
0.30	11.11	8.5×10^{-4}
0.37	7.31	4.9×10^{-4}

It is clear from the present measurement that the behavior of high field conduction is different at 4 at.% of Pb as compared to other composition. This is consistent with the earlier reported work where p to n transition has been observed at 5 at.% of Pb [19, 20].

4. Conclusion

I-V characteristics have been studied in amorphous thin films of a-$Se_{75}In_{25-x}Pb_x$ ($0 < x < 10$). At low fields, ohmic behaviour is observed. However, at higher fields ($\sim 10^4$ V/cm) superohmic behaviour is observed.

Analysis of the observed data shows the existence of SCLC in the sample where Pb is 4 at.% . From the fitting of the data to the SCLC theory, the density of localized states near Fermi-level is calculated. The different behavior at 4 at.% of Pb is consistent with a transition from p to n near to this composition as reported by other workers.

Acknowledgement

S. Kumar is grateful to U.G.C., New Delhi for providing a major research project during the course of this work.

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Received 8 April 2002
and accepted 12 August 2004