Journal of Optoelectronics and Advanced Materials Vol. 6, No. 4, December 2004, p. 1153 - 1157

SPACE CHARGE LIMITED CONDUCTION IN a-Se_{90} Ge_{10-X} In_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 $Se_{90}Ge_{10-x}In_x$ (x=0,4,6,10). Current-voltage (I-V) characteristics have been measured at various fixed temperatures. In all the samples, at low electric fields, ohmic behaviour is observed. However, at high electric fields (E~10^4 V/cm), non-ohmic behaviour is observed. An analysis of the experimental data indicates that upto 4 at.% of In, in these materials, space charge limited conduction (SCLC) is observed and beyond it heating effects are dominant due to which SCLC is not found. From the fitting of the data, the density of defect states (DOS) near Fermi level is calculated in the first two samples. The different behaviour of In incorporation with its lower and higher concentration is attributed with the change in structure of Ge-Se-In glassy network.

(Received July 13, 2004; accepted November 29, 2004)

Keywords: Thin films, Space charge limited conduction, Amorphous Se-Ge-In

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 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 chalcogenide 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 alloys of the Se_{90} Ge_{10-x} In_x, and found that up to 4 at.% of In, the experimental data fits well with the theory of SCLC. However, heating effect is dominant at higher concentration of In.

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 $Se_{90}Ge_{10-x}In_x$ (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

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* work supported by University Grants Commission, New Delhi

ampoules (length ~ 5 cm and internal dia ~ 8 mm) with a vacuum ~ 10^{-5} Torr. The ampoules containing the materials are heated to 1000°C and held at that temperature for 10 - 12 hours. 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 glassy alloys.

After rocking for about 10 hours, the obtained melts are cooled rapidly by removing the ampoules from the furnace and dropping to ice-cooled water. The quenched samples of $Se_{90}Ge_{10-x}In_x$ are taken out by breaking the quartz ampoules. The amorphous nature of the samples was confirmed by the absence of any sharp peaks 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 ~ 10^{-2} Torr is maintained in the entire temperature range (297- 337 K).

Before measuring the d. c. conductivity, the films are first annealed at 340 K for one hour in a vacuum ~ 10^{-2} Torr. I - V characteristics are found to be linear and symmetric up to 10 V. The present measurements are, however, made by applying a voltage from 10 V 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 ~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, up to 10 V. However, at higher voltages (E ~ 10^4 V/cm), a superohmic 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. 1-3 show such curves in case of a-$Se_{90}Ge_{10}$, $Se_{90}Ge_6In_4$ and $Se_{90}Ge_4In_6$ respectively. Similar curves were found in other glassy samples studied here (results not shown).

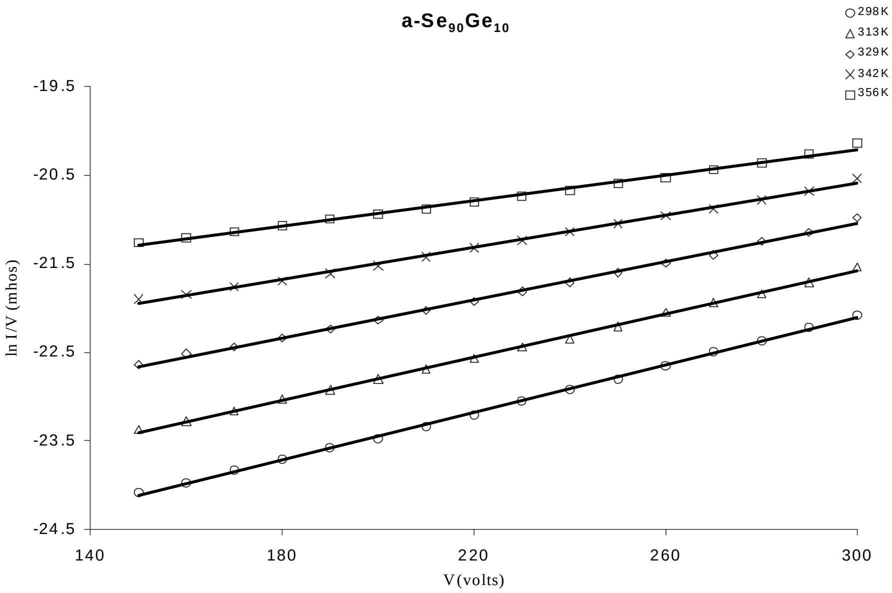

Fig. 1. Plots of ln(I/V) vs. V curves for a-$Se_{90}Ge_{10}$ at different temperatures.

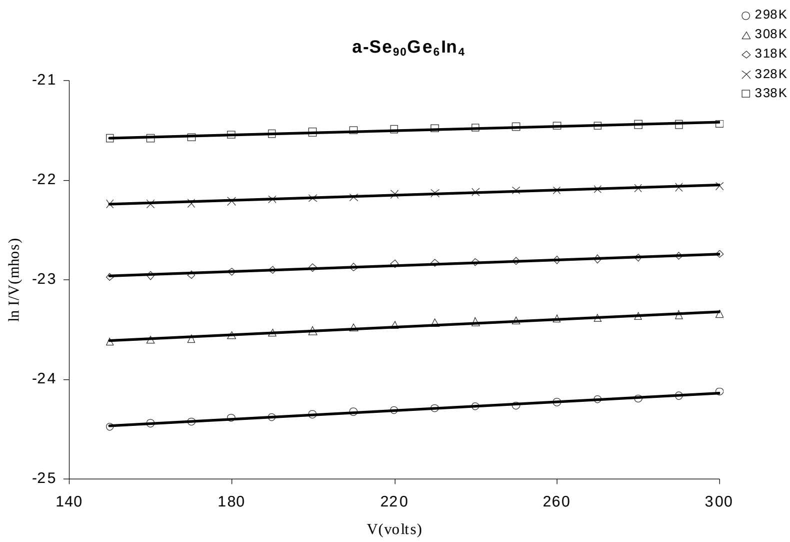

Fig. 2. Plots of ln(I/V) vs. V curves for a-$Se_{90}Ge_6In_4$ at different temperatures.

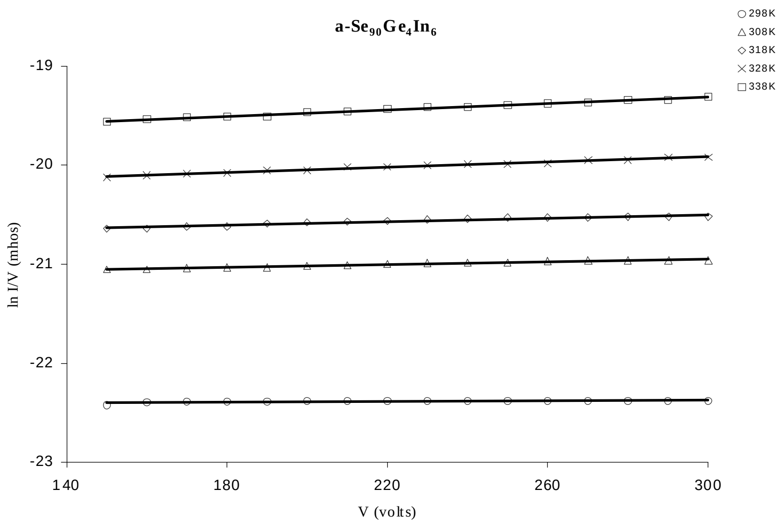

Fig. 3. Plots of ln(I/V) vs. V curves for a-$Se_{90}Ge_4In_6$ at different temperatures.

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]:

$$I = 2\, e\, A\, \mu\, n_0\, V / d\, [\, \exp\, (SV)] \quad (1)$$

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:

$$S = 2\, \varepsilon_r\, \varepsilon_0 / e\, g_0\, k\, T\, d^2 \quad (2)$$

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 eq. 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 eq. 1, ln(I/V) vs. V curves should be a straight lines whose slope should decrease with increase in temperature as evident from eq. 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 of a-$Se_{90}Ge_{10}$ and $Se_{90}Ge_6In_4$. In other samples, slope increases with increase in temperature (see Fig. 3 for example). This indicates that SCLC mechanism is not predominant in these alloys due to excessive joule heating. This may probably be the reason for an increase in the slope with increase in temperature instead of a decrease. Only in case of the first two samples, slopes follow the trend as required by eq. 2 . This shows that the theory of SCLC is applicable in these cases.

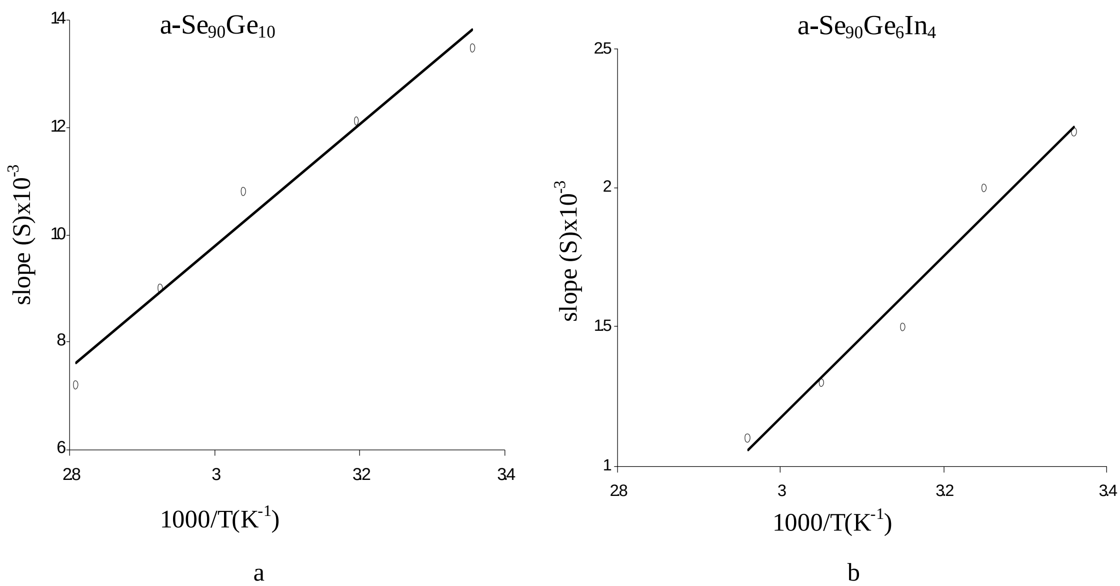

Fig. 4. (a)-(b): Plots of S vs. 1000/T for a-$Se_{90}Ge_{10}$ and a-$Se_{90}Ge_6In_4$ glassy system.

Using eq. 2 , we have calculated the density of localized states from the slope of Fig. 4. 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 results of the calculations are present in Table 1.

Table 1. Density of defect states (DOS) in a-Se_{90} Ge_{10-x} In_x glassy system.

Glassy alloys	slope of S vs. 1000/T curve	ε_r (1KHz, 305K)	g_0 ($eV^{-1}cm^{-3}$)
Se_{90} Ge_{10}	11.32 x 10^{-3}	3.61	2.80 x 10^{13}
Se_{90} Ge_6 In_4	2.90 x 10^{-3}	11	3.44 x 10^{14}

It is evident from Table 1 that density of localized states increases with In incorporation in pure $Se_{90}Ge_{10}$ binary system. During their measurement of high field and optical energy gap in Se-In system, Sayed et al [16] have reported that density of charged defects increases with increased In content and they have applied chemical bond approach to examine the structure and properties of the above system. The increase in values of dielectric constant (ε′) and dielectric loss (ε″) with In content in Se-Ge-In system have also been reported by Choudhary et al [19].

When the third element (In in the present case) is added to the Ge-Se system, the atoms of the third element induce structural changes in the host network Ge-Se. This leads to readjustments in the local environment, which disturbs the balance of characteristic charged defect states which may result in the shift of the Fermi level as observed by Kosek et al [20], in the Ge-Se-In system. In addition to the above readjustments, some new trap states can also appear in the mobility gap of the host material on addition of the third element, as evident from the present measurements.

Observation of SCLC in the composition where In is upto 4 at.% only indicates that defect states may be smaller as compared to other glass composition. Hence, the effect of incorporation of In in the Ge-Se system seems to be quite different at low and high concentration.

4. Conclusion

I-V characteristics have been studied in amorphous thin films of a-$Se_{90}Ge_{10-x}In_x$ (x = 0,4,6,10). At low fields, ohmic behaviour is observed. However, at higher fields (~ 10^4 V/cm) superohmic behaviour is observed.

Analysis of the observed data shows the existence of SCLC in the samples where In is upto 4 at.% only and excessive joule heating is found to be dominant in rest of the samples. From the fitting of the data in the sample where SCLC is being found, the density of localized states near Fermi-level is calculated. Observation of SCLC in some particular glass composition indicates that defect states may be lower in this case as compared to the other glass compositions.

Acknowledgements

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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