

Study of Density of Localized States in a-Se_{100-x}Sb_x Films by Space Charge Limited Conduction Measurements

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Abstract

The present paper reports dc conductivity measurements at high electric fields in vacuum evaporated a-Se_{1-x}Sb_x ($x = 0, 0.02, 0.04, 0.06$) thin films. At high fields ($\sim 10^4 \text{V}\cdot\text{cm}^{-1}$), current can be modelled by the theory of space charge-limited conduction in the case of uniform distribution of localised states in the mobility gap of these materials. The addition of Sb in Se results in an increase in the density of localised states.

Key Words: Chalcogenide glasses, Localised states, Space charge limited conduction, High field conduction.

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 for their greater hardness, high photosensitivity, higher crystallization temperature and smaller aging 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 Joule heating is negligibly small in these materials at moderate temperatures. Some studies have been reported in chalcogenide glassy semiconductors [5–15] and the results have been interpreted in terms of space charge-limited conduction (SCLC) 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 glassy system Se-Sb, where properties have been found to be highly composition dependent. Using SCLC theory 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 $\text{Se}_{1-x}\text{Sb}_x$ ($x = 0, 0.02, 0.04, 0.06$) were prepared by quenching technique. Component materials of high purity (99.999%) were weighed according to their atomic fractions and were sealed in quartz ampoules of length ~ 5 cm and internal diameter ~ 8 mm with a vacuum $\sim 10^{-5}$ Torr. The sealed ampoules were heated to and held at 900°C for 10–12 hours. The temperature of the furnace was raised slowly at a rate of $\sim 3\text{--}4^\circ\text{C}/\text{min}$. During heating, all the ampoules were constantly rocked by rotating a ceramic rod to which the ampoules were attached. 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 dropped into ice-cooled water. The quartz ampoules were then broken and the quenched samples were recovered. 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 the 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 samples so that a comparison of results could be made for the various glassy samples. The amorphous nature of the thin films was ascertained by x-ray diffraction.

For the measurements of high field conduction, thin film samples were mounted in a specially designed sample holder. A vacuum $\sim 10^{-2}$ Torr was maintained throughout the measurements. A dc voltage (0 to 300 V) was applied across the sample and the resultant current was measured by digital picoammeter. I-V characteristics were measured at various fixed temperatures (285–328 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 very near the films. Before measuring I-V characteristics, thin films were annealed in a vacuum $\sim 10^{-2}$ 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- $\text{Se}_{1-x}\text{Sb}_x$ ($x = 0, 0.02, 0.04, 0.06$) are examined at various temperatures (285–328 K). At low fields ($< 10^3$ V/cm), ohmic behaviour is observed in all the samples. However, at higher fields ($\sim 10^4$ V/cm), a superohmic behaviour is observed at all measuring temperatures.

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

$$S = 2\varepsilon_r\varepsilon_0/eg_0kTd^2. \quad (2)$$

As evident from equations (1) and (2), in the case of space charge limited conduction, a plot of $\ln I/V$ vs. V should be linear and slope S of these lines should decrease inversely with temperature.

Measurement data for $\ln(I/V)$ as a function of V for the various temperatures under which the samples were examined—is plotted in Figure 1. Indeed, as one can observe, $\ln(I/V)$ does largely vary linearly with V in all samples and at all temperatures. In Figure 2, note how the slope s of all linear fits increase linearly with *inverse* temperature for all samples. These results indicate the presence of space charge limited conduction in all the samples. Thin films contain a large number of defects due to dangling bonds that give rise to a large number of localised defect states. These localised states act as carrier trapping centres, and after trapping the injected charge from 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.

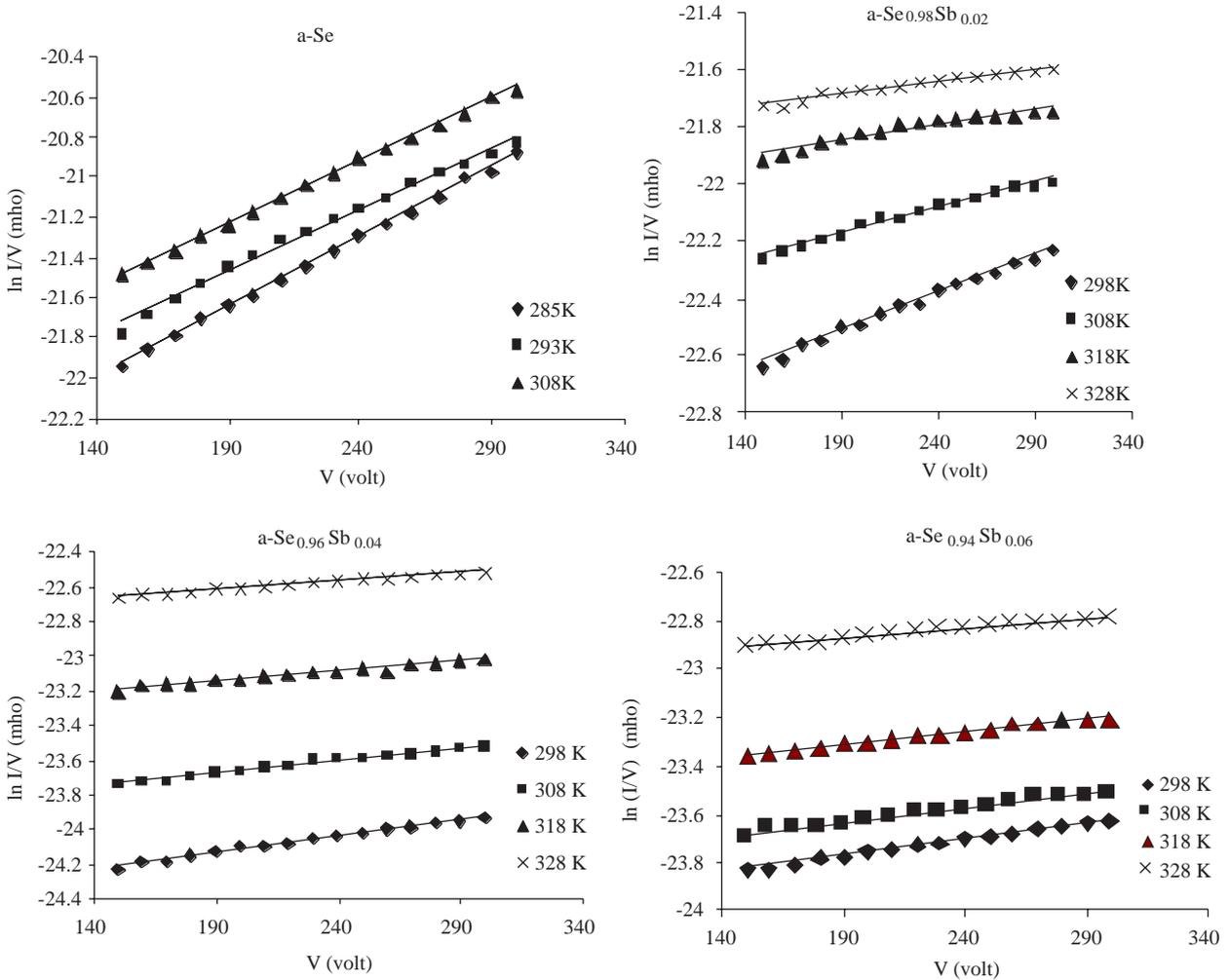


Figure 1. Plots of $\ln I/V$ vs. V for $a\text{-Se}_{1-x}\text{Sb}_x$ films at different temperatures.

Using equation 2, we have calculated the density of localised states from the slopes of Figure 2. The value of the relative dielectric constant ε_r are measured using a General Radio model 1620-AP capacitance bridge, employing the three terminal technique. The result of these calculations and measurements are given in Table and plotted in Figure 3 as a function of S_b concentration. It is clear that g_o increases with Sb concentration in a-Se-Sb system.

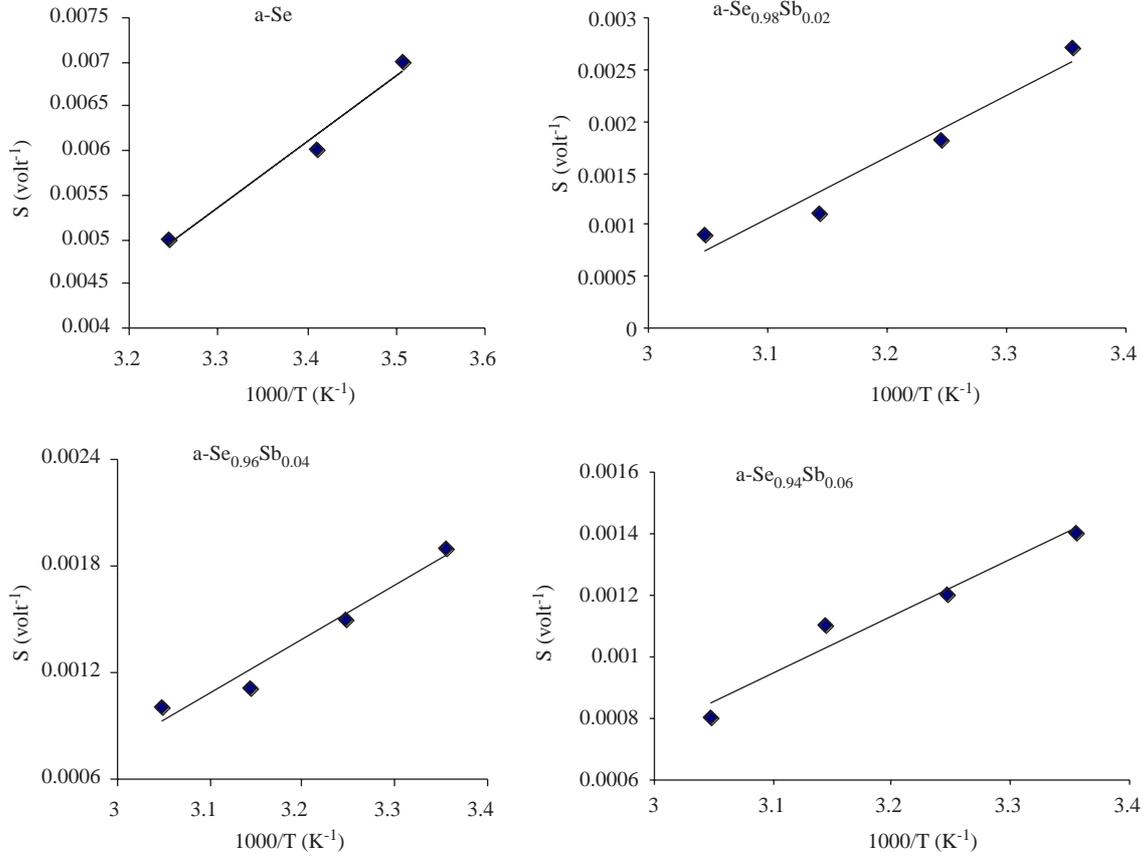


Figure 2. Plots of slope S of $\ln(I/V)$ as a function of V curves against $1000/T$ for $\text{a-Se}_{1-x}\text{Sb}_x$ films.

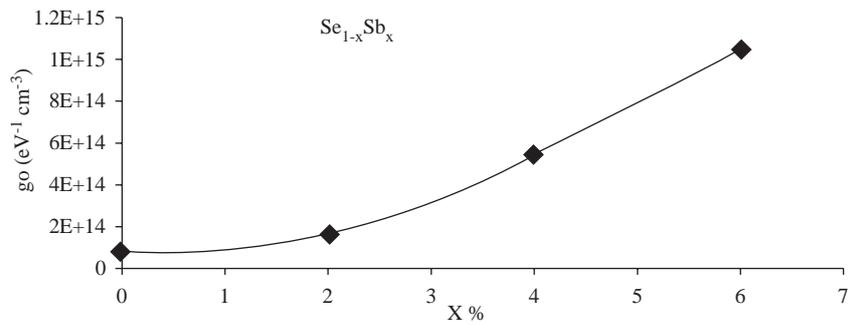


Figure 3. Plot of density of localised states g_0 vs. Sb concentration in $\text{a-Se}_{1-x}\text{Sb}_x$.

Table. Composition dependence of density of localised states g_0 in $\text{a-Se}_{1-x}\text{Sb}_x$.

Samples	Slope of s ($1000/T$)	ε_r ($f = 120$ Hz, $T = 303$ K)	g_0 (density of localised states, in units of $\text{eV}^{-1}\cdot\text{cm}^{-3}$)
a-Se	7.5×10^{-3}	6.2	7.34×10^{13}
a-Se _{0.98} Sb _{0.02}	6×10^{-3}	10.8	1.60×10^{14}
a-Se _{0.96} Sb _{0.04}	3×10^{-3}	18.4	5.45×10^{14}
a-Se _{0.94} Sb _{0.06}	1.8×10^{-3}	21.3	1.05×10^{15}

When isoelectronic atom Te is added to amorphous Selenium, the density of defect states increases, and hence the residual potential increases in xerographic characterizations. Onozuka et al. [22] observed that, on introducing Cl to Se-Te system, the residual potential is decreased. 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 electronegativity than Selenium [23], form negatively charged impurities, thereby compensating the effect of Te [22].

Along the same lines, one can expect that when Sb—having lower electronegativity than Se [23]—is introduced, positively charged defects are being created, thus increasing the density of defect states in the binary Se-Sb system beyond pure Se.

4. Conclusions

I-V characteristics have been studied in amorphous thin films of $\text{Se}_{1-x}\text{Sb}_x$ ($x = 0, 0.02, 0.04, 0.06$). At low fields ($< 10^3 \text{ V/cm}$), an ohmic behaviour is observed. At high fields ($\sim 10^4 \text{ V/cm}$), 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 for the case of a uniform distribution of localised states. The incorporation of Sb in a-Se results in an increase in the density of localised states around the Fermi level.

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