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Ohmic and space-charge-limited conduction in doped amorphous arsenic triselenide

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The electrical characteristics of arsenic triselenide have been studied. A number of parameters is evaluated on the basis of the theory of space-charge limited conduction and the following values were obtained: hole mobility $\mu = 2.68 \times 10^{-13} \text{m}^2\text{v}^{-1}\text{s}^{-1}$, room temperature hole concentration $p_o = 7.21 \times 10^{23} \text{m}^{-3}$, temperature parameter of trapping distribution $T_t = 790 \text{K}$; total trap concentration $N_t = 3.49 \times 10^{25} \text{m}^{-3}$. Both deep and shallow traps were present. The latter extended to a depth of $0.27 \pm 0.03 \text{eV}$ while the deep traps were at about the fermi level.

Key words: ohmic; space; charge; limited; conduction; semiconductors; amorphous; substances; doped; selenium

INTRODUCTION

A lot of work has been done on the semiconducting chalcogenide glasses in the past (Lakatos & Abkowitz, 1971; Seager et al., 1973; Marshall & Owen, 1976; Scharfe, 1970). This is because of their low cost of production. Some chalcogenides have been used as photoconductors in xerography and in vidicon tubes. There is a lot of literature on space charge limited conduction in organic materials (Gutmann & Lyons, 1981; Kao & Hwang, 1970; Lampert & Mark, 1970) but little in inorganic ones. One of the early works on SCLC was done by Bube (Bube, 1962) on cadmium sulfide in crystal form. He determined the depth of the traps ($E_t$) to lie between 0.49 eV and 0.53 eV and a trap density ($N_t$) of $10^{19} \text{m}^{-3}$. However, from the log $j^{-1}/T$ plot, he found $E_t = 0.28 \text{eV}$ and $N_t = 5 \times 10^{22} \text{m}^{-1}$. Hartke (1962), working on amorphous selenium films, found that capture centres were distributed uniformly in energy in a region extending downwards from 1 eV above the valence band edge. The densities of these levels were $10^{21} \text{m}^{-3}$ eV$^{-1}$ in the energy distribution width from 0.15 to 1 eV above the fermi level. The hole traps were $10^{27} \text{m}^{-3}$ extending from valence band to a depth of 0.14 eV while electron traps were $10^{25} \text{m}^{-3}$ extending 0.285 eV from valence band. However little has been done in doped selenium. Hence the relevancy of our research. We endeavored to get insight of charge transport mechanisms and carrier trapping in these materials. Such information is of particular importance in further utilization of the materials.

EXPERIMENTAL

The samples that we used were prepared by Dr. S. C. Katyal, H. P. University, India. The samples are prepared by quenching from melt. Pure arsenic and selenium of 99.99% purity in powder form are mixed in quartz tube. The tube is then evacuated, sealed and put in an electric furnace and heated at 1123 K for about 8 hours and shaken was from time to time to ensure proper mixing of the composition. After about 8 hours the melt is quenched in ice-cold water. The sample doped with aluminium is prepared by taking 1 at. % of aluminium (Al) in a mole of $\text{As}_2\text{Se}_3$ and the above procedure repeated.

We tested the samples for the amorphous nature using an x-ray diffractometer (Philips equipment, pw 1710).

The samples were then ground to plane parallel platelets of 5 x 5 mm$^2$ area and 1 mm thick .The surface of the material was roughed with emery paper to achieve an ohmic contact. The electrical contacts of the connecting wires to the specimen were made with the help of silver conducting paste. The two-probe dc method was employed because the materials have very high resistance. For temperature above room temperature, an electric oven whose temperature was controlled through a variac transformer was used. The temperature
was measured with a Cu/Ni thermocouple. For current measurements, ADC microvolt/ammeter model Philips 425A was used while a 7045 Digital Multimeter was used to measure the voltage. A dc stabilized power supply type VV315/A was used as a voltage source. For space-charge-limited current measurements, the values of the current were recorded when the value had settled to a steady one. Both values of current, that is, when the voltage was increased from minimum to maximum and vice versa were recorded and then the average value calculated.

RESULTS AND DISCUSSION

Figures 1 and 2 show that current-voltage relation at room temperature of pure and Al-doped As₂Se₃ respectively is ohmic at low fields but becomes non-ohmic at higher values of the applied field (Ahmad & Collins, 1991). Space Charge Limited (SCL) currents start at 250 volts for pure sample and at 400 volts for aluminium doped sample. At low voltages the slope of the log. I-log. V plot is approximately equal to unity (1.13 for pure & 1.08 for Al.) for both samples, while at higher voltages, above a well-defined transition voltage $V_{\text{trans}}$, the slope for Al-doped sample is 3.7 and for pure sample is 1.4. For both samples in the non-ohmic region, current was found to be approximately proportional to the square of voltage. This implies that there are single discrete shallow traps (Lampert & Mark, 1970) in agreement with the finding of Hartke (1962), who said that addition of arsenic to the amorphous films of As₂Se₃ introduced shallow electron traps. Since these shallow level of traps have been attributed to imperfections, and since it is doubtful that each imperfection would have an identical environment, a continuous distribution energy of the shallow traps seems more likely. The transition voltage $V_{\text{trans}}$ marks the end of the ohmic region and given by (Gutmann & Lyons, 1981)

$$V_{\text{trans}} = e n_{\text{eo}} d^2 / 2 \varepsilon_0$$

where $e$ is electronic charge, $n_{\text{eo}}$ concentration of majority carriers at equilibrium, $d$ the interelectrode distance, $\varepsilon_0$ the permittivity of free space and $\varepsilon$ permittivity of the solid. The value of $n_{\text{eo}}$ was found from experimental data to be $1.30 \times 10^{23} \text{ m}^{-3}$ and $1.33 \times 10^{23} \text{ m}^{-3}$ for pure and Al-doped samples respectively. Doping does not affect these carriers at this stage.
Further on the relation reaches another critical voltage $V_{\text{tr}}$, given by

$$V_{\text{tr}} = e\frac{d^2N_t}{2e}$$  \hspace{1cm} (2)

where $N_t$ stands for the total concentration of traps. For pure sample $V_{\text{tr}} = 1862$ volts and using (2), $N_t$ is found to be $3.49 \times 10^{25} \text{m}^{-3}$. This is in agreement with other researchers. Experiments on chalcogenides through photoconductivity (Mott & Davis, 1979), pinned fermi energy (Mott & Davis, 1979), and photoluminescence (Street et al., 1974) confirm that these materials have high concentration of defect states in the range $10^{24} - 10^{25}\text{m}^{-3}$. Hartke (1962) found the electron traps to be $10^{25}\text{m}^{-3}$ lying $0.285\text{ eV}$ below the conduction band. However, this traps-filled limit was not observed with the Al-doped sample because the sample burnt out at voltage $= 1514$ volts. This could have been due to the break down field or the heating effects of the high currents involved but from our work we could not be certain.

From the conductivity equation given by

$$\sigma = \sigma_0 \exp \left(-\frac{E_i}{K_B T}\right)$$ \hspace{1cm} (3)

and fig. 3 (In $\sigma$-T graph), the depth of shallow traps $E_i$ was obtained in the non-ohmic region at 600 volts. From the slope, $E_i$ was determined to be $0.27\text{ eV}$. However, from the information given we can not tell if these were electron traps or hole traps. Hartke (1962) found electron traps at depth of $0.285\text{ eV}$ in conduction band. This suggests that our result might be hole traps, but of course this can be verified which more experimental work. The number of traps in the range $K_B T$ near the fermi level was found from the theory that change voltage needed to double the current in the space charge limited region is a good measure of the trap density($N_T$) per $K_BT$ at the position of the quasi-fermi level (Lampert & Mark, 1970).These values were $1.25 \times 10^{20}\text{m}^{-3}$, $1.75 \times 10^{20}\text{m}^{-3}$ for pure and Al. doped respectively. Hartke (1962) value was $10^{21}\text{m}^{-3}$ for arsenic, one order higher than ours.

At low voltages we expect Ohm’s law given by Ahmad & Collins (1991) as

$$j = eP_0\mu\left(\frac{V}{d}\right)$$ \hspace{1cm} (4)

will be observed. In the above equation $j$ is the current density due to holes, $V$ the applied voltage, $P_0$ the concentration of thermally activated holes in the valence band, the hole mobility, $e$ and $d$ have been defined already. From the above theory, the SCLC characterstics for a p-type semi conductor with an exponential distribution of trapping levels is given by

$$j = eP_0\mu\left(\frac{V}{d}\right)$$ \hspace{1cm} (5)

where $N_v$ is the effective density of states in the valence band edge, $K_B$ is the Boltzmann constant, $P_0$ the trap density per unit energy range at the valence band edge. The constant $I$ is the temperature parameter describing the exponential trapping distribution $P(E)$ equal...
P(E) = P₀ exp.(-E / kₜ Tₜ)  \hspace{1cm} (6)

where P(E) is the trap density per unit energy range at an energy E above the valence band edge. From eq. (4) a slope of 3.7 implies that l = 2.7 and thus the parameter Tₜ = 790 k for T = 297k for Al. doped sample. Ahmad & Collins (1991) found Tₜ = 770 k for T = 300 k for lead phthalocyanine thin films. This analysis did not work for the pure sample. Probably doping the sample with aluminium introduces defects that interfere with trap arrangement already there. It appears the smeared continuous energy levels became exponential trapping levels after doping.

Temperature parameters allow us to obtain more information about the sample under investigation. From fig. 3 (lg j versus T⁻¹ graph) (3), the intercept on the lg j axis is given by:

\[ \text{lg} jₒ = \text{lg}(\text{eg} NᵥV/d) \]  \hspace{1cm} (7)

where jₒ represents the current density at infinite temperature. In order to determine the mobility \( \mu \), a value for Nᵥ is needed. Hartke (1962) evaluated Nᵥ = 10²⁶ m⁻³ for amorphous selenium films. The intercept extrapolated on the lg jₒ axis is -1.32 for 600 V. Substituting these values in eq.(7), the measured value of mobility is 2.68 x 10⁻¹³ m² v⁻¹ s⁻¹. Hartke’s drift mobility was (2-4) x 10⁻¹² m² v⁻¹. The room temperature concentration Pₒ of holes in the valence band can be calculated using the ohmic characteristics of graph 1 and by substituting the derived value of mobility in eq. (4). The value found was 7.21 x 10²³ m⁻³, which compares well with 1.30 x 10²³ m⁻³, concentration of majority carriers at equilibrium found earlier on.

CONCLUSION

Investigations were done on amorphous arsenic triselenide in pure form and doped with aluminium through measurements of ohmic and SCLC. The findings showed that the samples had both shallow and deep traps. The shallow traps lie 0.27 ± 0.03 eV in the energy gap and were arranged in smeared continuous energy level in pure sample. The deep traps were at about the fermi level. In the doped sample traps appeared to lie in exponential trapping levels. We suggest that more work be done on traps in doped in arsenic triselenide samples to find out what how arrangement of traps is modified after doping.
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REFERENCES


