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Density of Charged Defect States Using A.C. Conductivity Measurement in Glassy Alloys of $Se_{90}M_{10}$ (M = Ag, In)

Physics

KEYWORDS: A.C. Conductivity, Chalcogenide glasses, Charged Defect States, CBH model, Hopping Conduction

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ABSTRACT

Frequency and temperature dependence of a. c. conductivity is studied in glassy alloys of Se90Ag10 and $Se_{90}In_{10}$ in the temperature range (289 K - 402 K) and frequency range from 500 Hz to 500 kHz. The theory of a. c. conductivity is used to fit the observed experimental results and from this fitting, density of charged defect states has been calculated. It has been found that the bipolaron hopping dominates over single polaron hopping. It has also been found that the Density of charged defects is three times more in $Se_{90}In_{10}$ alloy as compared to $Se_{90}Ag_{10}$ alloy.

INTRODUCTION

Due to technologically important electrical properties chalcogenide glasses have drawn enormous attention in recent years. These materials show IR transparency and electrical properties, which makes them potentially useful for applications such as threshold and memory switching [1]. These glasses are widely used in various other applications such as ultra high density phase change memories, computer memories, X-Ray imaging, erasable high density optical memories, photodetectors, photoreceptors, thermal imaging, [2-11]. Ag doped chalcogenide glasses are widely used in various applications such as gratings, solid electrolytes, microlenses, bio-sensors, waveguides, optical memories, etc. Silver containing chalcogenide glasses exhibits single glass and crystallization peaks [12-14]. This is very important condition for the use of these materials in rewritable disk [15]. Studies of the electronic nature of amorphous material give information about its electrical behavior and this may be related to structural properties.

Since charge carriers are localized so their behavior can be studied by a. c. conductivity measurements [16-17]. The a. c. conduction in amorphous glassy semiconductors has been studied by various workers [18-23]. In a-semiconductors the a. c. conductivity (a.c.) is expressed by the usual relation () = As where is angular frequency and A and s are constants. Different conduction mechanisms can lead to s type of behavior for a. c. conductivity, but it is not easy to decide which of those mechanisms is responsible for the observed conduction mechanism. In order to explain the mechanism of a. c. conduction in a-semiconductors a number of models [24-26] have been proposed. Recently, the variation of a.c. with frequency and temperature in majority of chalcogenide glasses have been explained on the basis of correlated barrier hopping (CBH) model [26] which is based on the concept of the charged defect states in these glasses. From the experimental data, density of defect states has also been evaluated. In the present work, the a. c. conductivity is measured in Se90Ag10 and Se90In10 glasses as a function of frequency and temperature. Density of defect states has also been evaluated.

EXPERIMENTAL

The glassy alloys of Se90Ag10 and Se94Ag6 are prepared by melt quenching technique. To achieve this, exact proportions of high purity (99.999%) Se, In and Ag elements were weighed according to their respective atomic percentages., An electronic balance (LIBROR, AEG-120) having least count of 10-4g is used for weighing. The materials were sealed in evacuated quartz ampoules having dimensions of approximately 5 cm in length and 8 mm internal diameter. These ampoules along with materials were then heated upto a temperature of 800 0C at the rate of 3-4 0C per minute and kept at the same temperature for about 12 hours. While heating, the ampoules were constantly rocked for the preparation of homogeneous alloys. Quenching was done by dropping hot ampoule suddenly in ice cooled water.

These glassy alloys were then ground to get very fine powder and pellets were prepared by compressing the powdered alloy under a pressure of 5 - 6 tons in a die. For good electrical contacts, both faces of the pellets were coated with Indium. A. C. conductivity measurements were done by mounting the pellets inside a sample holder where a vacuum ~ 10-2Torr is maintained during measurements. The temperature inside the sample holder is measured by a copper - constantan thermocouple mounted near the pellet. A digital LCR meter WAYNE KERR-4255 is used to measure A. C. conductance at different frequencies.

THEORETICAL BASIS

In these materials A. C. conductivity is found to be frequency dependent which is due to the presence of localized charged defect states. Total conductivity in these materials is given by following relation [17]:

$$\sigma(\omega) = \sigma_{dc} + \sigma_{ac} \qquad \ldots 1$$

Since the obtained dc conductivity σ_dc is much smaller than total electrical conductivity**σ(ω)**, therefore it can be neglected in Eq. (1), so **σ(ω)** is considered to be ~**σ**_ac (**ω**).

It is assumed that the a. c. conductivity in semiconductors is caused due to a pair of localized states, where electrons move back and forth with a particular relaxation time. But in amorphous semiconductors, the back and forth movement is again valid but the relaxation time is not fixed, it is directly proportional to the randomness of these charged defect states since these charged defect states are randomly distributed. Therefore, the A. C. conductivity then will be sum of contributions from all such pairs.

$$\sigma_{ac} \propto \omega^s \qquad \ldots 2$$

or, $$\sigma_{ac} = A\omega^s$$

here A and s are temperature dependent constants.

According to CBH model [26], charge carriers hop over a potential barrier. For the electron to hop over a barrier of height W, the

relaxation time τ , is given by

$$\tau = \tau_0 \exp\left(\frac{W}{kT}\right), \quad \ldots 3$$

Here τ _0 is a characteristic relaxation time, which is of the order of an atomic vibrational period and k is the Boltzmann constant.

For single polaron hopping, the a. c. conductivity σ _ac (ω) originating from randomly distributed defects centers can be expressed as

$$\sigma_{ac}(\omega) = \pi^3 \varepsilon\varepsilon_0 N N_p \omega (R_\omega)^6/6 \quad \ldots 4$$

Where

$$R_\omega = \left(\frac{e^2}{\pi\varepsilon\varepsilon_0 W_m}\right)\left[1 + \frac{kT}{W_m \ln(\tau_0\omega)}\right]^{-1} \quad \ldots 5$$

For single polaron hopping process, the maximum barrier height,W_m, is taken as half of the band gap. For bipolaron hopping, the A. C. conductivity, σ_ac (ω), originating from intimate D+ and D- pairs having a non-random distribution can be written as

$$\sigma_{ac}(\omega) = \left(\frac{1}{6}\right) n\pi^3 \varepsilon\varepsilon_0 N N_p \omega (R_\omega)^6 \exp\left(e^2/ 4\pi\varepsilon\varepsilon_0 k T_g R_\omega\right) \quad \ldots 6$$

here, n = 1 for single polaron and n = 2 for bipolaron hopping. T_g is the glass transition temperature. The maximum height W_m is regarded as the band gap. Here N is the density of localized states at which carriers exists, N_P is the density of localized states to which the carriers hop. N_P is considered to be equal to N/2.

The hopping distanceR_ω is given by

$$R_\omega = \left(\frac{ne^2}{\pi\varepsilon\varepsilon_0 W_m}\right)\left[1 + \frac{kT}{W_m \ln(\tau_0\omega)}\right]^{-1} \quad \ldots 7$$

CBH model predicts the frequency exponent 's' to be

$$s = 1 - \frac{6kT}{W_m - kT\ln\left(\frac{1}{\omega\tau_0}\right)} \quad \ldots 8$$

It has been found that the frequency exponent 's' is both frequency and temperature dependent. For small values of W_m/kT, 's' increases with increasing frequency and for large values of W_m/kT 's' is nearly unity and the increase is so small that 's' is effectively independent of frequency.

RESULTS AND DISCUSSION:

Frequency Dependence of A. C. Conductivity:

The frequency dependence of a. c. conductivity for the investigated samples is studied in the frequency range (500 Hz – 500 kHz) for different values of temperature in the range (289 K –402 K). Figs. 1 - 2 show a frequency dependence of A. C. conductivity at different temperatures for both the compositions. It is clear from these figures that ln(_(a.c.)) increases linearly with ln(ω). The decrease in slope indicates that the frequency exponent 's' decreases with increase in temperature. This is consistent with the CBH model as mentioned above.

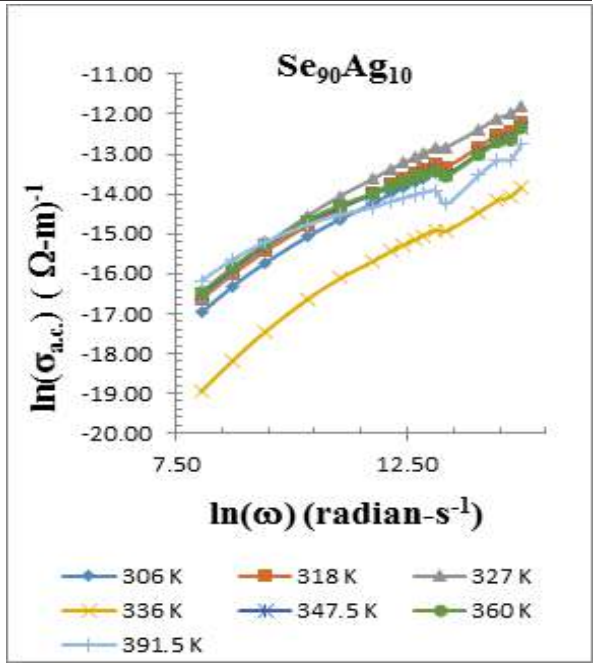

Figure : Frequency dependence of A. C. conductivity for Se90Ag10 glassy alloy at different temperatures.

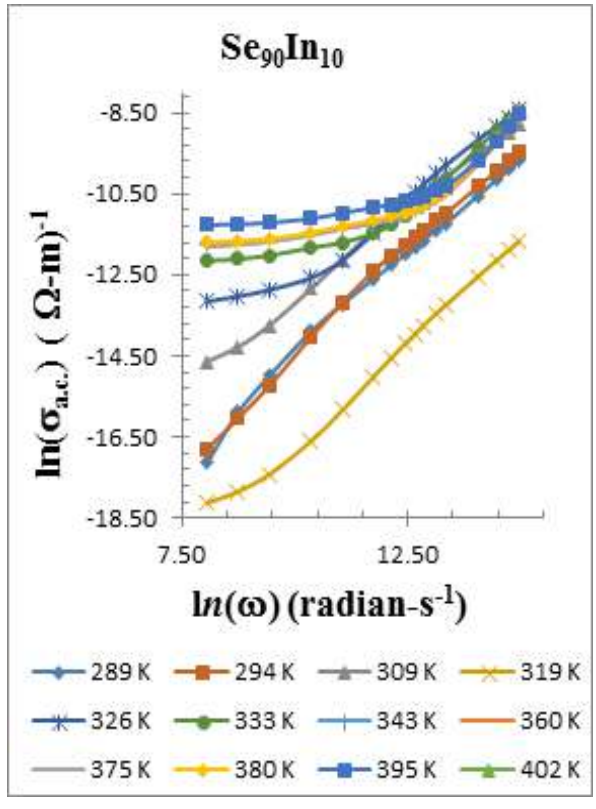

Figure 2: Frequency dependence of A. C. conductivity for Se90In10 glassy alloy at different temperatures.

Temperature Dependence of A. C. Conductivity

The temperature dependence of A. C. conductivity for glassy alloys of Se90Ag10 and Se90In10 at different frequencies ranging from 500 Hz to 500 kHz is shown in Figs. 3 and 4. These curves show that A. C. conductivity increases with temperature.

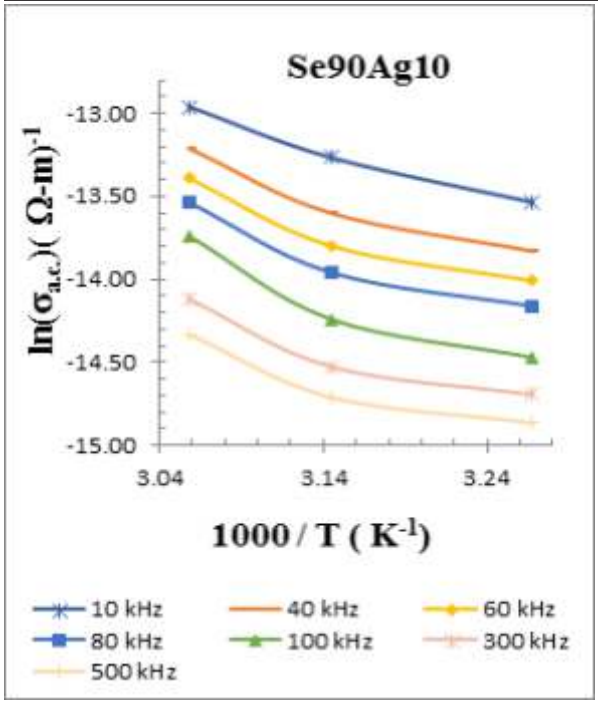

Figure 3: Temperature dependence of A. C. conductivity for Se90Ag10 glassy alloy at different frequencies.

In amorphous chalcogenide alloys the A. C. conductivity may be considered as the sum of two types of conduction mechanisms, i.e., single polaron hopping and bipolaron hopping. Considering this, the experimental data has been fitted to CBH model. The values of NNP and W_m have been adjusted to obtain a best fit of the theoretical curves to the experimental data. The fitting is done for one frequency and the same values are used for different frequencies. The different parameters used are given in Table 1

TABLE: 1
DIFFERENT PARAMETERS USED FOR CURVE FITTING OF Se90Ag10 AND Se90In10 ALLOYS USING SINGLE POLARON (n = 1) AND BIPOLARON (n = 2) THEORY AT 500 Hz.

Sample	Wm (eV)	NNp $[m^{-3}]^2$	NNp $[cm^{-3}]^2$	
	n = 1	n = 2		
$Se90Ag_{10}$	0.60	1.20	1.1 x 1051	1.1 x 10^{45}
$Se90In_{10}$	0.60	1.20	8.5 x 1051	8.5 x 10^{45}

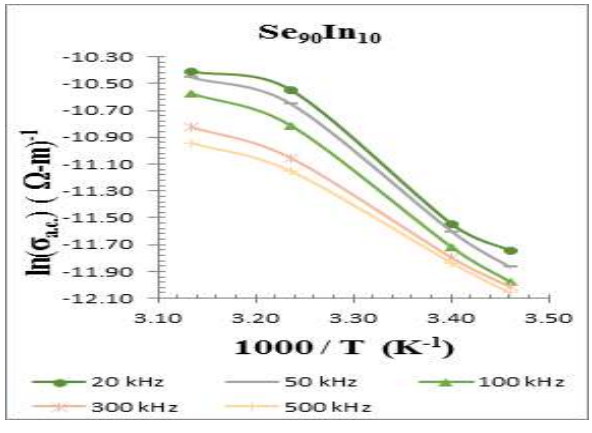

Figure 4: Temperature dependence of A. C. conductivity for Se94Ag6glassy alloy at different frequencies

The fitting is shown in Figs. 5 and 6, in case of Se90In10 and Se90Ag10. These figures show that the experimental values of A. C. conductivity in Se90Ag10 and Se90In10 are in close agreement with bipolaron hopping mechanism.

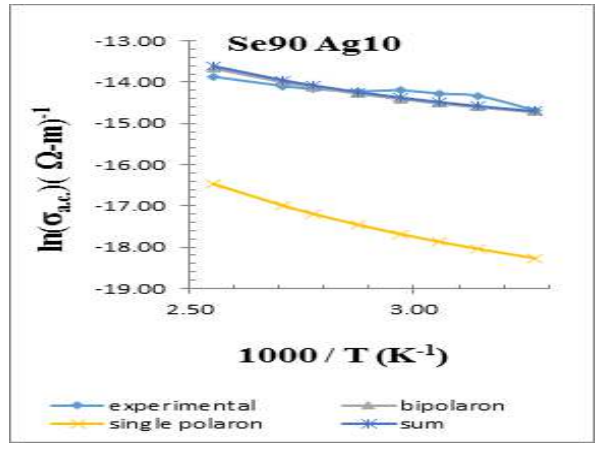

Figure 5: Curve fitting of A. C. conductivity with theory for Se90Ag10glassy alloy at 500 Hz

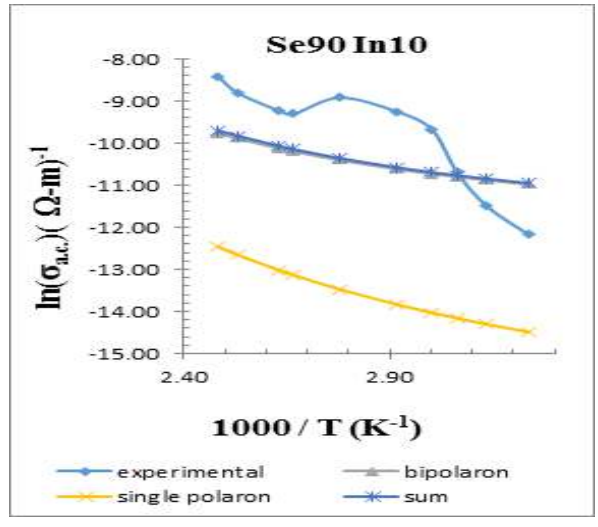

Figure 6: Curve fitting of A. C. conductivity with theory for Se90In10 glassy alloy at 500 Hz.

The sum of the contributions in A. C. conduction from single polaron and bipolaron hopping mechanisms are also in close agreement with experimental data. The contribution of single polaron hopping mechanism in A. C. conduction is very small.

The density of charged defect states N is obtained from the values of NNP which have been arbitrarily chosen to fit the curve for all the glassy alloys. These values for two different alloys are also given in Table 2.

TABLE-2
VALUES OF DENSITY OF CHARGED DEFECT STATES FOR Se90Ag10 AND Se90In10 ALLOYS AT 500 kHz.

Sample	NNp $[m^{-3}]^2$	N (cm^{-3})
$Se90Ag_{10}$	1.1 x 10^{45}	3.31 x 10^{22}
$Se90In_{10}$	8.5 x 10^{45}	9.21 x 10^{22}

CONCLUSION

The A. C. conductivity in glassy alloys of Se90Ag10 and Se90In10 has been studied as a function of frequency and temperature. A. C. conductivity is found to be dependent on temperature. The results

could be explained using correlated barrier hopping (CBH) model. It has been observed that A. C. conduction in these glassy alloys may be well explained in terms of single polaron and bipolaron hopping mechanism. We have observed that the contribution in A. C. conductivity due to bipolaron hopping is much more dominating than the contribution due to single polaron. The experimental data has been fitted to the theory of A. C. conductivity and from this fitting the density of defect states has been calculated. Density of defects is three times more in Se90In10 alloy as compared to Se90Ag10 alloy.

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