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AC CONDUCTIVITY OF Se_{85-x} $Te_{15}Ge_x$ GLASSES

ANIL KUMAR, MANOHAR LAL[a], KANCHAN SHARMA,
NAVDEEP GOYAL[*]
Department of Physics, Panjab University, Chandigarh, India-160014
[a]*GGD SD College, Sector-32, Chandigarh-160030, India.*

Frequency and temperature dependent ac conductivity studies of Se_{85-x} $Te_{15}Ge_x$ (x= 0, 2,6,10, 15) glasses have been reported. Results indicate that Se_{83} Te_{15} Ge_2 obeys well established relation $\sigma_{ac}\alpha\omega^s$, frequency exponent *s* is found to decrease with increase in temperature and results have been explained as the basis of correlated barrier hopping (CBH) model and numerical calculations agree well with experimental results. But a different type of behavior has been observed for Se_{85-x} $Te_{15}Ge_x$(x = 6, 10, 15). Distinct peaks have been observed in the plots of temperature dependence of ac conductivity for Se_{85-x} $Te_{15}Ge_x$ (x = 10, 15). The results have been interpreted using CHB and "simple pair" model. Ge introduced localized states deep in the band gap and form simple pairs. Theoretical results obtained by using these models have been found to be in agreement with experimental results.

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1. Introduction

The effect of an impurity doping of amorphous semiconductors is of great interest. The effect of impurity depends on the conduction mechanism and the structure of the material [1]. Measurement of frequency and temperature dependence of ac conductivity has been found to be powerful tool to investigate the gap states in amorphous semiconductors. CBH model is generally used to explain the ac conductivity behavior in chalcogenide glasses [2-11]. We report the temperature and frequency dependent ac conductivity studies of Se_{85-x} $Te_{15}Ge_x$ and results have been explained using CBH model [2-4] and simple pair model [12, 13].

2. Experimental detail

For the present work, the glassy materials have been prepared by melt quench method. The constituent elements (5N pure) were weighed in the required atomic weight percentages using electronic balance with accuracy of 0.0001gm.The weighed components were sealed in properly cleaned quartz ampoules under a vacuum of 10^{-5} mbar. The sealed ampoules were heated gradually upto 1000^0C in the rocking furnace and later the temperature of the furnace was raised upto 1050^0C and this temperature was maintained for about 24hrs. Rocking has been done to insure proper mixing and homogeneity of samples. Heated ampoules were immediately quenched in ice cold water. The amorphous nature of the samples was confirmed using X-ray diffraction technique.Compressed pellets were prepared by grinding bulk-ingots into fine powder and compressing the powder in a die under a hydraulic press (10^3kg/m^2). A three terminal sample holder has been fabricated for the measurement of ac and dc conductivity of pellet- shaped

[*]Corresponding author: ngoyal@pu.ac.in

samples. A thermocouple has been inserted inside the sample holder which has been kept close to the sample for the measurement of correct temperature. A vacuum of the order of 10^{-4} to 10^{-5} Torr was achieved inside the sample holder using vacuum pump. A general radio bridge (Model 1615-A) was used for the measurements of frequency –dependent ac conductivity and dielectric constant of the materials. This bridge is designed for the precise measurements of capacitance and conductance. Its direct read-out system minimizes the reading errors and permits rapid operation.

3. Results and discussion

The frequency and temperature dependent behaviour of Se_{85} Te_{15} had been studied [11] and obeys the well-known relation

$$\sigma_{ac} = A\omega^{s} \quad (1)$$

where *s* is frequency exponent and A is constant.

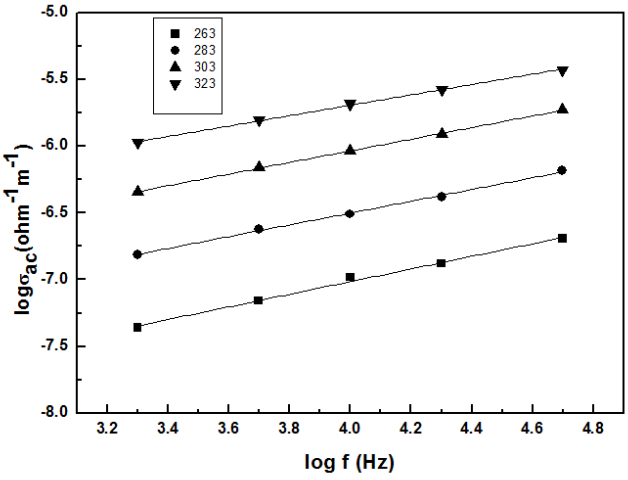

Fig.1. logσ_{ac}versus log f for Se_{83} Te_{15} Ge_2 glass.

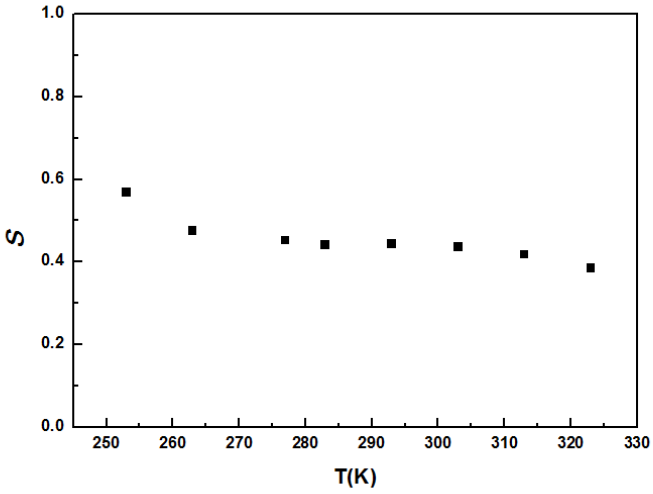

Fig. 2. Temperature dependence of frequency exponent s for $Se_{83}Te_{15}Ge_2$

Figure(1) shows the frequency dependent behavior of ac conductivity of $Se_{83}Te_{15}Ge_2$ at different temperatures. From figure, it is clear that relation (1) is obeyed for this glass. The

temperature dependence of *s* for $Se_{83}Te_{15}Ge_2$ is given in figure 2. The value of *s* decreases with rise in temperature. So it is clear that the behaviour of $Se_{85}Te_{15}Ge_2$ is similar to that of $Se_{85}Te_{15}$. But the relation (1) has not been followed for Se_{85} $Te_{15}Ge_x$ (x = 6, 10, 15) and prominent peaks appear in the log σ_{ac} versus 1000/T graphs for Se_{75} Te_{15} Ge_{10} and Se_{70} Te_{15} Ge_{15} glasses. The peaks are not prominent in case of Se_{75} Te_{15} Ge_6. In the following discussion, an attempt has been made to explain this deviation from the general behavior of amorphous semiconductors.

The a.c. conductivity in semiconductors has been interpreted in terms of pair approximation [12,13]. In this approximation, it is assumed that a pair consists of two localized states between which carriers hop back and forth. Each pair has its own relaxation time τ which is related to the transition probability of the carrier. The contribution of one such pair to a.c. conductivity is given by $C\omega^2\tau/(1+\omega^2\tau^2)$ where C is independent of ω&τ. If the relaxation time is same for all the pairs then the above relation will hold good in the simple pair model [12,13]. But in amorphous semiconductors, the conduction is due to hopping between defect states [2-4]. According to correlated barrier hopping(CHB) model, the a.c. conductivity is given by

$$\sigma_{ac} = n\pi^2 NN_p \kappa \omega R_\omega^6/24 \tag{2}$$

Where n is the number of polarons involved in the hopping process, R_ω is the hopping distance for the condition $\omega\tau$ =1 and is given by [4].

$$R_\omega = \frac{4ne^2}{k(W + kT\ln(\omega\tau_0)} \tag{3}$$

NN_P is given by

$NN_P = N_T^2$ (for bipolaron hopping)

$NN_P = N_T^2 \exp(-U_{eff}/2kT)$ (for single polaron hopping)

where N_T is the number of density of states and U_{eff} is the Hubbard intrasite correlation energy.
The value of frequency exponent *s* is calculated from equation (2) and (3) and is approximately equal to [3]

$$s = \frac{d(\ln \sigma_{ac})}{d(\ln w)} = 1 - \frac{6kT}{W - kT\ln(1/wT_0)} \tag{4}$$

$W = W_M$ (for bipolaron hopping)

$W = W_1$ or W_2 (for single polaron hopping)

W_1 is the energy(distortion+electronic) required to take an electron from the valence band and place it on a D^0centre thereby turning it into D^- and W_2 is the total energy necessary to take an electron from D^0centreand place it in the conduction band.W_M can also be understood as simplytwice the energy difference between Fermi level and conduction band [3].
In general, the experimentally obtained ac conductivity is given by

$$\sigma_{ac} = \sigma_b + \sigma_s \tag{5}$$

Where σ_b is contribution from bipolaron hopping and σ_s is contribution from single polaron hopping. Single polaron hopping is dominant at higher temperature and bipolaron hopping is dominant at lower temperature.

CBH model explains the frequency and temperature dependent behavior in Se_{85} Te_{15} and is predominantly due to bipolaron hopping in the studied temperature range (213K to 283 K)[11].

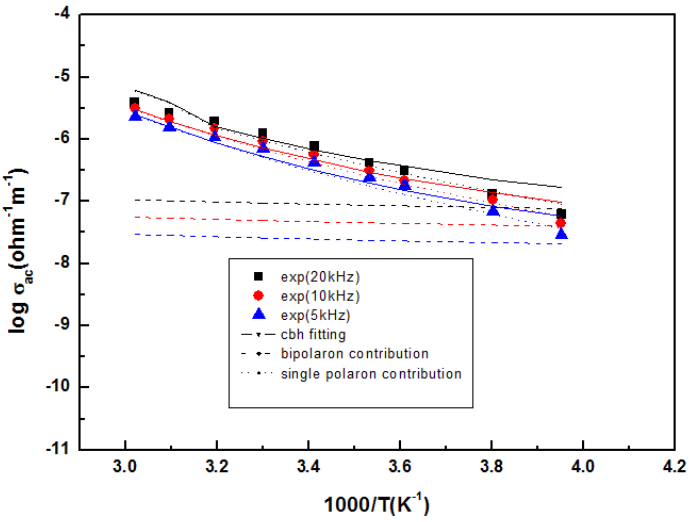

Fig. 3.Temperature dependence of ac conductivity for Se_{83} Te_{15} Ge_2

Fig. (3) shows the experimental data and calculated curves (from CHB model fitting) for $Se_{83}Te_{15}Ge_2$. The two curves are in good agreement with each other. Parameters used in the calculations are shown in Table 1. CBH model shows that conductivity in Se_{83} Te_{15} Ge_2 is predominantly due to single polaron hopping in the studied temperature range (263 K to 333K).But the temperature and frequency dependent behavior of ac conductivity for S_{85-x} $Te_{15}Ge_x$ (x = 6, 10, 15) in the temperature range 263K-333K and frequency range 5kHz - 20kHz cannot be explained solely on the basis of CBH model of randomly distributed centers.

To explain the ac conductivity in S_{85-x} $Te_{15}Ge_x$ (x = 6, 10, 15),we assume that Gemanium doping gives rise to some neutral defect states called simple pairs in the band gap and ac conductivity due to these defect pairs (σ_{SP}) is given by [12].

$$\sigma_{SP} = \frac{e^2 N_S r_0^2}{12 kT} \quad \frac{\omega^2 \tau}{1+\omega^2 \tau^2} \tag{6}$$

where N_S is concentration of pairs and r_0 is hopping distance.
σ_{SP} in (6) is considered to be function of τ with constant ω , then σ_{SP} has a peak at $\omega\tau = 1$. Therefore the pronounced peaks observed in figures 5&6 are justified by equation (6)
The relaxation time τ for hopping of electrons over a barrier height W is given by

$$\tau = \tau_0 \exp (W/kT)$$

Where τ_0 is characteristic relaxation time and k is Boltzmann constant.
Recently it has been reported [14] that for thermally activated ac conduction in chalcogenide glasses,

$$\tau = \tau_0 \exp (W/kT) \exp (-W/kT_0) \tag{7}$$

wherek $T_0 = E_{MN}$ is Mayer-Neldel energy.

Considering that the contribution to ac conductivity due to simple pairs is thermally activated, relation (7) has been used to calculate τ by using E_{MN} = 40.67 meV . E_{MN} has been calculated from dc conductivity measurements of the glasses under consideration [15].
So, the ac conductivity for Ge doped Se_{85} $Te_{15}$is expected to be

$$\sigma_{ac} = \sigma_b + \sigma_s + \sigma_{SP} \quad (8)$$

Where σ_{SP} is simple pair contribution.

Theoretical calculation of ac conductivity in case of Se_{85-x} $T_{15}Ge_x$ (x = 6,10, 15) have been done using (2), (3), (6) and (7) and the parameters obtained for best fits are listed in Table 1.

Table1. Parameters for CBH and Simple Pair model.

Glass	κ	W_M(eV)	W_1(eV)	W_2(eV)	U_{eff}(eV)	$N_T(m^3)$	W(eV)	τ_o(s)	$N_s r^2_0(m^{-1})$
Se_{83} Te_{15} Ge_2	7.5	2.78	0.78	1.74	0.26	$2*10^{20}$	-	$1.0*10^{-13}$	-
Se_{79} Te_{15} Ge_6	7.0	2.70	0.80	1.52	0.38	$9*10^{19}$	1.10	$1.8*10^{-13}$	$1.60*10^5$
$Se_{75}Te_{15}$ Ge_{10}	6.0	2.40	0.90	1.02	0.48	$4*10^{19}$	1.34	$1.0*10^{-14}$	$1.44*10^7$
$Se_{70}Te_{15}$ Ge_{15}	5.5	2.24	0.80	1.12	0.40	$9*10^{18}$	1.06	$2.0*10^{-14}$	$7.84*10^6$

Figs. 4, 5, 6 show the experimental data and calculated curves (from CHB model plus simple pair contribution fitting) for Se_{79} Te_{15} Ge_6 ,Se_{75} Te_{15} Ge_{10} and Se_{70} Te_{15} Ge_{15} respectively . In each case, the two results are in good agreement with each other.

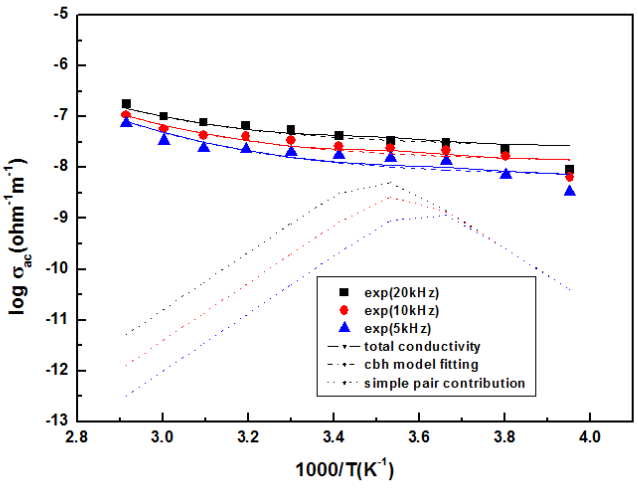

Fig. 4. Temperature dependence of ac conductivity for Se_{79} Te_{15} Ge_6

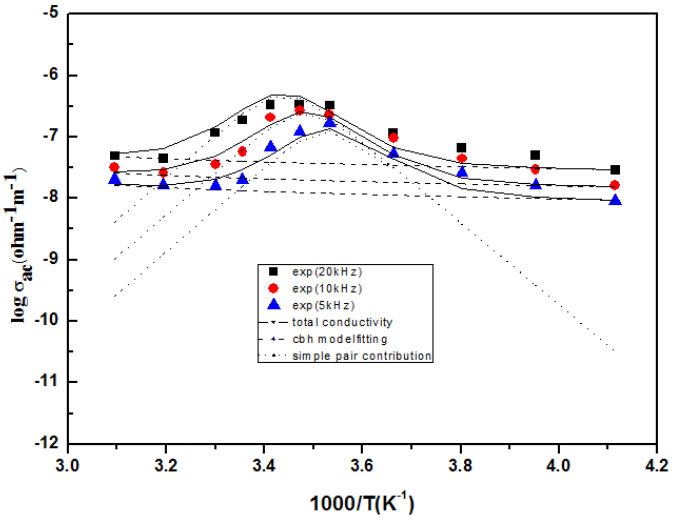

Fig5. Temperature dependence of ac conductivity for Se_{75} Te_{15} Ge_{10}

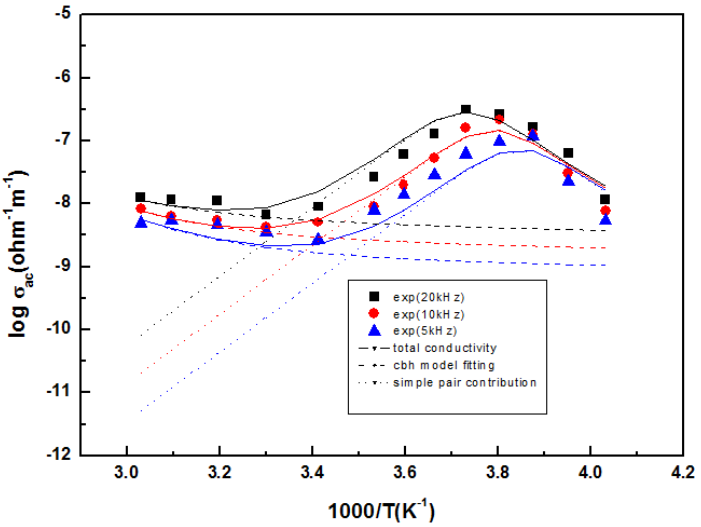

Fig. 6. Temperature dependence of ac conductivity for Se_{70} Te_{15} Ge_{15}

An agreement between theoretical and experimental results clearly indicates that the behaviourof S_{85-x} $Te_{15}Ge_x$ (x = 6, 10, 15) seems to be well explained by introducing the concept of simple paired states. From the fits it is clear that single polaron hopping is dominant in case of Se_{83} Te_{15} Ge_2 and Se_{79} Te_{15} Ge_6 in the studied temperature range and simple pair contribution to conductivity is very small due to lack of sufficient number of paired states. But the sufficient numbers of paired states are available in case of Se_{75} Te_{15} Ge_{10} and Se_{70} Te_{10} Ge_{15} to contribute significantly to the ac conductivity in addition to polaron hopping in the studied temperatureand frequency range.

4. Conclusion

The ac conductivity behaviour of Se_{85} Te_{15} and Se_{85} Te_{15} Ge_2 has been explained well by CBH model. But higher doping of Gemanium leads to the formation of simple pairs in the band

gap which contribute significantly to the ac conductivity in addition to the polaron hopping as suggested by CBH model.

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